

Quantum thermodynamics and metrology with mechanical degrees of freedom

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Abstract

Quantum metrology and quantum thermodynamics are two relatively young fields that, in the spirit of the second quantum revolution, apply quantum theory to address fundamental questions and to develop technological applications. While the research in these fields is predominantly conducted with discrete, finite-dimensional quantum systems in mind, this thesis makes the case for continuous variable mechanical degrees of freedom.

In the first of three projects, we present two realizations of the Otto cycle with a planar rotor as the working medium. As a mechanical system, the planar rotor has a well-defined classical analogue that allows the identification of genuine quantum effects by comparing classical and quantum machine. Here, we mainly focus on the parameter regimes, where the machine admits a certain operation mode, i.e. an engine, a refrigerator, or a heater. In the first realization, we find a systematic disadvantage of the quantum machine. The opposite is true for the second realization: It can be shown that the classical machine can, in general, not be run in a useful operation mode. The quantum machine, on the other hand, admits an engine operation mode for sufficiently cold temperatures of the cold bath.

The second project is devoted to the dynamics of a quantum system subjected to a thermal gas. In contrast to repeated interaction models, we consider any gas particle as a motional degree of freedom. We Employ quantum mechanical scattering theory to derive a low-density limit master equation, including gases with internal structure. Then, the thermodynamic consistency of the master equation is shown. A comparison with repeated interaction models makes evident that the inclusion of motional degrees of freedom of the gas plays a curial role for the consistency. Finally, we consider a nonequilibrium scenario, where the internal and motional degrees of freedom of the gas are thermal with respect to different temperatures. We show that, under the influence of this gas, the ergotropy of the system can increase.

In the last project, we apply quantum metrology on the measurement of magnetic moments in an electron microscope. We consider two types of sample, one that is static and does not change under the influence of the electron and another one that is described by a quantum system and experiences quantum backaction. For both samples, we derive the scattering operator from first principles. Then, two metrological tasks are considered: First, the sensing of the strength of the magnetic moment. We derive the Fisher information in several bases and find that momentum measurements are already optimal in the case without backaction. With backaction included, a measurement of angular momentum is optimal. The second task, we consider, is the optimal discrimination of the scattered and unscattered motional electron state. We derive the trace distance and find the experimentally achievable basis with the highest classical trace distance, which is still significantly worse than the theoretical optimum.

Zusammenfassung

Quantenmetrologie und Quantenthermodynamik sind zwei relativ junge Felder, die sich im Geiste der zweiten Quantenrevolution fundamentalen Fragen und technologischen Anwendungen widmen. Während die Forschung in diesen beiden Feldern vorwiegend auf diskreten, endlichdimensionalen Quantensystemen beruht, betrachte ich in dieser Arbeit mechanische Freiheitsgrade, die durch unendlichdimensionale Systeme beschrieben werden.

Im ersten von drei Projekten präsentiere ich zwei Realisierungen des Otto-Kreisprozesses, bei denen ein planarer Rotor als Arbeitsmedium dient. Als mechanisches System besitzt der Rotor ein wohldefiniertes klassisches Analogon, welches man verwenden kann, um reine Quanteneffekte durch den Vergleich von klassischem und quantenmechanischen Arbeitsmedium zu ermitteln. Das Hauptaugenmerk liegt in diesem Projekt auf dem Parameterregime, in dem der Kreisprozess einen bestimmten Betriebsmodus (Motor, Kühltank, oder “Heater”) annimmt. In der ersten Realisierung des Otto-Kreisprozesses zeige ich einen systematischen Nachteil der Maschine mit quantenmechanischem Arbeitsmedium. Das Gegenteil ist der Fall für die zweite Realisierung: Man kann zeigen, dass die klassische Maschine unter keinen Umständen einen nützlichen Betriebsmodus (Motor, Kühltank) erlaubt. Wird jedoch das quantisierte Arbeitsmedium verwendet, ist ein Betrieb als Motor für hinreichend kalte Temperaturen am kalten Bad möglich.

Das zweite Projekt dieser Arbeit widmet sich der Dynamik von Quantensystemen, die einem thermischen Gas ausgesetzt sind. Im Gegensatz zu sogenannten *repeated interaction models*, betrachte ich jedes Gasteilchen als einen dreidimensionalen Bewegungsfreiheitsgrad. Ich verwende quantenmechanische Streutheorie, um eine Mastergleichung für den Grenzfall stark verdünnter Gase herzuleiten, wobei das Gas selbst eine innere Struktur besitzen darf. Im Anschluss zeige ich, dass diese Mastergleichung mit den Hauptsätzen der Thermodynamik kompatibel ist. Der Vergleich mit den *repeated interaction models* offenbart, dass das Miteinbeziehen von Bewegungsfreiheitsgraden eine wichtige Voraussetzung für diese Kompatibilität ist. Abschließend betrachte ich ein Nichtgleichgewichtsszenario, in dem die Bewegungsfreiheitsgrade und die internen Freiheitsgrade der Gasteilchen zwar thermisch sind, aber in Bezug auf verschiedene Temperaturen. Ich zeige dann, dass Quantensysteme unter dem Einfluss dieses Gases ihre Ergotropie erhöhen können.

Im letzten Projekt wende ich Quantenmetrologie auf die Messung von magnetischen Momenten in der Elektronenmikroskopie an. Dazu unterscheide ich zwei Arten von Proben: Eine, die statisch ist und durch den Elektronenstrahl nicht beeinflusst wird, und eine, die selbst ein Quantensystem ist und zusammen mit dem Elektronenstrahl der Zeitentwicklung unterliegt. Im zweiten Fall spricht man auch von Quantenrückkopplung. Für beide Proben berechne ich den Streuoperator aus grundlegenden Prinzipien. Darauf-

hin werden zwei Szenarien untersucht: Im ersten Szenario ist das Ziel, das magnetische Moment möglichst präzise aus Messdaten des Elektronenmikroskops zu schätzen. Ich berechne die Fisherinformation für verschiedene Messbasen und zeige, dass eine Messung im Impulsraum im rückkopplungsfreien Fall bereits optimal ist. In dem zweiten Szenario untersuche ich die optimale Strategie zur Unterscheidung zwischen gestreuten und ungestreuten Zustand des Elektrons. Ich berechne die “trace distance” und finde die beste Messung, die experimentell durchführbar ist. Diese weicht jedoch signifikant vom theoretischen Optimum ab.

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Introduction

The discovery of quantum mechanics over 100 years ago brought great advancements in a wide range of fields. Especially, the insight into the quantized energy spectrum of many-body systems influenced the understanding of chemistry and solid state physics, eventually leading to pioneering new technologies such as semi-conductor materials. These advancements, mainly based on the understanding of quantization, material waves and quantum statistics, are known as the *first quantum revolution*. Today, we are in the middle of the *second quantum revolution*, where quantum phenomena like superposition and entanglement are used to create new technologies [1]. The fields of *quantum metrology* [2] and *quantum thermodynamics* [3] are, among others like quantum information theory, an integral part of this prospective new era. The research presented in this thesis focuses on these two areas that we now outline.

Quantum metrology, a successful new branch of quantum applications, is concerned with quantum enhancements of high precision sensing. It proved useful for the detection of gravitational waves [4], the sensing of magnetic fields with nitrogen-vacancy centers [5], and atom interferometry [6]. Another remarkable result of quantum metrology is that the distance of two incoherent point sources radiating light, understood as a quantum parameter estimation problem, can be estimated beyond the Rayleigh diffraction limit [7].

A perfect example for the distinction between the first and the second quantum revolution in the context of metrology can be drawn from electron microscopy [8]: Based on the wave-particle duality, electron microscopes were build in the course of the first quantum revolution. However, a consistent improvement of the beam focus, spatiotemporal manipulation, and detection techniques are now enabling the coherent manipulation of quantum systems [9], bringing forth novel technological applications today. This development formed the rising field of *free-electron quantum optics* that considers free electrons as a quantum resource [10]. Yet it is only recently that advanced methods from quantum metrology and phase estimation theory are being applied to electron microscopy [11,12], so far only for the sensing of a static sample's spatial profile.

Quantum thermodynamics (QTD), on the other hand, strives to apply thermodynamic concepts on quantum mechanical systems: Is concerned with concepts like thermal equilibration, the exchange of work and heat, and the performance of thermal machines when the working medium is chosen to be a quantum system. Since heat is always exchanged with a thermal environment, QTD is strongly linked with the theory of open quantum systems, where the effect of the environment on the thermodynamic system is usually modeled by so-called *master equations* [13].

Two research directions are especially promising. One of them is concerned with the fundamental question of how thermodynamics behavior can emerge in microscopic

quantum systems. Works in this field often consider a type of environmental model and proof the consistency of the laws of thermodynamics or, vice versa, show how the laws of thermodynamics have to be understood to be applicable [14–18]. The examined settings range from specified environments in form of master equations to more abstract scenarios with so-called *thermal operations* [19]. The other research direction is focused on potential technological application for the future: For example, how quantum resources can be used to optimize or improve thermodynamic processes and thermal machines. The Carnot cycle [20–24] and the Otto cycle [25–31] are simple machine models that are often used to single out quantum effects. To this end, the quantum machine is compared with a classical analogue of itself, which is usually understood as a version that operates without quantum coherence [32].

Since QTD is a rather young field, meaningful technological applications of QTD remain to be found. However, recent proof-of-principle experiments are laying the foundations for promising applications in the future: For heat engines there are, among others, experiments based on trapped ions [33–35], nitrogen-vacancy centers [36], nano mechanical systems [37], spins with nuclear magnetic resonance techniques [38–40], optomechanical systems [41, 42], quantum gases [43] and Josephson junctions [44].

A common theme in both outlined research fields is that the majority of models and findings concerns discrete, finite-dimensional quantum systems. While the simpler mathematical structure makes them a suitable choice for toy models, they often lack the complexity to represent physically realistic scenarios. In this thesis, we present three projects, one on quantum metrology and two on quantum thermodynamics. In each project, we consider mechanical degrees of freedom that are described by continuous-variable infinite-dimensional quantum systems.

In the project on *quantum metrology* (Chapter 4), we explore the possibilities of sensing magnetic moments in an electron microscope, building on the work of the Haslinger group at TU Wien [45]. We employ scattering theory in order to describe how the motional state of the electron changes due to the presence of the magnetic moment. Here, we distinguish two types of samples: a static one that is sufficiently large so as not to be influenced by the electron, and a magnetic moment sourced by a quantum mechanical spin. In the latter case, there is a quantum backaction on the spin that changes the analysis qualitatively. Given the scattering operator in either case, we derive the outcome statistics for measurements in position, momentum, and angular momentum and calculate the *Fisher information* from these, respectively. This not only characterizes the precision with which the magnetic moment can be sensed, but also yields, by comparison with the *quantum Fisher information*, a criterion whether a chosen measurement scheme is optimal. We will show that, without backaction, optimal precision can be achieved by measurements in momentum space, which in practice corresponds to measuring the far-field diffraction pattern of the electrons. With backaction included, this is not true anymore and higher precision would be achieved by discerning the orbital angular momentum components of the diffracted electron beam. However, it is not clear how such a measurement could be realized.

Besides sensing, we also explore quantum state discrimination of the scattered and unscattered state. Here, the strength of the magnetic moment is assumed known, but

it is uncertain whether a sample is present on the beam axis or not. The optimal success probability of deciding if there was a sample in a single measurement shot of the experiment is characterized by the (quantum) *trace distance*. As in the sensing case, we calculate the (classical) trace distance in the mentioned measurement bases and compare them with the optimal quantum value. In contrast to the sensing case, the quantum optimum can never be reached with available momentum-space measurements or with angular momentum measurements.

These *metrological* studies for electron microscopes have been conducted in close collaboration with the experimental group of Philipp Haslinger, where cutting-edge experiments in free-electron quantum optics are performed [46–48]. We are confident that our findings will support the design of experiments in the near future and, by that, have a direct impact on the field.

The first project on *quantum thermodynamics* theoretically investigates two realizations of the Otto cycle with quantum planar rotors as the working media. A working medium based on a mechanical degree of freedom, such as the planar rotor, has the advantage that there is an evident classical analogue. This allows for a fair comparison between the classical and the quantized machine and thus to identify genuine quantum effects. In the first realization of the Otto cycle, the rotor is subjected to an angle-dependent potential that is periodically modulated to mimic work exchange with a moving piston, and to an alternating hot and cold bath to allow for heat flow in and out of the rotor system. Physically, this setting could represent, e.g., an electric dipole rotating in an external electric field. Our investigations show that the quantum version of this rotor machine is systematically worse in terms of work output and operation regimes than the classical machine. The opposite is true for the second realization, where we consider a planar rotor with an interaction term that is linear in angular momentum, e.g. a charged rotor that rotates with varying angular frequency in a magnetic field. Classically, this realization of an Otto cycle does not work as an engine, for any choice of parameters. With a quantum rotor, on the other hand, we show that there is a parameter regime in which the system operates as an engine and outputs work.

Summarizing, this project demonstrates quantum disadvantage as well as advantage in terms of operation regimes for planar rotor-based thermal machines. It provides a working example for more general studies of quantum advantage. The project also makes the case for more complicated mechanical systems as working media, because these admit a fair comparison between classical and quantum machine, in contrast to finite-dimensional systems with no direct classical analogue. Since state-of-the-art experiments are able to cool all motional degrees of freedom of levitated particles close to the ground state [49] or even to prepare a planar rotor in a quantum state [50], proofs of principle for our proposed machine models should be experimentally feasible.

The second *quantum thermodynamics* project deals with open system dynamics and the thermodynamically consistent description of thermal environments. A common model of environments is based on the weak coupling to a large system [14], e.g. a continuum of harmonic oscillator modes. The most widely used physical example is the thermal radiation field in free space or in microwave circuits, which exchanges excitations with atoms or resonators. Besides this radiative environment, many systems

comprised of mechanical degrees of freedom are exposed to a surrounding gas and experience random collisions with the gas particles. In the framework of *repeated interaction models*, such collisions are described by a short joint evolution of the system of interest and an ancilla degree of freedom, after which the ancilla is traced out [51]. This framework completely neglects the motion of the particles that determines whether or not a collision occurs and replaces it by an external time dependence. This can result in thermodynamically inconsistent behavior such as thermalization to the wrong state [52], if the interactions are not tailored for the specific system. Jacob et al. [53] demonstrated that the problem can be alleviated by incorporating a one-dimensional motional degree of freedom. However, the authors have to use the so-called *effusion* distribution [54] instead of the Maxwell-Boltzmann distribution. In our work, we consider a three-dimensional gas and derive a master equation in the low-density limit [55]. We present a detailed derivation starting from the many-body description of the gas, including internal degrees of freedom of the gas particles. We further show that the master equation is consistent with the laws of thermodynamics. Finally, we go beyond the equilibrium case and consider a structured non-thermal bath comprised of motional and internal degrees of freedom that are thermal with respect to different temperatures. For properly chosen parameters, this bath constitutes an uncontrolled work reservoir.

In summary, this project is about a thermodynamically consistent collisional master equation based on scattering theory. The detailed derivation presented in this thesis is intended to lower the entrance barrier for those without prior knowledge of scattering theory. The master equation itself provides a realistic approach for collisional open system dynamics, which is an alternative to the operational, and often rather artificial, repeated interaction models. It also expands upon previous one-dimensional collision models and translates them into a more realistic, three dimensional scattering framework. In addition, our case study with a two temperature non-equilibrium reservoir highlights the possibility to perform work without an external control or timing.

The thesis is structured as follows: Chapter 1 reviews the basic concepts in quantum mechanics, scattering theory, open systems, thermodynamics, and metrology are presented. Chapter 2 is about the rotor-based Otto cycles. The collisional master equation is discussed in 3. In Chapter 4, we show the metrology for the sensing of magnetic moments with electron microscopes. Finally, we conclude in Chapter 5.

1. Basic concepts

In this chapter, we recall the important mathematical and physical concepts that will be needed throughout this thesis and fix the notation. The mathematical framework of quantum mechanics is discussed in Section 1.1. This discussion is intended as a concise review of elementary quantum mechanics. Experts in the field may find it redundant and can skip ahead without loss of continuity.

The second section (Section 1.2) focuses on the description of linear motion and scattering theory. We emphasize, in particular, the inelastic scattering scenario and derive the scattering operator, scattering amplitudes, and cross sections. The contents of this section will be needed in Chapter 3, where we describe collisional dynamics of thermal gases, and Chapter 4, where we infer information from a scattered electron.

The theory of open quantum systems is presented in Section 1.3. In order to consistently describe thermal or more general environments, we discuss Markov chains and local detailed balance classically and quantum channels, quantum semi-groups and master equations quantum mechanically. These basics will mainly be needed in Chapter 3.

Next, we present a short introduction into thermodynamics in Section 1.4. After briefly reviewing the main concepts of classical thermodynamics, we discuss the microscopic picture. Finally, we outline the conceptual problem in the definition of quantum work and present those definitions that we will need in Chapter 2 to describe the performance of an engine.

The final section of the basics chapter, Section 1.5, is about metrology. There, we discuss classical and quantum concepts for parameter estimation and state discrimination. This will be exclusively needed in Chapter 4, where we apply metrology to electron microscopes.

1.1. Mathematical framework of quantum mechanics

Over the course of 100 years after the seminal works of Heisenberg, Born and Jordan [56–58], a powerful mathematical framework for the use of quantum mechanics has been developed. We proceed with a review of the relevant concepts.

In most physical theories, we can identify three categories of objects: states, transformations and measurements. In quantum mechanics, all three can be formulated in terms of operators on a complex and separable Hilbert space $\mathcal{H}$. The details of this formulation will be elaborated upon in the following subsections, where we stay close to [59] for the most time.

1. Basic concepts

1.1.1. Hilbert spaces

Consider the complex, separable and potentially infinite dimensional Hilbert space $\mathcal{H}$, where we denote elements of this space as "ket" vectors $|\psi\rangle \in \mathcal{H}$. Since the space is separable, we can always find a countable orthonormal basis (ONB) $(|n\rangle)_n$ in which we can expand vectors

$$|\psi\rangle = \sum_n \psi_n |n\rangle, \quad (1.1.1)$$

where the expansion coefficients $\psi_n = \langle n|\psi\rangle$ are complex numbers. The space is equipped with a scalar product that maps two vectors onto the underlying field, i.e. the complex numbers, $\langle\phi|\psi\rangle \in \mathbb{C}$. The scalar product can explicitly be written in terms of the expansion coefficients ϕ_n of $|\phi\rangle$ and ψ_n of $|\psi\rangle$

$$\langle\phi|\psi\rangle = \sum_n \phi_n^* \psi_n, \quad (1.1.2)$$

where $()^*$ denotes the complex conjugate. Alternatively, we can read (1.1.2) as the linear functional

$$\langle\phi| = \sum_n \phi_n^* \langle n| \quad (1.1.3)$$

applied to the vector $|\psi\rangle$. We say $\langle\phi| \in \mathcal{H}'$ is a element of the dual vector space and call it a "bra" vector. By means of the scalar product, we can define a norm in $\mathcal{H}$ by taking the square root of a vector's overlap with itself

$$\| |\psi\rangle \|_2 = \sqrt{\langle\psi|\psi\rangle} = \sqrt{\sum_n |\psi_n|^2}. \quad (1.1.4)$$

Linear maps from a subset $D(\mathcal{H})$ in $\mathcal{H}$ are called operators and are denoted with a hat, $\hat{A} : D(\mathcal{H}) \rightarrow \mathcal{H}$. We can expand operators in any ONB by

$$\hat{A} = \sum_{n,k} A_{nk} |n\rangle \langle k|, \quad (1.1.5)$$

where the matrix elements are $A_{nk} = \langle n|\hat{A}|k\rangle$. The *identity* operator plays an important role and can be expressed by a *resolution of identity* in any ONB,

$$\mathbb{1} = \sum_n |n\rangle \langle n|. \quad (1.1.6)$$

The significance of this expression comes from the fact that we can "insert an identity" in order to change basis: Assume we want to expand the state $|\psi\rangle$ in the basis $(|b_k\rangle)_k$,

1. Basic concepts

then

$$\begin{aligned} |\psi\rangle &= \mathbb{1} |\psi\rangle = \sum_k |b_k\rangle \langle b_k | \psi \rangle = \sum_k |b_k\rangle \sum_n \psi_n \langle b_k | n \rangle \\ &= \sum_k \left(\sum_n \langle b_k | n \rangle \psi_n \right) |b_k\rangle = \sum_k \tilde{\psi}_k |b_k\rangle, \end{aligned} \quad (1.1.7)$$

where we multiplied with identity, resolved the identity in the new basis, expanded $|\psi\rangle$ in the old basis and finally identified the expansion coefficients $\tilde{\psi}_k$ in the new basis.

An important map on the space of operators is the so called Hermitean conjugate,

$$\hat{A} = \sum_{n,k} A_{nk} |n\rangle \langle k| \mapsto \hat{A}^\dagger = \sum_{n,k} A_{kn}^* |n\rangle \langle k|, \quad (1.1.8)$$

which can be seen as a transposition and a complex conjugate of the matrix elements. This allows us to read quadratic forms in two different ways,

$$\langle \phi | \hat{A} \psi \rangle = \langle \hat{A}^\dagger \phi | \psi \rangle, \quad \forall |\psi\rangle, |\phi\rangle, \quad (1.1.9)$$

where we either apply $\hat{A}$ to $|\psi\rangle$ and then multiply the result with $|\phi\rangle$, or we can apply $\hat{A}^\dagger$ to $|\phi\rangle$ and then multiply with $|\psi\rangle$. This freedom is especially helpful if we only know the effect of $\hat{A}$ or $\hat{A}^\dagger$ on one of the states.

Based on the behavior under the Hermitean conjugate we can define two classes of operators: An operator that satisfies $\hat{A}^\dagger = \hat{A}$ is called *self-adjoint* or *Hermitean*. If the operator becomes its own inverse, $\hat{A}^\dagger \hat{A} = \hat{A} \hat{A}^\dagger = \mathbb{1}$, it is called *unitary*. Both cases have in common that the operator and its inverse commute, $[\hat{A}, \hat{A}^\dagger] = 0$.

1.1.2. Spectral decomposition

In the last subsection we have seen different characterizations of operators based on their behavior under the adjoint operation. For the class of self-adjoint operators¹, i.e. operators such that $\hat{A}^\dagger = \hat{A}$, we will now discuss the important *spectral theorem*. The spectral theorem is a direct generalization of the eigenvalue decomposition in finite-dimensional spaces, which we assume to be known. The rest of this subsection follows along the line of Ref. [60], where the reader can find a concise explanation of the most general case. Given that $\hat{A}$ is self-adjoint, it can be expanded in terms of the operator-valued integral,

$$\hat{A} = \int_{-\infty}^{\infty} a \, d\hat{E}_a, \quad (1.1.10)$$

¹Note that $\hat{A}^\dagger = \hat{A}$ is meant for the linear maps. In the infinite-dimensional case, this additionally requires that the two operators live on the same domain. For the well behaved operators appearing in physics, however, we will assume this to be always true.

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where $a \in \mathbb{R}$ is a real number indexing the so-called *spectral family* $(\hat{E}_a)_a$. The spectral family contains mutually commuting orthogonal projections that are monotonously increasing, i.e. $\hat{E}'_a > \hat{E}_a$ for $a' > a$, continuous from the right, i.e. $\hat{E}_{a'} \rightarrow \hat{E}_a$ for $a' \rightarrow a$ from above, and fulfill the limits

$$\lim_{a \rightarrow -\infty} \hat{E}_a = 0 \quad (1.1.11)$$

$$\lim_{a \rightarrow +\infty} \hat{E}_a = 1. \quad (1.1.12)$$

The right hand side of (1.1.10) has to be understood in the sense that, for any two vectors $|\phi\rangle, |\psi\rangle \in \mathcal{H}$, it holds

$$\langle \phi | \hat{A} | \psi \rangle = \int_{-\infty}^{\infty} a \, d \langle \phi | \hat{E}_a | \psi \rangle, \quad (1.1.13)$$

where $d \langle \phi | \hat{E}_a | \psi \rangle$ is a $\mathbb{C}$ -valued measure and the integral is over the real numbers. We will now present with two examples how the so-called *spectrum* of $\hat{A}$ is encoded in the spectral family: First, let's consider a diagonal operator on a two-dimensional Hilbert space,

$$\hat{A} = 2 |2\rangle \langle 2| + 1 |1\rangle \langle 1|. \quad (1.1.14)$$

The spectral family in this case becomes

$$\hat{E}_a = \begin{cases} 0 & \text{for } 1 > a \\ |1\rangle \langle 1| & \text{for } 2 > a \geq 1 \\ |1\rangle \langle 1| + |2\rangle \langle 2| & \text{for } a \geq 2 \end{cases}. \quad (1.1.15)$$

We observe that at points where a is an eigenvalue of $\hat{A}$ the spectral family is discontinuous and changes by a projector on an eigen space, e.g. $\hat{E}_{1+\epsilon} - \hat{E}_{1-\epsilon} = |1\rangle \langle 1|$. This holds in general: A point a_{discrete} , where the spectral family is discontinuous, belongs to the so-called *point spectrum* of the operator. The space, on which $\hat{E}_{a+\epsilon} - \hat{E}_{a-\epsilon}$ projects, is spanned by proper eigen vectors $|a\rangle \in \mathcal{H}$ in the sense that $\hat{A} |a\rangle = a |a\rangle$. A complementary example can be found by assuming a multiplication operator on a space of square integrable functions $\hat{x} : \psi(x) \mapsto x\psi(x)$: Then the spectral family acts like

$$(\hat{E}_y \psi)(x) = \Theta(y - x) \psi(x), \quad (1.1.16)$$

where the original function is set to zero for every point where $x \geq y$, denoted by the Heaviside function Θ . This spectral family is continuously changing at every point. In general, we say a point, at which $\hat{E}_x$ changes continuously, belongs to the continuous spectrum of the operator. In this specific example, the whole spectrum of $\hat{x}$ is continuous and coincides with the real numbers. Note that, in contrast to the point spectrum, the continuous spectrum does not have corresponding proper eigenvectors: Imagine we had

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an eigen function $\psi(x)$ of $\hat{x}$ with the eigenvalue x_0 , then this would mean

$$x\psi(x) = x_0\psi(x), \quad (1.1.17)$$

which can not be fulfilled for any square integrable function. The only option would be to concentrate the function around x_0 to a single point, leading to the one-dimensional delta function $\psi(x) = \delta^{(1)}(x - x_0)$, which indeed fulfills

$$x\delta^{(1)}(x - x_0) = x_0\delta^{(1)}(x - x_0). \quad (1.1.18)$$

On the contrary, the delta function is not a proper function and in general not square integrable. In physics, however, there is a powerful formal tool that allows us to treat the continuous spectrum and the point spectrum on the same footing, improper vectors. The main idea is to introduce non-normalizable eigenvectors of $\hat{x}$ (in this case) $(|x\rangle)_x$ that fulfill

$$\hat{x}|x\rangle = x|x\rangle \quad (1.1.19)$$

and have a scalar product of

$$\langle y|x\rangle = \delta^{(1)}(x - y). \quad (1.1.20)$$

The spectral family $\hat{E}_y$ in this formalism can be written as

$$\hat{E}_y = \int_{-\infty}^y dx |x\rangle \langle x| \quad (1.1.21)$$

and conclusively

$$\mathbb{1} = \hat{E}_{\infty} = \int_{-\infty}^{+\infty} dx |x\rangle \langle x|. \quad (1.1.22)$$

Analogous to any proper basis (1.1.7), we can expand states and operators in the improper basis by using the resolution of identity (1.1.22). Note that the strength of the improper eigenvectors lies in the fact that in equations like (1.1.22) the left hand side is a well-defined mathematical object, while the right hand side can be intuitively understood in the same fashion as for a proper eigenbasis. Finally, we want to define functions of operators. Consider a function $f : \mathbb{C} \mapsto \mathbb{C}$ that admits an expansion as a power series

$$f(z) = \sum_{k=0}^{\infty} f_k z^k. \quad (1.1.23)$$

Then, the function of the operator can simply be defined as

$$f(\hat{A}) = \sum_{k=0}^{\infty} f_k \hat{A}^k. \quad (1.1.24)$$

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If the operator admits a spectral decomposition, this simplifies to just applying the function to the eigen values,

$$f(\hat{A}) = \int_{-\infty}^{\infty} f(a) d\hat{E}_a. \quad (1.1.25)$$

In case the function can not be expanded in a power series, we will use expression (1.1.25) as the definition instead.

1.1.3. States and measurements

In quantum theory, we have to distinguish two types of states, *pure states* and *mixed states*: A *pure state* can be expressed as a vector $\psi \in \mathcal{H}$ that is normalized to one, i.e. $\|\psi\| = 1$. Vectors that only differ by a global phase $|\psi'\rangle = e^{i\varphi} |\psi\rangle$ correspond to the same physical state. The formalism of pure states is very useful if we have full classical information and control of our quantum system. In practice, however, we often lack this precise knowledge and ability: For example, the initial state might not be precisely known or there might be uncontrolled interactions with the environment. In principle, this lack of knowledge could be encoded in a probability distribution over the actual quantum state, but this would force us to tediously update the distribution in every step of the calculation, too. Fortunately, we can use the so-called *mixed states* to treat classical and quantum uncertainty on the same footing:

An operator $\hat{\rho}$ that is positive semi definite, i.e. $\langle \phi | \hat{\rho} | \phi \rangle \geq 0, \forall |\phi\rangle \in \mathcal{H}$, and has unit trace ² $\text{tr}\{\hat{\rho}\} = 1$ is called a *density matrix*, *density operator*, or just *state*. It encodes all the information that we have about a particular physical system. A single pure state in this formalism becomes the rank-one operator $\hat{\rho} = |\psi\rangle \langle \psi|$ and in the example from above, where we have a probability distribution $(p_n)_n$ over pure states $|\psi_n\rangle$, the corresponding density operator is the convex combination

$$\hat{\rho} = \sum p_n |\psi_n\rangle \langle \psi_n|. \quad (1.1.26)$$

Given the definition of density operators, we can now introduce measurements in form of *positive operator-valued measure* (POVM). Consider a sample space X , which is the set of all possible measurement results, e.g. the real line $X = \mathbb{R}$ for the position of a particle or $X = \{1, \dots, n\}$ for some internal levels. An *event* is a subset of the sample space $A \subseteq X$ and we denote the set of all events to be $\mathcal{A}$ ³. A POVM is a map $A \mapsto \hat{E}(A)$

²In the infinite dimensional case, the operator needs to fulfill $\text{tr}\{|\hat{\rho}|\} < \infty$, too

³Strictly speaking, $\mathcal{A}$ has to be a so-called sigma algebra. This basically means that $\mathcal{A}$ is closed under logical operations on events like "and" (union), "not" (complement), etc.

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from $\mathcal{A}$ into the set of positive operators, which are bounded from above by $\mathbb{1}$ such that

$$\hat{E}(\{\}) = 0 \quad (1.1.27)$$

$$\hat{E}(X) = \mathbb{1} \quad (1.1.28)$$

$$\hat{E}\left(\bigcup_j A_j\right) = \sum_j \hat{E}(A_j), \quad (1.1.29)$$

where the family of sets in the last line $(A_j)_j$ has to be disjoint. The operators $\hat{E}(A)$ are called *effects*. These properties ensure that for any state $\hat{\rho}$

$$\mathbb{P}_{\hat{\rho}}(A) = \text{tr} \left\{ \hat{\rho} \hat{E}(A) \right\} \quad (1.1.30)$$

is a probability measure. Conclusively, if we perform a measurement described by the POVM $\hat{E}$ on a quantum system described by state $\hat{\rho}$, the probability that the event A occurs is $\text{tr} \left\{ \hat{\rho} \hat{E}(A) \right\}$. If we choose a disjoint sequence $(A_n)_n \subseteq X$ that fulfill $\bigcup A_n = X$, we can define $\hat{E}_n = \hat{E}(A_n)$. In finite-dimensional scenarios, the collection $(\hat{E}_n)_n$ is often referred to as POVM. An infinite dimensional example is given in form of a particle position, where $A \subseteq \mathbb{R}$ is a region where the particle is potentially located and

$$\hat{E}(A) = \int_A dx |x\rangle \langle x| \quad (1.1.31)$$

is the associated operator⁴. Given a state $|\psi\rangle$, the probability to find the particle in the region A is

$$\mathbb{P}(A) = \text{tr} \left\{ |\psi\rangle \langle \psi| \hat{E}(A) \right\} = \langle \psi | \hat{E}(A) | \psi \rangle = \int_A dx \langle x | \psi \rangle \langle \psi | x \rangle = \int_A dx |\psi(x)|^2. \quad (1.1.32)$$

With the preceding concepts we can describe the outcome statistics of any quantum measurement. In general, the system is not discarded after the measurement and impact on the system state has to be discussed: We define so-called *measurement operators* $\hat{M}(A)$ that can be related to the effects,

$$\hat{E}(A) = \hat{M}^\dagger(A) \hat{M}(A). \quad (1.1.33)$$

Then, the post measurement state can expressed by the transformation,

$$\hat{\rho} \mapsto \frac{\hat{M}(A) \hat{\rho} \hat{M}^\dagger(A)}{\text{tr} \left\{ \hat{\rho} \hat{E}(A) \right\}}, \quad (1.1.34)$$

⁴Note the connection to the spectral family (1.1.10) $\hat{E}_y = \hat{E}((-\infty, y])$: The spectral family relates to a POVM in the same fashion as the cumulative distribution function relates to a probability density.

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when event A is measured. Note that the denominator, which normalizes the transformed state by the probability for event A to occur, makes the evolution non-linear.

Given any set of measurement operators ($\hat{M}(A)$), we can apply an outcome-dependent feedback unitary $\hat{U}(A)$ to obtain another set of measurement operators $\hat{M}'(A) = \hat{U}(A)\hat{M}(A)$ that correspond to the same set of effects

$$\hat{M}'^\dagger(A)\hat{M}'(A) = \hat{M}^\dagger(A) \underbrace{\hat{U}^\dagger(A)\hat{U}(A)}_{=\mathbb{1}} \hat{M}(A) = \hat{E}(A). \quad (1.1.35)$$

Such a feedback unitary can be, for example, used to generate measurement feedback processes, see [61]. However, the simplest choice of measurement operators is given by the *Lüders rule*, where we set $\hat{M}(A) = \sqrt{\hat{E}(A)}$.

1.1.4. Time evolution and Schrödinger equation

Besides states and measurements, physical theories provide rules under which the states transform. In the case of quantum mechanics, the evolution of a *closed* system, i.e. a system isolated from any environment and described by a pure state, evolves according to the Schrödinger equation,

$$i\hbar \frac{d}{dt} |\psi(t)\rangle = \hat{H} |\psi(t)\rangle. \quad (1.1.36)$$

Here, $\hat{H}$ is a self-adjoint operator, the so-called *Hamiltonian* or *Hamilton operator*. This differential equation can be formally solved by the operator exponential

$$|\psi(t)\rangle = \underbrace{\exp\left(-\frac{i}{\hbar}\hat{H}(t-t_0)\right)}_{=\hat{U}(t,t_0)} |\psi(t_0)\rangle, \quad (1.1.37)$$

where we call $\hat{U}(t, t_0)$ the *time evolution operator* or *propagator*. From the fact that $\hat{H}$ is self-adjoint and from the properties of the exponential, we can deduce that the time evolution operator is unitary. Another implication of $\hat{H}$ being self-adjoint is that it admits a spectral decomposition

$$\hat{H} = \int E \, d\hat{P}_E = \sum_n E_n |E_n\rangle \langle E_n|, \quad (1.1.38)$$

where we assume, for simplicity, the spectrum to be discrete with energy levels E_n and energy eigenstates $|E_n\rangle$. The significance of the energy eigenstates lies in the fact that they obtain a phase proportional under time evolution. Thus, given the eigen decomposition of the Hamilton operator, the time evolution can be trivially solved in

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the energy eigenbasis

$$\hat{U}(t, t_0) = \sum_n \exp\left(-\frac{i}{\hbar} E_n(t - t_0)\right) |E_n\rangle \langle E_n|. \quad (1.1.39)$$

The situation gets more complicated if we consider a time dependent Hamiltonian $\hat{H}(t)$. This can occur, for example, if we have externally controlled parameters in our system, or if we describe a system in a co-moving frame. If we think about this time evolution as composed of many short time intervals of length Δt over which the Hamiltonian is constant, it is plausible that the resulting time evolution is again unitary as the product of many unitaries. We denote the time evolution again as $\hat{U}(t, t_0)$. By comparison with the Schrödinger equation for state vectors, we find,

$$i\hbar \frac{d}{dt} \hat{U}(t, t_0) |\psi(t_0)\rangle = i\hbar \frac{d}{dt} |\psi(t)\rangle = \hat{H}(t) |\psi(t)\rangle = \hat{H}(t) \hat{U}(t, t_0) |\psi(t_0)\rangle, \quad (1.1.40)$$

the operator-valued differential equation for the propagator,

$$i\hbar \frac{d}{dt} \hat{U}(t, t_0) = \hat{H} \hat{U}(t, t_0), \quad (1.1.41)$$

where we concluded the equality of the operators from the fact that their images have to coincide for any initial $|\psi(t_0)\rangle \in \mathcal{H}$. Integrating (1.1.41) and implementing the initial condition $\hat{U}(t_0, t_0) = \mathbb{1}$, yields an integral equation,

$$\hat{U}(t, t_0) = \mathbb{1} - \frac{i}{\hbar} \int_{t_0}^t dt' \hat{H}(t') \hat{U}(t', t_0), \quad (1.1.42)$$

that can be iterated and truncated in order to generate an infinite series of terms, the so-called *Dyson series*,

$$\hat{U}(t, t_0) = \sum_{n=0}^{\infty} \hat{U}^{(n)}(t, t_0). \quad (1.1.43)$$

The first few terms are given by,

$$\begin{aligned} \hat{U}^{(0)}(t, t_0) &= \mathbb{1} \\ \hat{U}^{(1)}(t, t_0) &= -\frac{i}{\hbar} \int_{t_0}^t dt' \hat{H}(t') \\ \hat{U}^{(2)}(t, t_0) &= \left(-\frac{i}{\hbar}\right)^2 \int_{t_0}^t dt' \hat{H}(t') \int_{t_0}^{t'} dt'' \hat{H}(t''). \end{aligned} \quad (1.1.44)$$

In the second order term, the order of the operators becomes important for the first time since $\hat{H}(t')$ and $\hat{H}(t'')$ might not commute unless $t' = t''$. We can simplify this by noting

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$$\begin{aligned}
\int_{t_0}^t dt' \int_{t_0}^{t'} dt'' \hat{H}(t') \hat{H}(t'') &= \int_{t_0}^t dt' \int_{t_0}^{t'} dt'' 1_{\{t' > t''\}} \hat{H}(t') \hat{H}(t'') \\
&= \int_{t_0}^t dt' \int_{t_0}^{t'} dt'' \frac{1}{2} \left(1_{\{t' > t''\}} \hat{H}(t') \hat{H}(t'') + 1_{\{t'' > t'\}} \hat{H}(t'') \hat{H}(t') \right) \\
&= \int_{t_0}^t dt' \int_{t_0}^{t'} dt'' \frac{1}{2} \mathcal{T} \left\{ \hat{H}(t') \hat{H}(t'') \right\} \\
&= \frac{1}{2} \mathcal{T} \left\{ \left(\int_{t_0}^t dt' \hat{H}(t') \right)^2 \right\}, \tag{1.1.45}
\end{aligned}$$

where we first used a characteristic function to absorb the integral limit into the integrand and second made the expression symmetric in t' and t'' by switching $t' \leftrightarrow t''$ in one half of the integral expression. In the third step the *time ordering operator*, which orders operator functions from left to right at decreasing time points, was introduced. Finally, with some abuse of notation, we pulled the time ordering out of the integral. This has to be read as a formal expression where, for the actual calculation, the double integral has to be written out again. The generalization of this is straightforward, leading to the expression of the n -th term in the Dyson series,

$$\hat{U}^{(n)}(t, t_0) = \frac{1}{n!} \left(-\frac{i}{\hbar} \right)^n \mathcal{T} \left\{ \left(\int_{t_0}^t dt' \hat{H}(t') \right)^n \right\}. \tag{1.1.46}$$

This can now be absorbed in the Dyson series to yield a formal exponential function

$$\hat{U}(t, t_0) = \mathcal{T} \left\{ \exp \left(-\frac{i}{\hbar} \int_{t_0}^t dt' \hat{H}(t') \right) \right\}. \tag{1.1.47}$$

Although this formal series can be calculated exactly for some cases, in most situations it will be used to calculate approximate solutions by truncating the series at some point.

We can even use the Dyson series, in order to derive approximate solutions, if the original Hamiltonian $\hat{H}$ is not time-dependent: An often encountered situation is where the Hamiltonian is the sum of a well-understood *free Hamiltonian* $\hat{H}_0$ and a more complicated *interaction Hamiltonian* $\hat{V}$,

$$\hat{H} = \hat{H}_0 + \hat{V}. \tag{1.1.48}$$

In such a case, it can be beneficial to go into a co-moving frame, called *Interaction picture* and denoted with a superscript (I). Here, states and operators are connected to the original *Schrödinger picture* by

$$\begin{aligned}
|\psi^{(I)}(t)\rangle &= e^{i\hat{H}_0 t/\hbar} |\psi(t)\rangle \\
\hat{V}^{(I)}(t) &= e^{i\hat{H}_0 t/\hbar} \hat{V} e^{-i\hat{H}_0 t/\hbar}. \tag{1.1.49}
\end{aligned}$$

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Multiplying the Schrödinger equation from the left with $e^{i\hat{H}_0 t/\hbar}$ yields on the left hand side,

$$i\hbar e^{i\hat{H}_0 t/\hbar} \frac{d}{dt} e^{-i\hat{H}_0 t/\hbar} |\psi^{(I)}(t)\rangle = \left(i\hbar \frac{d}{dt} + \hat{H}_0 \right) |\psi^{(I)}(t)\rangle. \quad (1.1.50)$$

On the right hand side, we find,

$$e^{i\hat{H}_0 t/\hbar} \left(\hat{H}_0 + \hat{V} \right) e^{-i\hat{H}_0 t/\hbar} |\psi^{(I)}(t)\rangle = \left(\hat{H}_0 + \hat{V}^{(I)}(t) \right) |\psi^{(I)}(t)\rangle. \quad (1.1.51)$$

On both sides of the equation, the $\hat{H}_0$ term cancels and we find the interaction picture Schrödinger equation,

$$i\hbar \frac{d}{dt} |\psi^{(I)}(t)\rangle = \hat{V}^{(I)}(t) |\psi^{(I)}(t)\rangle. \quad (1.1.52)$$

This equation can now be solved or approximated by the Dyson Series.

As a final remark in this subsection, we shortly rephrase the closed system dynamics in terms of density matrices: The Schrödinger equation can directly translated to the *Von Neumann equation*,

$$i\hbar \frac{d}{dt} \hat{\rho}(t) = [\hat{H}, \hat{\rho}(t)]. \quad (1.1.53)$$

Whether $\hat{H}$ is time-dependent or not, we can use the corresponding time evolution operator for pure states and obtain

$$\hat{\rho}(t) = \hat{U}(t, t_0) \hat{\rho}(t_0) \hat{U}^\dagger(t, t_0). \quad (1.1.54)$$

1.1.5. Composite systems

Physical systems typically consist of many degrees of freedom. Therefore, we will show how to mathematically combine subsystems into a bigger composite system by means of the *tensor product*. In case that some of the systems are not accessible experimentally, we introduce the *partial trace*, which averages over the unknown degrees of freedom. This eventually leads to the notion of open system dynamics.

Consider two quantum physical systems, one described by states from a Hilbert space $\mathcal{H}_1$ and the other with states from the Hilbert space $\mathcal{H}_2$. Consistently, both systems together are still a quantum system and should be described by a Hilbert space $\mathcal{H}$. The new Hilbert space is called the *tensor product* $\mathcal{H} = \mathcal{H}_1 \otimes \mathcal{H}_2$ of the former ones. A straightforward construction of the space can be given in terms of orthogonal bases $(|n\rangle_1)_{n \in \mathbb{N}}$ and $(|k\rangle_2)_{k \in \mathbb{N}}$ of the respective spaces. We define a new basis,

$$(|n\rangle_1 \otimes |k\rangle_2)_{n, k \in \mathbb{N}}, \quad (1.1.55)$$

which is orthonormal with respect to the scalar product,

$$(\langle n'|_1 \otimes \langle k'|_2) (|n\rangle_1 \otimes |k\rangle_2) = \langle n'|_1 \langle k'|_2 \rangle_1 \langle k|_2 = \delta_{n'n} \delta_{k'k}. \quad (1.1.56)$$

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By linear combination, we now construct any state $|\Psi\rangle \in \mathcal{H}_1 \otimes \mathcal{H}_2$ as

$$|\Psi\rangle = \sum_{n,k} \Psi_{nk} |n\rangle_1 \otimes |k\rangle_2. \quad (1.1.57)$$

Vice versa, if the first system is prepared in state $|\psi\rangle_1$ and system two in $|\phi\rangle_2$, the compound state $|\psi\rangle_1 \otimes |\phi\rangle_2$ can be brought into the form (1.1.57) by the linearity of the tensor product for both states,

$$\begin{aligned} |\psi\rangle_1 \otimes |\phi\rangle_2 &= \left(\sum_n \psi_n |n\rangle_1 \right) \otimes |\phi\rangle_2 = \sum_n \psi_n (|n\rangle_1 \otimes |\phi\rangle_2) \\ &= \sum_n \psi_n \left(|n\rangle_1 \otimes \left(\sum_k \phi_k |k\rangle_2 \right) \right) = \sum_{n,k} \psi_n \phi_k (|n\rangle_1 \otimes |k\rangle_2). \end{aligned} \quad (1.1.58)$$

Note that we will also use the notations

$$|\psi\rangle \otimes |\phi\rangle = |\psi\rangle |\phi\rangle = |\psi, \phi\rangle. \quad (1.1.59)$$

The tensor product can not only combine vectors, but also operators: Given two operators $\hat{A}_1$ and $\hat{A}_2$ on the respective Hilbert spaces $\mathcal{H}_1$ and $\mathcal{H}_2$, we can define an operator on the compound Hilbert space $\hat{A}_1 \otimes \hat{A}_2$ that acts like,

$$\hat{A}_1 \otimes \hat{A}_2 (|n\rangle_1 \otimes |k\rangle_2) = \left(\hat{A}_1 |n\rangle_1 \right) \otimes \left(\hat{A}_2 |k\rangle_2 \right). \quad (1.1.60)$$

This enables us to extend the formalism of mixed states to compound systems, where a crucial distinction can be made: Consider a state of product form,

$$\hat{\rho}_1 \otimes \hat{\rho}_2. \quad (1.1.61)$$

This just describes system one in state $\hat{\rho}_1$ and system two in state $\hat{\rho}_2$. We can also assume a common random source that prepares states like this. The overall state then becomes the convex combination,

$$\sum_n p_n \hat{\rho}_1^{(n)} \otimes \hat{\rho}_2^{(n)}, \quad (1.1.62)$$

which we call *separable*. Separable states can be fully understood classically. On the other hand, there are states ρ on $\mathcal{H}_1 \otimes \mathcal{H}_2$, that are not of the form (1.1.62). These states are called *entangled*. While the correlations in some entangled states can still be understood classically, there are known scenarios where local realism has to be given up [62].

We have now seen how quantum systems can be combined to bigger quantum systems by means of the tensor product. Typically, many of the degrees of freedom are relevant for the overall time evolution of the system but are not accessible experimentally. In

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this situation, we can apply the so-called *partial trace* to eliminate inaccessible degrees of freedom while preserving the correct statistics of the remaining ones. Mathematically, the partial trace is given by the formula

$$\mathrm{tr}_2 \left\{ \hat{X} \right\} = \sum_{n,k,l} \langle n, l | \hat{X} | k, l \rangle | n \rangle \langle k |. \quad (1.1.63)$$

The significance comes from the following identity: Consider a state $\hat{\rho}$ on $\mathcal{H}_1 \otimes \mathcal{H}_2$, where we want to measure an effect $\hat{E}$ regarding the degrees of freedom described by $\mathcal{H}_1$. Since, by assumptions, the degrees of freedom in $\mathcal{H}_2$ are inaccessible, we can only perform the trivial measurement with effect $\mathbb{1}$ on that space. Short calculation shows

$$\mathrm{tr} \left\{ (\hat{E} \otimes \mathbb{1}) \hat{\rho} \right\} = \mathrm{tr} \left\{ \hat{E} \mathrm{tr}_2 \{ \hat{\rho} \} \right\}, \quad (1.1.64)$$

where we see that, instead of the measurement $\hat{E} \otimes \mathbb{1}$ on $\mathcal{H}_1 \otimes \mathcal{H}_2$, we can perform the measurement $\hat{E}$ on $\mathcal{H}_1$ by using the *reduced state* $\mathrm{tr}_2 \{ \hat{\rho} \}$. Note that the ordinary trace $\mathrm{tr} \{ \}$ is always meant to be applied on the whole space. Hence in (1.1.64), the trace on the left hand side is applied to the full Hilbert space $\mathcal{H}_1 \otimes \mathcal{H}_2$, while the trace on the right hand side goes over the remaining space $\mathcal{H}_1$.

1.1.6. The two-level system as a toy model

The quantum system with the smallest Hilbert space dimension is the two-level system, also known as *qubit*. Due to the limited dimension, most physically interesting objects—such as self-adjoint operators, unitary operators, and states—are fully characterized and well understood. For this reason, the two-level system is a convenient choice for the illustration of new and complicated concepts. Here, we summarize the most important facts.

In the two-dimensional Hilbert space $\mathbb{C}^2$, all operators can be identified with 2×2 matrices. Strictly speaking, such a matrix just expresses the matrix elements of the operator in a certain basis, i.e. $\langle \phi | \hat{H} | \psi \rangle$. However, for convenience we think of a fixed basis and use $\hat{=}$ to identify operators with matrices.

The most general self-adjoint operator is parametrized by four real numbers a_0 , a_x , a_y , and a_z ,

$$\hat{H} \hat{=} \begin{pmatrix} a_0 + a_z & a_x - \mathrm{i}a_y \\ a_x + \mathrm{i}a_y & a_0 - a_z \end{pmatrix}. \quad (1.1.65)$$

It is customary to decompose this general operator in a basis consisting of the identity operator $\mathbb{1}$ and the three Pauli operators,

$$\hat{\sigma}_x \hat{=} \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}, \quad \hat{\sigma}_y \hat{=} \begin{pmatrix} 0 & -\mathrm{i} \\ \mathrm{i} & 0 \end{pmatrix}, \quad \hat{\sigma}_z \hat{=} \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}. \quad (1.1.66)$$

Combining the Pauli operators in a vector $\hat{\sigma} = (\hat{\sigma}_x, \hat{\sigma}_y, \hat{\sigma}_z)$ and setting the, in most cases

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irrelevant, shift $a_0 = 0$, a general self-adjoint operator becomes

$$\hat{H} = \mathbf{a} \cdot \hat{\boldsymbol{\sigma}}. \quad (1.1.67)$$

It can be verified by straightforward calculation that these operators are self-adjoint and fulfill the commutation relations,

$$[\hat{\sigma}_i, \hat{\sigma}_j] = 2i\epsilon_{ijk}\hat{\sigma}_k \quad (1.1.68)$$

and anti-commutation relations,

$$\{\hat{\sigma}_i, \hat{\sigma}_j\} = 2\delta_{jk}\mathbb{1}. \quad (1.1.69)$$

The general unitary is given by the exponential of the general Hamiltonian (1.1.65) and can be, under the assumption $|\mathbf{a}| = 1$, expressed as

$$\hat{U} = \exp(-i\varphi(\mathbf{a} \cdot \hat{\boldsymbol{\sigma}})) = \mathbb{1} \cos(\varphi) - i(\mathbf{a} \cdot \hat{\boldsymbol{\sigma}}) \sin(\varphi). \quad (1.1.70)$$

It can be checked that the expression,

$$\hat{\rho} = \frac{1}{2}(\mathbb{1} + \mathbf{r} \cdot \hat{\boldsymbol{\sigma}}), \quad (1.1.71)$$

is a quantum state if and only if $|\mathbf{r}| \leq 1$ and it is pure for $|\mathbf{r}| = 1$. The characterizing vector $\mathbf{r}$ is called *Bloch* vector.

1.2. Linear motion and scattering theory

Scattering theory is an important part of modern physics that allows to single out the net effect of short time interactions. Here, we present a short recap on the algebra and eigenstates of position and momentum operator in Subsection 1.2.1, before we discuss scattering theory: In Subsection 1.2.2, we introduce the scattering operator as a map from asymptotic incoming to asymptotic outgoing states. Next in Subsection 1.2.3, we discuss abstract properties like energy conservation of scattering events and the optical theorem. Then, scattering amplitudes and cross sections are introduced in Subsection 1.2.4 and the Born approximation is discussed in Subsection 1.2.5. Finally, we discuss how to regularize the delta function that appears in the matrix elements of the scattering operator due to energy conservation in Subsection 1.2.6. The presented concepts can be found in [63]. Note that our presentation is focused on inelastic scattering as we need it in Chapter 3

1.2.1. The canonical commutation relation

The canonical commutation relation (CCR) for the position and momentum operator, proposed in [57] in its modern form, is the key ingredient in order to describe translational

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motion and its implications will be used frequently throughout this thesis. A detailed discussion on the matter can be found in most quantum textbooks, such as [64]. In one dimension the CCR can be phrased as

$$[\hat{x}, \hat{p}] = i\hbar \mathbf{1}. \quad (1.2.1)$$

Equation (1.2.1) has already several implications: First of all, $\hat{x}$ and $\hat{p}$ can not be operators on a finite-dimensional Hilbert space⁵. Second, both operators can not be diagonal in the same basis. Thus, the uncertainty in both observables can not simultaneously be arbitrarily small. This is known as the *Heisenberg uncertainty principle* and can be quantified in terms of the standard deviations of $\hat{x}$ and $\hat{p}$ in any state $|\psi\rangle$,

$$\sqrt{\mathbb{V}[\hat{x}]_{|\psi\rangle}} \sqrt{\mathbb{V}[\hat{p}]_{|\psi\rangle}} \geq \frac{\hbar}{2} \quad (1.2.2)$$

We can further derive the commutator between $\hat{p}$ and any function of $\hat{x}$ and vice versa,

$$[f(\hat{x}), \hat{p}] = i\hbar f'(\hat{x}) \quad (1.2.3)$$

$$[\hat{x}, g(\hat{p})] = i\hbar g'(\hat{p}). \quad (1.2.4)$$

The first line in (1.2.4) can be derived by first noting that $[\hat{x}^n, \hat{p}] = i\hbar n\hat{x}^{n-1}$ for all $n \in \mathbb{N}$. Due to the linearity of the commutator, this yields the claim for any function f that can be written as a power series. The second line follows analogously.

From this one can straightforwardly conclude that

$$e^{iy\hat{p}/\hbar} \hat{x} e^{-iy\hat{p}/\hbar} = \hat{x} + y \quad (1.2.5)$$

$$e^{iq\hat{x}/\hbar} \hat{p} e^{-iq\hat{x}/\hbar} = \hat{p} - q, \quad (1.2.6)$$

which illuminates the connection of the CCR with translational motion: The momentum operator is the generator of translations and the position operator is the generator of (Galilean) boosts. Equivalently, we find the transformation of (improper) position and momentum eigen states

$$e^{iy\hat{p}/\hbar} |x\rangle = |x - y\rangle \quad (1.2.7)$$

$$e^{iq\hat{x}/\hbar} |p\rangle = |p + q\rangle \quad (1.2.8)$$

The advantage of the aforementioned properties is that they follow exclusively from the CCR and allow us to do many calculations on a very abstract level. In practice, however, we have to express the operators in a certain basis in order to carry out more

⁵Proof: Assume the opposite and take the trace on both sides of the equation leading to a contradiction

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complicated calculations. To this end, we can use the position representation

$$\langle x | \hat{x} | \psi \rangle = x \langle x | \psi \rangle \quad (1.2.9)$$

$$\langle x | \hat{p} | \psi \rangle = -i\hbar \frac{d}{dx} \langle x | \psi \rangle \quad (1.2.10)$$

or the momentum representation

$$\langle p | \hat{x} | \psi \rangle = i\hbar \frac{d}{dp} \langle p | \psi \rangle \quad (1.2.11)$$

$$\langle p | \hat{p} | \psi \rangle = p \langle p | \psi \rangle, \quad (1.2.12)$$

where in either case one operator becomes a multiplication and the other a differentiation on the space of square integrable functions. Both representations are connected via the Fourier transform,

$$\langle p | \psi \rangle = \langle p | \mathbb{1} | \psi \rangle = \int dx \langle p | x \rangle \langle x | \psi \rangle = \int \frac{dx}{\sqrt{2\pi\hbar}} e^{-ixp/\hbar} \langle x | \psi \rangle, \quad (1.2.13)$$

where it is easy to verify that

$$\langle x | p \rangle = \frac{1}{\sqrt{2\pi\hbar}} e^{ixp/\hbar} \quad (1.2.14)$$

is in fact the eigen function with eigenvalue p of the position space representation of the momentum operator (1.2.10).

1.2.2. Time dependent scattering theory at a potential

Many physical interactions appear in the form of a collision, where two particles approach each other, interact over a short time span, and then get spatially separated due to their motion. The time evolution of the two particles can then, in principle, be exactly solved in form of the Schrödinger equation, discussed in Subsection 1.1.4. However, in many cases, our primary interest lies not in the detailed time evolution, but in the overall change in the trajectory and internal state of the particles resulting from the entire interaction. In such cases, *scattering theory* is a sophisticated method to single out this net effect of the interaction. In this and the following Subsections we will derive the scattering operator for a stationary particle at the origin interacting with a moving particle and discuss its properties.

Consider a particle with a free Hamiltonian $\hat{H}_0$ and an interaction Hamiltonian $\hat{V}$,

$$\hat{H} = \hat{H}_0 + \hat{V}. \quad (1.2.15)$$

The free Hamiltonian contains the kinetic term $\hat{\mathbf{p}}^2/(2m)$, while the interaction Hamiltonian represents a position dependent potential that might also introduce changes of the internal states of the particles, $\langle \mathbf{x} | \hat{V} | \mathbf{y} \rangle \propto \delta^{(3)}(\mathbf{x} - \mathbf{y})$. In many physical scenarios in

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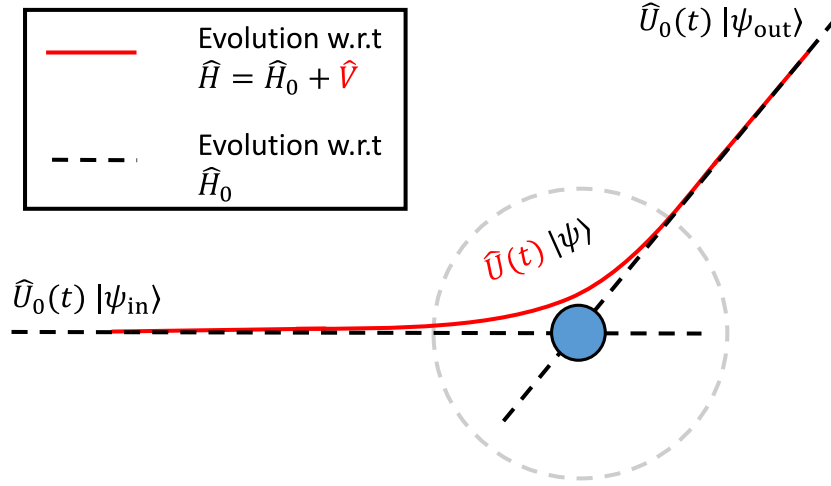


Figure 1.1.: Scattering of a state $|\psi\rangle$ at a Potential. The red line represents the trajectory of a state evolving with respect to the full Hamiltonian $\hat{H}$. The horizontal dashed line is the in-asymptote which is approached by the red line in the distance past. The other dashed line represents the out-asymptote that is approached by the red line in the distant future.

which the particle interacts with a physical target or scatterer, the potential can be assumed to be localized to a region around the origin where the scatterer is placed. Far away from that region, we can assume the particle to behave approximately like a free particle. To this end, we define two time evolution operators,

$$\hat{U}_0(t) = \exp\left(-\frac{i}{\hbar}\hat{H}_0 t\right) \quad (1.2.16)$$

$$\hat{U}(t) = \exp\left(-\frac{i}{\hbar}(\hat{H}_0 + \hat{V})t\right), \quad (1.2.17)$$

where the first describes the free time evolution and the latter the full time evolution. With these operators in mind, we can understand the trajectory of the particle state as depicted in Fig. 1.1: Consider a particle evolving with respect to the full theory, depicted by the red line. Close to the scattering center, the blue dot at the origin, the particle interacts with the potential and its state gets 'deflected' off the free trajectory. In the distant past however, the particle must have evolved almost freely because the potential had been negligible. Thus, there must have been a state $|\psi_{\text{in}}\rangle$, called the asymptotic in-state, and a time $t = -T$ such that $\hat{U}_0(-T)|\psi_{\text{in}}\rangle \approx \hat{U}(-T)|\psi\rangle$, or to be more precise,

$$|\psi\rangle = \lim_{t \rightarrow -\infty} \hat{U}^\dagger(t) \hat{U}_0(t) |\psi_{\text{in}}\rangle = \hat{\Omega}_+ |\psi_{\text{in}}\rangle, \quad (1.2.18)$$

where we defined the so-called Møller operator Ω_+ . This operator maps the asymptotic in-state onto the state $|\psi\rangle$ that evolves according to the full Hamiltonian. In Fig. 1.1, this corresponds to a jump from the horizontal dashed line onto the red curved line. Analogously, we can find the asymptotic out-state in the distant future and define the

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respective Møller operator as

$$|\psi\rangle = \lim_{t \rightarrow +\infty} \hat{U}^\dagger(t) \hat{U}_0(t) |\psi_{\text{out}}\rangle = \hat{\Omega}_- |\psi_{\text{out}}\rangle, \quad (1.2.19)$$

which corresponds to jump from the inclined dashed line onto the red line. In summary, we have defined two operators that map asymptotic states onto the state evolving under the full theory,

$$|\psi\rangle = \hat{\Omega}_\pm |\psi_{\text{in/out}}\rangle. \quad (1.2.20)$$

The key idea is now to invert the $\hat{\Omega}_-$ operator in order to get a connection between the in- and out-asymptote,

$$|\psi_{\text{out}}\rangle = \hat{\Omega}_-^\dagger \hat{\Omega}_+ |\psi_{\text{in}}\rangle, \quad (1.2.21)$$

which extracts the net effect of the scattering by jumping from the horizontal dashed line to the inclined dashed line in Fig. 1.1. Before we can do so, we have to be careful with the inversion of $\hat{\Omega}_-$: If the potential $\hat{V}$ admits bound states, by definition, a subset of our Hilbert space $\mathcal{H}_{\text{Bound}}$ will not have asymptotic out-states and $\hat{\Omega}_-^\dagger$ is not defined⁶. Fortunately, most physically motivated potentials fulfill the condition of *asymptotic completeness*, which states that the set of asymptotic in-states coincides with the set of asymptotic out-states and is orthogonal to the set of bound states,

$$\mathcal{H} = \mathcal{H}_{\text{Asympt}} \oplus \mathcal{H}_{\text{Bound}}. \quad (1.2.22)$$

Thus, we get the following chain of mappings,

$$\mathcal{H} \xrightarrow{\hat{\Omega}_+} \mathcal{H}_{\text{Asympt}} \xrightarrow{\hat{\Omega}_-^\dagger} \mathcal{H}, \quad (1.2.23)$$

where $\hat{\Omega}_-^\dagger$ is exactly defined on the range of $\hat{\Omega}_+$. Hence, the object $\hat{S} = \hat{\Omega}_-^\dagger \hat{\Omega}_+$ is a well defined linear map from $\mathcal{H}$ in itself. We call it the *scattering operator*, which directly connects the in- to the out-asymptote,

$$|\psi_{\text{out}}\rangle = \hat{S} |\psi_{\text{in}}\rangle. \quad (1.2.24)$$

The scattering operator is, as a concatenation of the norm preserving Møller operators, norm preserving and thus, as a map from $\mathcal{H}$ in itself, a unitary map.

Physically speaking, the scattering operator solves the particle interaction in an input output formalism without considering the details of the evolution while the particles interact.

⁶Mathematically speaking, the Møller operators are in general only isometric ($\hat{\Omega}^\dagger \hat{\Omega} = \mathbf{1}$) but not necessarily unitary ($\hat{\Omega}^\dagger \hat{\Omega} = \hat{\Omega} \hat{\Omega}^\dagger = \mathbf{1}$). Note that isometric, non-unitary operators are exclusive to infinite-dimensional spaces.

1.2.3. Scattering operator: Abstract properties

In the last subsection, we introduced the scattering operator $\hat{S}$ as the unitary map between asymptotic in- and out-states. While being the core concept of scattering theory, the exact calculations can be hard to perform and are often practically impossible. This makes the following collection of abstract properties, which are generally true, especially useful.

First, we note that, instead of using the Møller operators, we can define the scattering operator directly in terms of free and full time evolution

$$\hat{S} = \lim_{t \rightarrow \infty} \hat{U}_0^\dagger(t) \hat{U}(2t) \hat{U}_0^\dagger(t) \quad (1.2.25)$$

by writing out the Møller operators and unifying the two limits. From here we can derive the very useful form

$$\hat{S} = \mathcal{T} \exp \left(-\frac{i}{\hbar} \int_{-\infty}^{+\infty} dt \hat{V}^{(I)}(t) \right), \quad (1.2.26)$$

where $\hat{V}^{(I)}(t)$ is the interaction operator in interaction picture and $\mathcal{T}$ is the time ordering operator, as it is derived in Appendix A.1.1. The advantage of this form is that, for weak interactions, we can expand the exponential in its power series and, in principle, get approximations to arbitrary high precision by integrating term by term. An important property of the scattering operator is how it relates the free and full time evolution. On operator level, this reads

$$\begin{aligned} \hat{U}(\tau) \hat{\Omega}_\pm &= e^{-i\hat{H}\tau/\hbar} \lim_{t \rightarrow \mp\infty} \left(e^{i\hat{H}t/\hbar} e^{-i\hat{H}_0 t/\hbar} \right) = \lim_{t \rightarrow \mp\infty} \left(e^{i\hat{H}(t-\tau)/\hbar} e^{-i\hat{H}_0 t/\hbar} \right) \\ &= \lim_{t' \rightarrow \mp\infty} \left(e^{i\hat{H}t'/\hbar} e^{-i\hat{H}_0(t'+\tau)/\hbar} \right) = \lim_{t' \rightarrow \mp\infty} \left(e^{i\hat{H}t'/\hbar} e^{-i\hat{H}_0 t'/\hbar} \right) e^{-i\hat{H}_0 \tau/\hbar} \\ &= \hat{\Omega}_\pm \hat{U}_0(\tau), \end{aligned} \quad (1.2.27)$$

where we absorbed the full time evolution in the limit, shifted the variable of the limit by $t' = t - \tau$ and then factored the free time evolution on the right side of the limit. Taking the derivative with respect to τ on both sides of the equation yields the *intertwining relation*,

$$\hat{H} \hat{\Omega}_\pm = \hat{\Omega}_\pm \hat{H}_0. \quad (1.2.28)$$

From this we can immediately conclude,

$$\begin{aligned} \hat{H}_0 \hat{S} &= \hat{H}_0 \hat{\Omega}_-^\dagger \hat{\Omega}_+ = \left(\hat{\Omega}_- \hat{H}_0 \right)^\dagger \hat{\Omega}_+ = \left(\hat{H} \hat{\Omega}_- \right)^\dagger \hat{\Omega}_+ = \hat{\Omega}_-^\dagger \hat{H} \hat{\Omega}_+ \\ &= \hat{\Omega}_-^\dagger \hat{\Omega}_+ \hat{H}_0 = \hat{S} \hat{H}_0, \end{aligned} \quad (1.2.29)$$

where we made use of the intertwining relation twice. In short, this tells us that the free Hamiltonian commutes with the scattering operator,

$$\left[\hat{S}, \hat{H}_0 \right] = 0. \quad (1.2.30)$$

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Hence, energy is conserved in the scattering process.

In literature, the scattering operator is typically expressed as

$$\hat{S} = \mathbb{1} + i\hat{T}, \quad (1.2.31)$$

where $\hat{T}$ is simply called the T-operator. From the unitarity of $\hat{S}$ follows,

$$\mathbb{1} = \hat{S}^\dagger \hat{S} = (\mathbb{1} - i\hat{T}^\dagger)(\mathbb{1} + i\hat{T}) = \mathbb{1} + i(\hat{T} - \hat{T}^\dagger) + \hat{T}^\dagger \hat{T}, \quad (1.2.32)$$

which relates the anti-hermitean part $\text{Im}\{\hat{T}\}$ of $\hat{T}$ to its modulus squared $\hat{T}^\dagger \hat{T}$,

$$\text{Im}\{\hat{T}\} = \frac{\hat{T} - \hat{T}^\dagger}{2i} = \hat{T}^\dagger \hat{T}/2. \quad (1.2.33)$$

This fact is known as the *optical theorem*.

1.2.4. Scattering operator: Matrix elements, scattering amplitude and cross sections

Up to now, we considered the generic scattering scenario described by the Hamiltonian in (1.2.15). Now, we explicitly assume a particle with an internal structure, e.g. an atom with an excited and a ground state. These internal energy levels are given by the operator $\hat{h}$, which is added to the kinetic term,

$$\hat{H}_0 = \frac{\hat{\mathbf{p}}^2}{2m} + \hat{h}, \quad (1.2.34)$$

where

$$\hat{h} = \sum_{\nu} \epsilon_{\nu} |\nu\rangle \langle \nu|. \quad (1.2.35)$$

With this, we already know the eigen basis of the free Hamiltonian

$$\hat{H}_0 |\mathbf{p}, \nu\rangle = E_{\nu}(\mathbf{p}) |\mathbf{p}, \nu\rangle = \left(\frac{\mathbf{p}^2}{2m} + \epsilon_{\nu} \right) |\mathbf{p}, \nu\rangle. \quad (1.2.36)$$

Together with the energy conservation of the scattering operator (1.2.30), we find

$$0 = \langle \mathbf{p}_2, \nu | [\hat{H}_0, \hat{S}] | \mathbf{p}_1, \kappa \rangle = (E_{\nu}(\mathbf{p}_2) - E_{\kappa}(\mathbf{p}_1)) \langle \mathbf{p}_2, \nu | \hat{S} | \mathbf{p}_1, \kappa \rangle. \quad (1.2.37)$$

Thus, the matrix element $\langle \mathbf{p}_2, \nu | \hat{S} | \mathbf{p}_1, \kappa \rangle$ has to be zero unless $E_{\nu}(\mathbf{p}_2) = E_{\kappa}(\mathbf{p}_1)$. This can be built in explicitly by setting the matrix element proportional to the energy-conserving delta function $\delta^{(1)}(E_{\nu}(\mathbf{p}_2) - E_{\kappa}(\mathbf{p}_1))$. Using the expression of the scattering

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operator in terms of the T-operator (1.2.31), we can write

$$\langle \mathbf{p}_2, \nu | \hat{T} | \mathbf{p}_1, \kappa \rangle = \frac{1}{2\pi\hbar m} \delta^{(1)}(E_\nu(\mathbf{p}_2) - E_\kappa(\mathbf{p}_1)) f_{\nu\kappa}(\mathbf{p}_2, \mathbf{p}_1), \quad (1.2.38)$$

which defines the so-called *scattering amplitude* $f_{\nu\kappa}(\mathbf{p}_2, \mathbf{p}_1)$. Due to the energy-conserving delta function, the length of the ingoing momentum $\mathbf{p}_1$ and the outgoing momentum $\mathbf{p}_2$ are linked by,

$$|\mathbf{p}_2|^2 = |\mathbf{p}_1|^2 - 2m(\epsilon_\nu - \epsilon_\kappa) = p_1^2 - 2m\Delta_{\nu\kappa}, \quad (1.2.39)$$

where we defined $p_1 = |\mathbf{p}_1|$ and the energy gap $\Delta_{\nu\kappa} = \epsilon_\nu - \epsilon_\kappa$ of the internal transition. In the case of $p_1^2 < 2m\Delta_{\nu\kappa}$, the initial kinetic energy is too small to overcome the internal energy gap and the transition is not possible. For convenience, we define the outgoing absolute momentum as a function of ingoing momentum and energy gap

$$P_{\nu\kappa}(p_1) = \sqrt{p_1^2 - 2m\Delta_{\nu\kappa}}, \quad (1.2.40)$$

if $p_1^2 > 2m\Delta_{\nu\kappa}$ and $P_{\nu\kappa}(\mathbf{p}_1) = 0$ else. To include that the outgoing momentum can still have any direction, we also define

$$\mathbf{P}_{\nu\kappa}(p_1, \Omega) = P_{\nu\kappa}(p_1) \mathbf{n}(\Omega), \quad (1.2.41)$$

where $\mathbf{n}(\Omega)$ is the unit vector in direction of the solid angle $\Omega = (\vartheta, \varphi)$. Now, we have the terminology to define the differential cross section

$$\frac{d\sigma_{\nu\kappa}(\mathbf{p}_1)}{d\Omega} = \frac{P_{\nu\kappa}(p_1)}{p_1} |f_{\nu\kappa}(\mathbf{P}_{\nu\kappa}(\mathbf{p}_1, \Omega), \mathbf{p}_1)|^2 \quad (1.2.42)$$

and the total cross section

$$\sigma_{\nu\kappa}(\mathbf{p}_1) = \int d\Omega \frac{d\sigma_{\nu\kappa}(\mathbf{p}_1)}{d\Omega}. \quad (1.2.43)$$

The significance of these objects lies in the fact that they are experimentally observable: Consider a current density $\mathbf{j}(\mathbf{p}_1)$ of particles with momentum $\mathbf{p}_1$ that are initially in internal state $|\kappa\rangle$, then the number of particles in internal state κ that hit a distant detector placed in the direction of Ω and covering the small solid angle $\Delta\Omega$ within time Δt is

$$\Delta N_{\nu\kappa}(\Omega, \mathbf{p}_1) = \frac{d\sigma_{\nu\kappa}(\mathbf{p}_1)}{d\Omega} |\mathbf{j}(\mathbf{p}_1)| \Delta\Omega \Delta t. \quad (1.2.44)$$

If we are not interested in the deflection angle, the total cross section tells us the number of particles that scattered at all in Δt and had an internal transition $|\kappa\rangle \rightarrow |\nu\rangle$,

$$\Delta N_{\nu\kappa}^{\text{total}}(\mathbf{p}_1) = \sigma_{\nu\kappa}(\mathbf{p}_1) |\mathbf{j}(\mathbf{p}_1)| \Delta t. \quad (1.2.45)$$

Note that this interpretation of the cross section has to be handled with some care: It is only true if the momentum wave function of the particles is so narrow that the scattering

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amplitude can be assumed to be constant over the support of the wave packet. It is also necessary that the incoming beam of particles is distributed uniformly over a sufficiently large area of the plane perpendicular to the incoming momentum. If these conditions are not met, the differential and total cross section are meaningless. In fact, throughout this thesis the cross section will appear in some calculations, but we will not have to rely on the afore mentioned interpretations. Therefore, we will skip the derivation and refer the reader to the literature [65].

While it can be, in general, hard to find exact expressions for the scattering operator or equivalently the scattering amplitude, we can infer further properties from the symmetries of the system. In fact, symmetries of the (full) Hamiltonian, i.e. $[\hat{H}, \hat{U}] = 0$ for some unitary $\hat{U}$, are inherited by the scattering amplitude. In Chapter 3 we will assume the system to fulfill *time reversal symmetry*, which we will also call *microreversibility*. In that case, the full Hamiltonian commutes with the so-called *time-reversal operator* that acts on a position space wave function by taking the complex conjugate,

$$\psi(\mathbf{x}) \mapsto \psi^*(\mathbf{x}). \quad (1.2.46)$$

This map constitutes a *anti-unitary* transformation, which has to be treated with some care. Without going into the details of the derivation, we refer the reader once more to [63] and note that the scattering amplitude fulfills,

$$f_{\nu\kappa}(\mathbf{p}_2, \mathbf{p}_1) = f_{\kappa\nu}(-\mathbf{p}_1, -\mathbf{p}_2) \quad (1.2.47)$$

in this case.

1.2.5. Born approximation

In general, calculating the scattering operator is as hard as solving the time evolution of the particle as it passes and interacts with the scatterer. Fortunately, we can employ the so-called Born approximation if the potential is weak or our scatterer is fast. To this end, we can parametrize the potential with a small, dimensionless parameter $\hat{V} \rightarrow \lambda \hat{V}$ and expand the scattering amplitude as a power series in λ ,

$$f_{\nu\kappa}(\mathbf{p}_2, \mathbf{p}_1) = \sum_{n=1}^{\infty} \lambda^n f_{\nu\kappa}^{(n)}(\mathbf{p}_2, \mathbf{p}_1). \quad (1.2.48)$$

In literature, there are sophisticated methods to iteratively solve the so-called *Lippmann–Schwinger equation* in order to derive arbitrarily high orders in the Born series [65]. Throughout this thesis, however, we will not need the Born approximations of higher order. This allows us to circumvent these methods and immediately expand (1.2.26) in the Dyson series

$$\hat{S} = \mathcal{T} \exp \left(-\frac{i}{\hbar} \int_{-\infty}^{+\infty} dt \lambda \hat{V}^{(I)}(t) \right) \approx \mathbb{1} - \lambda \frac{i}{\hbar} \int_{-\infty}^{+\infty} dt \hat{V}^{(I)}(t) + \mathcal{O}(\lambda^2). \quad (1.2.49)$$

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The matrix elements of the term proportional to λ are

$$\begin{aligned}
\langle \mathbf{p}_2, \nu | \int_{-\infty}^{+\infty} dt \hat{V}^{(1)}(t) | \mathbf{p}_1, \kappa \rangle &= \int_{-\infty}^{+\infty} dt \langle \mathbf{p}_2, \nu | \hat{U}_0(t)^\dagger \hat{V} \hat{U}_0(t) | \mathbf{p}_1, \kappa \rangle \\
&= \int_{-\infty}^{+\infty} dt \exp \left(\frac{i}{\hbar} (E_\nu(\mathbf{p}_2) - E_\kappa(\mathbf{p}_1)t) \right) \langle \mathbf{p}_2, \nu | \hat{V} | \mathbf{p}_1, \kappa \rangle \\
&= 2\pi\hbar\delta^{(1)}(E_\nu(\mathbf{p}_2) - E_\kappa(\mathbf{p}_1)) \langle \mathbf{p}_2, \nu | \hat{V} | \mathbf{p}_1, \kappa \rangle,
\end{aligned} \tag{1.2.50}$$

where we used the Fourier integral representation of the delta function. Comparing this first order T operator with (1.2.38) yields the scattering amplitude in first-order Born approximation,

$$f_{\nu\kappa}^{(1)}(\mathbf{p}_2, \mathbf{p}_1) = -(2\pi)^2 \hbar m \langle \mathbf{p}_2, \nu | \hat{V} | \mathbf{p}_1, \kappa \rangle. \tag{1.2.51}$$

1.2.6. Delta regularization

Physical models are often significantly simplified by idealizations that are directly linked to mathematical limits: For example, crystals and gases are assumed to fill an infinite space in order to neglect boundary conditions, derivatives assume an infinitely fine resolution in time and space, and in the scattering operator an infinite interaction time is assumed. In general, different limits can not be interchanged and the wrong order can lead to ill-defined expressions. The goal of this subsection is to discuss the energy conserving delta function in the matrix elements from the T -operator, which is a sinc-function in the limit of large interaction times. We show how understanding this limit allows the regularization of a squared delta function.

Consider the delta distribution, which is a well-defined mathematical object that maps so-called *test functions* onto their value at $x = 0$

$$f \mapsto f(0). \tag{1.2.52}$$

This operation can be realized as the limit of integral operators

$$\lim_{n \rightarrow \infty} \int_{-\infty}^{\infty} dx g_n(x) f(x) = f(0), \tag{1.2.53}$$

where g_n is, for example, a sequence of centered Gaussians with standard deviation $1/n$. The larger n , the sharper the peak of g_n around $x = 0$. This motivates the definition of the *delta function* $\delta^{(1)}$ that is thought of as being infinitely large at $x = 0$ and zero everywhere else, such that its integral equals 1. While such a function cannot exist mathematically, there is a reason for its success in physics: This reason is the existence of relevant scales in physical theories. In classical mechanics, for example, the length scales are far beyond the size of an atom. If we now choose the "delta function" to be a Gaussian with standard deviation of the size of an atom, this is mathematically a smooth function but practically infinitely sharp on the relevant length scales of mechanical motion.

For the reasons mentioned above, the use of the idealized delta function is unproblem-

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atic in most cases. Unfortunately, throughout this thesis, we will encounter situations in which the idealization is too strong. Then, ill-defined objects like squared delta functions appear. However, if we know which limit led to the problematic expression, it is still possible to *regularize* these objects and turn them into a physically meaningful expression.

An example for such a procedure, which will be relevant Subsection 3.2.4, stems from the following Fourier representation of an energy delta function:

$$\delta^{(1)}(E) = \lim_{T \rightarrow \infty} \int_{-T/2}^{T/2} \frac{dt}{2\pi\hbar} e^{iEt/\hbar} = \lim_{T \rightarrow \infty} \frac{1}{\pi E} \sin\left(\frac{ET}{2\hbar}\right) = \lim_{T \rightarrow \infty} \delta_T(E). \quad (1.2.54)$$

In the course of the calculation, this delta function gets squared to

$$[\delta^{(1)}(E)]^2. \quad (1.2.55)$$

If one considers the original definition (1.2.52), the meaning of this object remains unclear. The problem can be attributed to the idealization that one lets the relevant evolution time T in a physical setting go to infinity. Hence, the remedy amounts to considering a finite, but sufficiently large T instead, so that the peak of $\delta_T(E)$ is narrow compared to the relevant energy scales of the physical problem. Inserting a physical test function that varies only over these scales, we can approximate $f(E)\delta_T^{(1)}(E) \approx f(0)\delta_T^{(1)}(E)$ and obtain,

$$\begin{aligned} \int_{-\infty}^{\infty} dE [\delta^{(1)}(E)]^2 f(E) &\mapsto \int_{-\infty}^{\infty} dE [\delta_T^{(1)}(E)]^2 f(E) \approx f(0) \int_{-\infty}^{\infty} dE [\delta_T^{(1)}(E)]^2 \\ &= f(0) \int_{-\infty}^{\infty} dE \left[\frac{1}{\pi E} \sin\left(\frac{ET}{2\hbar}\right) \right]^2 = \frac{T}{2\pi\hbar} f(0). \end{aligned} \quad (1.2.56)$$

In this sense the squared delta function got regularized and now reads as

$$[\delta^{(1)}(E)]^2 = \frac{T}{2\pi\hbar} \delta^{(1)}(E). \quad (1.2.57)$$

The normalization of the squared delta function can also be read as the definition of $\delta^{(1)}(0)$ since we can naively use one delta function to set the argument of the other delta function to zero

$$[\delta^{(1)}(E)]^2 = \delta^{(1)}(0) \delta^{(1)}(E) \quad (1.2.58)$$

and compare.

In Chapter 3, we encounter several ill-defined objects that include $\delta^{(1)}(0)$ in the scattering derivations. With the results of this subsection at hand, we will regularize those expression and obtain finite and physical expressions.

1.3. Open system dynamics

The theory of open quantum systems includes any situation, where a system of interest interacts with another, typically larger in size and uncontrolled, system that we call *environment* [13,66,67]. In this context, we refer to the system of interest as *open system*. While system and environment combined can be assumed to be closed, a solution of the Schrödinger equation for the composite system is, in most cases, practically impossible, due to the size of the environment.

In order to circumvent the calculation of the full dynamics, phenomenological descriptions of the dynamics of the open system have been developed. Due to incomplete knowledge of the environment, any description is unavoidably probabilistic in some sense.

Throughout this section we will recall all concepts from the theory of open quantum systems that will become important later. Starting with classical Markov processes and local detailed balance in Subsection 1.3.1. Then, we discuss channels and dynamical semi-groups in Subsection 1.3.2. Finally, we discuss quantum master equations in Subsection 1.3.3.

1.3.1. Markov processes and local detailed balance

In this subsection, we introduce classical stochastic processes. We will not only need these in Chapter 3, but they also serve as a classical analogy for channels and master equations, discussed in the following subsections. A detailed treatment of the matter can be found, for example, in [13,68]. Consider a classical system with N discrete states. We can express our knowledge about the system state by a probability distribution $(p_n)_n$. In general, this distribution can change in time and we express this stochastic transformation with a transition matrix $\mathbb{T}(t, t_0)$ evolving the distribution from time t_0 to t ,

$$p_n(t) = \sum_k \mathbb{T}_{nk}(t, t_0) p_k(t_0). \quad (1.3.1)$$

In order to guarantee positivity and normalization at any point in time, the transition matrix has to fulfill

$$\mathbb{T}_{nk}(t, t_0) \geq 0, \quad \sum_n \mathbb{T}_{nk}(t, t_0) = 1 \quad (1.3.2)$$

for all n, k, t, t_0 . We call this evolution of probabilities a *stochastic process*. We further call it *time-homogeneous* if $\mathbb{T}(t, t_0)$ only depends on the distance of the points in time $t - t_0$. A process fulfilling the *semi-group property*,

$$\mathbb{T}(t, t_0) = \mathbb{T}(t, t_1) \mathbb{T}(t_1, t_0), \quad \forall t > t_1 > t_0, \quad (1.3.3)$$

is called *Markovian* or *memory-less*. For a Markovian, time-homogeneous stochastic process we can define time-independent rates,

$$\gamma_{nk} = \lim_{\Delta t \rightarrow 0} \frac{\mathbb{T}_{nk}(t + \Delta t, t) - \delta_{nk}}{\Delta t}, \quad (1.3.4)$$

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and obtain a differential equation for the probabilities

$$\dot{p}_n(t) = \sum_{k \neq n} (\gamma_{nk} p_k(t) - \gamma_{kn} p_n(t)). \quad (1.3.5)$$

Such an equation is referred to as a *master equation* or *rate equation*. In order to avoid confusion with the notion of a quantum master equation, we will stick to the latter name. Note that the conservation of probability is automatically fulfilled in (1.3.5).

A probability distribution $(\tilde{p}_k)_k$ is a *steady state*, *fix point*, or *equilibrium* state of the process, if it makes the right hand side of (1.3.5) vanish and thus does not evolve in time. A stronger sufficient condition for a steady state is the so called *detailed balance* condition that requires a balance of the probability flow between each pair of states,

$$\gamma_{nk} \tilde{p}_k = \gamma_{kn} \tilde{p}_n. \quad (1.3.6)$$

A special case of detailed balance, often encountered in statistical physics, is the so-called *local detailed balance*,

$$\frac{\gamma_{nk}}{\gamma_{kn}} = \exp \left(-\frac{E_n - E_k}{k_B T} \right), \quad (1.3.7)$$

where E_n are the energies of the respective states and T is the temperature. This ensures that the *Gibbs distribution*,

$$\tilde{p}_n = \exp \left(-\frac{E_n}{k_B T} \right) / Z, \quad (1.3.8)$$

is a steady state of the process. This distribution will be discussed in Subsection 1.4.2.

1.3.2. Quantum channels

While the unitary time evolution in quantum mechanics, governed by the Schrödinger equation, is assumed to be fundamentally true, it can be beneficial to consider more general transformations. This is especially the case when our system of interest S is not isolated but coupled to an environment E of uncontrolled degrees of freedom. Then system and environment together can be considered to be a closed quantum system and the Schrödinger equation is applicable. After the joint evolution from time t_0 to time t with the time evolution operator $\hat{U}(t, t_0)$, the environment can be traced out and we find,

$$\hat{\rho}_S(t) = \text{tr}_E \left\{ \hat{U}(t, t_0) \hat{\rho}_{SE}(t_0) \hat{U}^\dagger(t, t_0) \right\}. \quad (1.3.9)$$

This map from state $\hat{\rho}_S(t_0) = \text{tr}_E \{ \hat{\rho}_{SE}(t_0) \}$ to state $\hat{\rho}_S(t)$ is called a *quantum channel* and we denote it by $\Phi(t, t_0)$. It is the quantum analogue of the stochastic transformation from Subsection 1.3.1 mapping quantum states onto quantum states. In order to do so, it has to be trace preserving and completely-positive⁷. In [69], Kraus proved that any

⁷This enforces that the state remains positive even when the channel is just applied to a sub system, e.g. $(\mathbb{1} \otimes \Phi(t, t_0)) \hat{\rho} \geq 0$. Surprisingly, this is not automatically true for positive maps.

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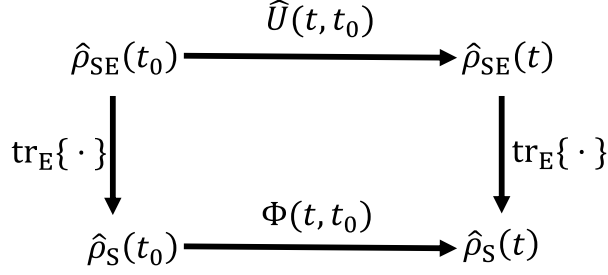


Figure 1.2.: Commutative diagram of an open quantum systems: The goal is to describe the evolution of a system S, which is in contact with an environment E. A direct way from t_0 to t is to evolve system and environment together with the unitary evolution $\hat{U}(t, t_0)$, according to the Schrödinger equation, and then trace out the environment. The same transformation can be achieved by first tracing out the environment and applying the corresponding channel $\Phi(t, t_0)$.

channel can be written in terms of so-called *Kraus operators*,

$$\Phi(t, t_0) [\hat{\rho}_S] = \sum_j \hat{K}_j(t, t_0) \hat{\rho}_S \hat{K}_j^\dagger(t, t_0), \quad (1.3.10)$$

which obey the normalization condition

$$\sum_j \hat{K}_j^\dagger(t, t_0) \hat{K}_j(t, t_0) = \mathbb{1} \quad (1.3.11)$$

in order to preserve the trace of the state. In the finite dimensional case, where $\Phi(t, t_0)$ maps from states on a d_{in} -dimensional space onto states on a d_{out} -dimensional space, the maximal number of Kraus operators is bounded by the product $d_{\text{in}} d_{\text{out}}$. As depicted in Fig. 1.2 a channel is basically a shortcut that encodes the effective evolution of the system. However, the direct calculation via (1.3.9) is as complicated as solving the full time evolution and approximations have to be made, as we will do in Chapters 3 and 4. Deeper discussions and further properties of channels can be found in [59, 70].

The family of Channels indexed by initial and final time, $\Phi(t, t_0)$, is the quantum analogue of a stochastic process and can be characterized accordingly: We call it *time-homogeneous* when it only depends on the difference $t - t_0$ and we say it is *Markovian*, a *dynamical semi-group*, or *CP-divisible* if it can be split like,

$$\Phi(t, t_0) = \Phi(t, t_1) \circ \Phi(t_1, t_0), \quad \forall t > t_1 > t_0. \quad (1.3.12)$$

Up to now, we used that an expression of the form (1.3.9) yields a Channel. In fact, the opposite is also true: According to the *Stinespring dilation theorem* [71], for any Channel Λ , there is a unitary $\hat{U}$ and an ancilla $\hat{\rho}_A$ such that,

$$\Lambda[\hat{\rho}] = \text{tr}_A \left\{ \hat{U} (\hat{\rho} \otimes \hat{\rho}_A) \hat{U}^\dagger \right\}, \quad \forall \hat{\rho}. \quad (1.3.13)$$

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Hence, *repeated interaction models*⁸ [51] that describe the evolution of quantum systems by the concatenation of maps of the form (1.3.13) are, in principle, capable of producing any possible quantum evolution. However, as we will discuss in Chapter 3, such operational reconstructions of open system dynamics must be tailored to the specifics of the system in order to achieve a desired outcome (e.g., thermalization). Conversely, unphysical behavior may arise if only a limited, albeit physically motivated, set of ancilla states and unitary operations is available.

Under certain conditions, often met in physics, the evolution given by a dynamical semi-group, $\Phi(t, t_0)$, can still be derived from a differential equation. This will be discussed in the next section.

1.3.3. Master equations

A dynamical semi-group of the form (1.3.10) is usually the solution of a complicated dynamical problem. Here, we present the general form of the generator of dynamical semi-groups that allows us to derive $\Phi(t, t_0)$ from a differential equation.

Gorini, Kossakowski, and Sudarshan [72], on the one hand, and Lindblad [73], on the other hand, found the generator of dynamical semi-groups at the same time: For a continuous, time-homogeneous dynamical semi-group, they derived the explicit form of the time-independent generator,

$$\mathcal{L} = \lim_{\Delta t \rightarrow 0} \frac{\Phi(t + \Delta t, t) - \text{id}}{\Delta t}, \quad (1.3.14)$$

where id is the identity channel. With the generator at hand, the dynamical semi-group can be obtained as the solution of the initial value problem,

$$\frac{d}{dt}\Phi(t, t_0) = \mathcal{L} \circ \Phi(t, t_0), \quad \Phi(t_0, t_0) = \text{id}. \quad (1.3.15)$$

Applied to the system state $\hat{\rho}_S$, we find the differential equation,

$$\frac{d}{dt}\hat{\rho}_S = -\frac{i}{\hbar} [\hat{H}, \hat{\rho}_S] + \sum_j \gamma_j \left(\hat{L}_j \hat{\rho}_S \hat{L}_j^\dagger - \frac{1}{2} \left\{ \hat{L}_j^\dagger \hat{L}_j, \hat{\rho}_S \right\} \right). \quad (1.3.16)$$

Here, $\hat{H}$ is a self-adjoint operator, $\hat{L}_j$ are called *jump operators* or *Lindblad operators* and γ_j are rates determining how strong the processes corresponding to $\hat{L}_j$ are. The first part of the equation resembles the Schrödinger equation and generates unitary evolution. The second part, on the other hand, is called *dissipator* and generates (in general) non-unitary dynamics. It is convenient to introduce the short hand notation,

$$\mathcal{D}[\hat{L}_j] \hat{\rho}_S = \left(\hat{L}_j \hat{\rho}_S \hat{L}_j^\dagger - \frac{1}{2} \left\{ \hat{L}_j^\dagger \hat{L}_j, \hat{\rho}_S \right\} \right). \quad (1.3.17)$$

⁸We try to avoid the notion *collision models* in this context to avoid confusion with the scattering based models in Chapter 3

The full equation (1.3.16) is known as the *GKLS-master equation* or often just *Lindblad equation*. It is of great importance in the field of open quantum systems [13].

The exact form of the Lindblad operators could be inferred heuristically or one often arrives at it with help of several approximations starting from an underlying microscopic theory. In Chapter 3, we derive the master equation of a quantum system subjected to a thermal gas environment.

1.4. Thermodynamics

Driven by the desire to better understand and improve the steam engine, Sadi Carnot lay the foundation of classical thermodynamics in 1824 [74]. From the modern point of view, however, the theory is not restricted to gases and steam and describes general macroscopic systems in equilibrium based on universal postulates that restrict thermodynamic processes [75]. For the laws of thermodynamics to apply, it was only required that a system be macroscopic. This changed with the seminal work of Scovil and Schulz-Du Bois [76] who interpreted the three-level maser as a thermal machine, exemplifying the utility of thermodynamics even for microscopic quantum systems and is often seen as the birth of *quantum thermodynamics* (QTD).

In this field, the concepts of thermodynamics are applied to quantum systems, which introduces a variety of open questions: In classical thermodynamics, the systems are assumed to equilibrate so fast that we can assume, for all practical purposes, the systems to be constantly in equilibrium [75]. With quantum mechanics, however, we have a theory that explicitly describes the system dynamics. Hence, models are required that properly describe the thermalization of quantum systems in order to consistently combine quantum theory and thermodynamics. This is, for example, explored in the weak coupling limit [14], but remains unclear for collisional models.

Another branch of QTD considers thermal machines with quantized working media. Quantum effects are singled out by comparing the quantum machine with its classical counterpart, which is usually understood as running the machine without coherences in the initial state. Here, an open problem is a proper definition of quantum work.

The goal of this section is to provide the reader with the necessary thermodynamic preliminaries to go through chapters 2 and 3. To this end, we set the stage with the discussion of classical, phenomenological thermodynamics in Subsection 1.4.1. Next, we discuss the microscopic description in Subsection 1.4.2. In the last subsection (Subsection 1.4.3) we discuss different notions of quantum work.

1.4.1. Phenomenological thermodynamics

Here, we review the most important concepts of classical thermodynamics, as described in [75, 77]. Phenomenological thermodynamics is an empirical theory that relates macroscopic quantities with each other, without further consideration of the underlying theory. A macroscopic portion of a gas, for example, consists easily of $N \sim 10^{23}$ particles, each with three position and three momentum degrees of freedom. Surprisingly, the number

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of relevant macroscopic observables such as particle number N , (empirical) temperature T , pressure p , and volume V is very small, compared to the $6N$ -dimensional microscopic phase space. A further reduction in complexity comes from the fact that the systems equilibrate fast on macroscopic time scales, in the sense that the relevant observables become constant. In equilibrium, the macroscopic observables are no longer independent but have to fulfill so-called equations of state

$$f(N, T, p, V) = 0. \quad (1.4.1)$$

In case of an ideal gas, this reads

$$pV = Nk_B T. \quad (1.4.2)$$

Hence, we can use any three observables of (N, T, p, V) , also called *state variables*, as coordinates on the manifold of equilibrium states. The remaining observable becomes a function on the manifold by virtue of the ideal gas law (1.4.2).

The existence of temperature is already an implicit assumption that is nowadays known as the *zeroth law of thermodynamics*: For two systems A and B , which are in thermal contact, we can define a relation that states $A \sim B$ if and only if the systems are in thermal equilibrium, i.e. the macroscopic observables do not change anymore. The zeroth law states that this relation is an equivalence relation. Besides the trivial properties of reflexivity, i.e. being in equilibrium with itself $A \sim A$, and symmetry, i.e. $A \sim B$ is equivalent to $B \sim A$, it postulates transitivity: If $A \sim B$ and if $B \sim C$, then automatically $A \sim C$. This is a necessary prerequisite in order to build a thermometer and define an empirical notion of temperature. Here, system B could be a mercury thermometer in which the liquid mercury expands as it equilibrates with system A . By means of the zeroth law, we can now conclude that $A \sim C$ if and only if the mercury expands the same amount when in contact with system C . The expansion of the liquid mercury can be read-off from a linear scale that we call, up to a proper choice of unit, (empirical) temperature T .

Next, we discuss transformations of the thermodynamic state. Since phenomenological thermodynamics is based on the concept of equilibrium, a consistent description requires changes to be *quasi-static*. This assumes the change to be so slow that the system is practically in equilibrium at any point in time. After choosing coordinates, say (T, V) for a gas with constant particle number N , we can express the change of any function in the language of differential forms. For example, the pressure p changes as a function of T and V according to

$$dp = \frac{Nk_B}{V} \left(dT - T \frac{dV}{V} \right). \quad (1.4.3)$$

The total pressure difference can be obtained by integrating this expression over a path in the manifold of states. Since pressure p itself is a state variable, its value does only depend on the coordinates and not on the integration path. As a direct consequence,

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any integral of dp over a closed curve is zero,

$$\oint dp = 0. \quad (1.4.4)$$

Since dp is the total differential of a function $p(T, V)$ it is called an *exact differential*. A general differential,

$$dK(T, V) = a(T, V)dT + b(T, V)dV, \quad (1.4.5)$$

is only exact if ⁹

$$\frac{\partial a(T, V)}{\partial V} - \frac{\partial b(T, V)}{\partial T} = 0. \quad (1.4.6)$$

If that is not true, the differential is *inexact*, denoted by the bar in $\bar{d}$, and the value of integrals depends on the chosen path. A discussion of differential forms can be found in the thermodynamic literature [75] or more in-depth in literature about differential geometry [78].

An important, and in general inexact, differential is the differential of work dW , which is known from mechanics [79] to be “force times displacement”,

$$dW = \mathbf{F} \cdot d\mathbf{x}. \quad (1.4.7)$$

For a cylinder of gas which is compressed by a piston, this translates to,

$$dW = -p dV, \quad (1.4.8)$$

with the sign convention that work is positive if energy flows into the system. If the system is isolated and the piston compresses the gas, the state of the system changes, here the temperature increases. The same can be observed when the piston is fixed in position and the cylinder is held over a flame. This makes evident that there is another change in energy dQ , which we call *heat*. The statement of the *first law of thermodynamics* is that the sum of the two inexact differentials of work and heat yields an exact differential,

$$dU = dW + dQ, \quad (1.4.9)$$

where dU is the change in the so-called *internal energy*. Since the differential is exact, integrals over dU do not depend on the path taken and define, up to a constant, a state function U . Hence, the first law states that the change in the energy of a system is exactly the amount of energy that entered the system in terms of work and heat. Put differently, energy does not get created or destroyed but can only be exchanged. This is a manifestation of energy conservation.

The first law narrows down which processes are energetically possible in nature. Note that the law is symmetric under time reversal, meaning a process is energetically possible if and only if its reverse is also energetically possible. However, violations of this symmetry are frequently seen in the macroscopic world: Heat always flows from hot to

⁹Note that this is a generalization of the fact that vector field can be expressed as the gradient of a scalar field if the rotation of the vector field vanishes.

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cold, gases always expand into the full space available, and friction stops any relative motion, while the reverse processes are not observed in nature. Hence, there must a law dictating in which direction certain processes can occur. This law is called the *second law of thermodynamics*, which reads in the version by Clausius “No process is possible whose sole result is the transfer of heat from a colder to a hotter body” [75].

In order to phrase the second law as a mathematical statement, we need some preparation: Assume we have experimental control over the state coordinates of our system and change these slightly. If system and environment return without any change to the initial state when we reverse the change of the parameters, the described process is called *reversible*. We can quantify the irreversibility of processes with help of the empirically tested *Clausius theorem*,

$$\oint_{\Gamma} \frac{\mathrm{d}Q}{T} \leq 0, \quad (1.4.10)$$

which holds for any thermodynamic cycle Γ , i.e., a closed loop of states. If this cycle Γ is reversible, we can apply Clausius theorem to the reverse cycle $-\Gamma$, which flips the sign of the heat,

$$\oint_{-\Gamma} \frac{\mathrm{d}Q}{T} = - \oint_{\Gamma} \frac{\mathrm{d}Q}{T} \leq 0. \quad (1.4.11)$$

Combining this with the original inequality (1.4.10), we find,

$$\oint_{\Gamma} \frac{\mathrm{d}Q}{T} = 0 \quad (1.4.12)$$

for reversible processes. Hence, we can define a function S on the manifold of states by,

$$S(2) - S(1) = \int_{\Gamma_{\text{rev}}(2,1)} \frac{\mathrm{d}Q}{T}, \quad (1.4.13)$$

given a reversible path, $\Gamma_{\text{rev}}(2, 1)$, between the states $2 \leftarrow 1$. This is well-defined because for another reversible path $\Gamma'_{\text{rev}}(2, 1)$ we find,

$$\int_{\Gamma_{\text{rev}}(2,1)} \frac{\mathrm{d}Q}{T} - \int_{\Gamma'_{\text{rev}}(2,1)} \frac{\mathrm{d}Q}{T} = \oint_{\Gamma_{\text{rev}}(2,1) + \Gamma'_{\text{rev}}(1,2)} \frac{\mathrm{d}Q}{T} = 0, \quad (1.4.14)$$

where we combined both paths to form a reversible cycle.

It is customary to use the differential form language denoting reversible heat exchange by a subscript,

$$\mathrm{d}S = \frac{\mathrm{d}Q_{\text{rev}}}{T}. \quad (1.4.15)$$

The so defined state function (up to an additive constant) is called *entropy*. If we now consider an arbitrary path between $2 \leftarrow 1$, we can complement it with a reversible path and apply Clausius theorem on the concatenation to find,

$$S(2) - S(1) = \int_1^2 \frac{\mathrm{d}Q_{\text{rev}}}{T} \geq \int_1^2 \frac{\mathrm{d}Q}{T}. \quad (1.4.16)$$

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The right hand side can be interpreted as the entropy exchange with the environment. We see that, in general, additional entropy is produced. We can complement the inequality by the missing term and write,

$$dS = dS_{\text{prod}} + \frac{dQ}{T} \geq \frac{dQ}{T}. \quad (1.4.17)$$

Since the left hand side is an exact differential, an integral over any closed loop shows the path-dependent entropy produced in one cycle,

$$S_{\text{prod}} = - \oint \frac{dQ}{T} \geq 0. \quad (1.4.18)$$

The fact that the entropy of a thermally isolated system always increases, i.e. $dS \geq 0$, breaks time reversal symmetry and is thus a mathematical manifestation of the second law of thermodynamics.

We now combine the first (1.4.9) and the second law (1.4.17) in order to bound the maximal work output in isothermal processes. We find,

$$dU \leq TdS + dW, \quad (1.4.19)$$

where equality holds for reversible processes. Next, we replace $TdS = d(TS) - SdT$ and bring the total differential on the other side,

$$d(U - TS) \leq -SdT + dW. \quad (1.4.20)$$

As a combination of state functions, $F = U - TS$ is a state function as well. It is called *free energy* and for an isothermal process ($dT = 0$) the negative change $-dF$ serves as an upper bound for the work performed by the system $-dW$,

$$-dW \leq -dF. \quad (1.4.21)$$

If the process is reversible, the work output is maximal and the bound saturated. Besides the practical implications, this inequality is remarkable because the fluctuation theorems by Jarzynski [80] and Crooks [81], we will encounter in Subsection 1.4.3, are stochastic generalizations of this inequality.

We conclude with the statement of the *third law of thermodynamics*. In a formulation known as the *unattainability principle*, it states that it is not possible to cool down a system to absolute zero in finite time. Besides the first and second law, it can be seen as another boundary for thermodynamically possible processes.

1.4.2. Microscopic point of view

As explained in Subsection 1.4.1, thermodynamics was originally purely macroscopic and phenomenological. However, in order to eventually understand thermodynamics in the quantum regime, a microscopic picture is required. To this end, we shortly outline

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how entropy was rediscovered as a statistical quantity also known in information theory and then present how thermodynamical systems are considered in statistical mechanics.

In 1877, Boltzmann laid the foundation for a statistically defined entropy [82]

$$S = k_B \log(\Omega), \quad (1.4.22)$$

where Ω is the number of accessible micro states. The famous formulation in (1.4.22), however, was published by Planck [83] in 1901. This formula already links the macroscopic quantity, entropy, to the statistics of microscopic quantities.

The influential work of Szilard in 1929 [84] proposed a machine that could beat the second law by exploiting additional information of the working medium. This made evident that information is relevant in thermodynamics. This discrepancy with the second law was resolved much later, in 1961 by Landauer who proved that the erasure of information is unavoidably accompanied with a flow of heat into the environment [85], ensuring the validity of the second law.

In 1948, Shannon established the field of *information theory* and the *Shannon entropy* as one of its main concepts [86]: Given a probability distribution $(p_n)_n$, the Shannon entropy is defined ¹⁰ as

$$\mathcal{H}[(p_n)_n] = - \sum_n p_n \log(p_n). \quad (1.4.23)$$

It is a measure of how uniform a distribution is: For a deterministic distribution $p_n = \delta_{nk}$, the entropy is zero and for the uniform distribution with N possible outcomes $p_n = 1/N$ the entropy reaches its maximal value of,

$$\mathcal{H}[(1/N)_n] = - \sum_{n=1}^N \frac{1}{N} \log\left(\frac{1}{N}\right) = - \log\left(\frac{1}{N}\right) = \log(N). \quad (1.4.24)$$

With the Shannon entropy at hand, we can now discuss a statistical approach to thermodynamics: We define the macroscopic state of a thermodynamic system to be a probability distribution over the microscopic states, e.g. the exact positions and momenta of the particles in a gas. A way to find such a probability distribution was proposed by Jaynes [87]. He suggested to treat the problem like a statistical inference problem by choosing the probability distribution which maximizes the Shannon entropy in accordance with all known constraints. Put differently, we model our knowledge by making the least assumption

Consider, for example, a system with Ω accessible states, completely isolated from any environment. Then, there are no constraints and the distribution with the maximal entropy is the uniform distribution with Shannon entropy $\mathcal{H}[(1/\Omega)_n] = \log(\Omega)$. Up to a constant, this explains the earlier result (1.4.22) of Boltzmann and Planck from a general point of view and motivates the identification of thermodynamic and Shannon entropy $S = k_B \mathcal{H}[(p_n)_n]$.

¹⁰Note that Shannon originally used the logarithm with base 2. We stick to the natural logarithm since it is customary in thermodynamics and does not change the qualitative results.

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A constraint that we will use frequently is a fixed average energy. Hence, we maximize the Shannon entropy over all probability distributions with a fixed value of,

$$U = \sum_n E_n p_n, \quad (1.4.25)$$

where E_n are the energies of the system. Using the method of Lagrange multipliers, we find a distribution with the probabilities

$$p_n = \exp(-\beta E_n) / Z, \quad (1.4.26)$$

where the Lagrange multiplier $\beta = 1/(k_B T)$ is known as the *inverse temperature* and the normalization constant Z is called *partition function*. The distribution $(p_n)_n$ is named after Gibbs¹¹, who first mentioned it in a formalized manner [88]. The ensemble corresponding to this distribution is called *canonical ensemble*.

Deriving the entropy of this probability distribution yields,

$$S = k_B \mathcal{H}[(p_n)_n] = k_B \sum_n p_n (\beta E_n + \log(Z)) = \frac{1}{T} U + k_B \log(Z), \quad (1.4.27)$$

where we identified the internal energy of the system as the mean energy. This can be rephrased as

$$-k_B T \log(Z) = U - TS = F, \quad (1.4.28)$$

making evident that it is sufficient to know the normalization constant Z in order to derive the free energy F and by that all thermodynamic quantities.

With the canonical framework at hand, we formulate the thermodynamic description of a classical and a quantum system. A classical system is characterized by a Hamiltonian function $H(\mathbf{x}, \mathbf{p})$ of the phase space variables $\mathbf{x}$ and $\mathbf{p}$, while a quantum system is characterized by a self-adjoint operator $\hat{H}$. For the sake of simplicity, we will refer to both $H(\mathbf{x}, \mathbf{p})$ and $\hat{H}$ as Hamiltonian. Classically, the ensemble state is a phase space probability density $P(\mathbf{x}, \mathbf{p})$, and the corresponding internal energy is given by the expectation value

$$U_{\text{cl}} = \int d^n \mathbf{x} d^n \mathbf{p} P(\mathbf{x}, \mathbf{p}) H(\mathbf{x}, \mathbf{p}). \quad (1.4.29)$$

On the other hand, the macroscopic state in the quantum case is a density matrix $\hat{\rho}$. The internal energy is given by the expectation value

$$U_{\text{q}} = \text{tr} \left\{ \hat{H} \hat{\rho} \right\}. \quad (1.4.30)$$

For a phase space density, the continuous version of the Shannon entropy has to be

¹¹Note that it is also called Boltzmann distribution due to Boltzmann's contributions to the foundations of the field.

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applied

$$\mathcal{H}[P(\mathbf{x}, \mathbf{p})_{\mathbf{x}, \mathbf{p}}] = - \int d^n \mathbf{x} d^n \mathbf{p} P(\mathbf{x}, \mathbf{p}) \log(P(\mathbf{x}, \mathbf{p})). \quad (1.4.31)$$

In quantum mechanics, the proper generalization of the Shannon entropy is the *von Neumann entropy*

$$\mathcal{S}[\hat{\rho}] = - \text{tr} \{ \hat{\rho} \log(\hat{\rho}) \}. \quad (1.4.32)$$

The connection between $\mathcal{S}$ and $\mathcal{H}$ becomes evident when one expresses $\hat{\rho}$ in its eigenbasis $(|n\rangle)_n$ with the corresponding populations $(p_n)_n$,

$$\mathcal{S}[\hat{\rho}] = \mathcal{H}[(p_n)_n]. \quad (1.4.33)$$

In both cases, classical and quantum, we can maximize the respective entropy with constraints and find the macroscopic states in the canonical ensemble at inverse temperature β ,

$$P(\mathbf{x}, \mathbf{p}) = e^{-\beta H(\mathbf{x}, \mathbf{p})} / Z_{\text{cl}} \quad (1.4.34)$$

$$\hat{\rho} = e^{-\beta \hat{H}} / Z_{\text{q}}. \quad (1.4.35)$$

Up to now, we treat classical and quantum systems on an equal footing. However, crucial differences will arise once we consider thermodynamic processes and attempt to characterize process quantities such as heat and work.

1.4.3. The different notions of (quantum) work

In our discussion up to now and in statistical mechanics in general [77], the systems are of macroscopic size. This allows us to neglect statistical fluctuations of macroscopic observables around their mean values, due to the law of large numbers. However, as the systems become smaller in size, the significance of random fluctuations increases. The thermodynamic consideration of these fluctuations gave rise to the classical field of *stochastic thermodynamics*, see the reviews [89, 90], with the *Fluctuation theorems* named after *Jarzynski* [80] and *Crooks* [81] at the heart of the field.

Jarzynski considers a system depending on a control parameter λ , initially in a Gibbs distribution at parameter λ_0 and temperature T . Then the parameter is driven, say $\lambda_0 \rightarrow \lambda_1$, while the system is isolated. At the new parameter λ_1 , the system thermalizes again and approaches a Gibbs distribution at temperature T . Note that the work W done on the system is a random variable because the trajectory that is taken by the system is random. However, this randomness solely originates from the random initial condition, drawn from the Gibbs distribution at λ_0 . The evolution itself is deterministic since the system is isolated. Hence, the random work can be identified as the entire change in system energy on a specific trajectory, labeled by an initial micro state. Jarzynski's achievement was to relate the statistics of Work to the change of free energy $\Delta F = F(T, \lambda_1) - F(T, \lambda_0)$,

$$\langle e^{-\beta W} \rangle = e^{-\beta \Delta F}. \quad (1.4.36)$$

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Note that the left hand side corresponds to the moment-generating function of the process quantity work, where $\langle \cdot \rangle$ denotes the ensemble average over the initial micro states. The right hand side only contains a difference of the state function F . The significance of this result becomes clearer by applying Jensen's inequality for expectation values of convex functions,

$$e^{-\beta \langle W \rangle} \leq \langle e^{-\beta W} \rangle. \quad (1.4.37)$$

This yields $-\langle W \rangle \leq -\Delta F$, which we found to be a consequence of the second law (1.4.21). In this sense, the Jarzynski equality can be seen as a stochastic refinement of the second law. Crooks' fluctuation theorem, on the other hand, is a refinement of the Jarzynski equation. The fluctuation theorems of Jarzynski and Crooks exemplify that a stochastic notion of work can be consistently included in classical thermodynamics.

However, in a quantum mechanical framework, the definition of quantum work is accompanied with new conceptual problems that led to various discussions [91–94], that still hold today [95–98]. The difficulty mainly arises from the conflict of two facts: On the one hand, work is a process quantity that has to be defined for the transitions between two states. On the other hand, quantum mechanical measurements are invasive in the sense that measurements change the state of the measured system. Thus, a determination of the underlying micro state at the beginning and the end of a process requires two measurements that will in general alter the evolution of the system. For the remainder of the subsection, we will discuss a selection of different notions of quantum work.

A definition of the average work in weakly coupled open quantum systems was already given in 1979 by Alicki [14]. For a system with a Hamiltonian $\hat{H}(\lambda)$, controlled by the parameter $\lambda(t)$, he applied the product rule to the internal energy $\hat{U} = \text{tr} \left\{ \hat{\rho} \hat{H}(\lambda) \right\}$,

$$\dot{\hat{U}} = \text{tr} \left\{ \dot{\hat{\rho}} \hat{H}(\lambda) \right\} + \text{tr} \left\{ \hat{\rho} \dot{\hat{H}}(\lambda) \right\}, \quad (1.4.38)$$

where the work corresponds to the energy change associated to a change in the control parameter $\lambda(t)$ and the heat corresponds to the change in the state $\hat{\rho}(t)$,

$$\dot{W} = \text{tr} \left\{ \hat{\rho} \dot{\hat{H}} \right\} = \text{tr} \left\{ \hat{\rho} \frac{\partial \hat{H}(\lambda)}{\partial \lambda} \right\} \dot{\lambda} \quad (1.4.39)$$

$$\dot{Q} = \text{tr} \left\{ \dot{\hat{\rho}} \hat{H}(\lambda) \right\}. \quad (1.4.40)$$

We employ this notion of work in Chapter 3.

Surprisingly, even a straightforward definition of mean work as in (1.4.39) can become problematic for quantum systems: There, a change in λ is often associated with a change in the energy of some control field, which might not be useful for any task. In fact, a dynamical exchange of work would require an interaction Hamiltonian, coupling system and load. This notion of work, however, is then dependent on the type of interaction and the pursued task. While this is, in principle, a problem in classical physics as well, it is an empirically fact that a classical control parameter, e.g. the motion of a piston, is

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capable of doing useful tasks. This is because, unlike in the quantum case, the “entropy-associated energy” of the control field (or piston) can be neglected, as discussed in [99]. Nevertheless, we may still take a general standpoint and seek quantitative statements or at least bounds on how much work one could in principle extract from a system in a given process. For a more general review discussing several definitions of work, see [100].

The *operator of work* $\hat{W}$ is one of the oldest concepts of quantum work [101–103] and in [94] Pinto Silva and Gelbwaser-Klimovsky make the case for this definition of work. Consider a thermally isolated system with a time-dependent Hamiltonian $\hat{H}(\lambda(t))$, unitarily evolving from $t_0 \rightarrow t_1$ with the time evolution operator $\hat{U}$. Due to the thermal isolation, the heat flow $\dot{Q} = 0$ vanishes and the integration of the mean work (1.4.39) has to be the change in internal energy. Making use of the linearity and cyclic invariance of the trace, we find the mean work,

$$\begin{aligned} W_{0 \rightarrow 1} &= \text{tr} \left\{ \hat{\rho}(t_1) \hat{H}(\lambda(t_1)) \right\} - \text{tr} \left\{ \hat{\rho}(t_0) \hat{H}(\lambda(t_0)) \right\} \\ &= \text{tr} \left\{ \hat{U} \hat{\rho}(t_0) \hat{U}^\dagger \hat{H}(\lambda(t_1)) \right\} - \text{tr} \left\{ \hat{\rho}(t_0) \hat{H}(\lambda(t_0)) \right\} \\ &= \text{tr} \left\{ \left(\hat{U}^\dagger \hat{H}(\lambda(t_1)) \hat{U} - \hat{H}(\lambda(t_0)) \right) \hat{\rho}(t_0) \right\}. \end{aligned} \quad (1.4.41)$$

Hence the operator of work for this process, defined as

$$\hat{W}_{0 \rightarrow 1} = \hat{U}^\dagger \hat{H}(\lambda(t_1)) \hat{U} - \hat{H}(\lambda(t_0)), \quad (1.4.42)$$

is by construction consistent with (1.4.39). Besides calculating the mean, we can use this self-adjoint operator to calculate higher moments of work $\langle \hat{W}_{0 \rightarrow 1}^k \rangle$ or the full work statistics $P(w_n) = \langle w_n | \hat{\rho}(t_0) | w_n \rangle$, where $|w_n\rangle$ are eigenvectors of $\hat{W}_{0 \rightarrow 1}$. We use this notion of quantum work to describe the fluctuations in the quantum heat engines in Chapter 2.

Another notion related to work, often used in the context of quantum batteries [104], is the *ergotropy* $\mathcal{E}$, which is used in Chapter 3 [105, 106]. For a given state $\hat{\rho}$ and Hamiltonian $\hat{H}$, it is defined as the maximal energy that can be extracted unitarily,

$$\mathcal{E} [\hat{\rho}, \hat{H}] = \text{tr} \left\{ \hat{\rho} \hat{H} \right\} - \min_{\hat{U}} \left\{ \text{tr} \left\{ \hat{U} \hat{\rho} \hat{U}^\dagger \hat{H} \right\} \right\}. \quad (1.4.43)$$

The minimization in (1.4.43) is explicitly solved in [105]. For this, it is convenient to arrange the eigenvalues of $\hat{\rho}$ in descending and the eigenvalues of $\hat{H}$ in ascending order,

$$\hat{\rho} = \sum_{n=1}^N P_n |P_n\rangle \langle P_n|, \quad P_1 \geq P_2 \geq \dots \geq P_N \quad (1.4.44)$$

$$\hat{H} = \sum_{k=1}^N E_k |E_k\rangle \langle E_k|, \quad E_1 \leq E_2 \leq \dots \leq E_N. \quad (1.4.45)$$

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Then the optimal unitary minimizing the second term of (1.4.43) can be written as

$$\hat{U}_{\min} = \sum_k |E_k\rangle \langle P_k|. \quad (1.4.46)$$

Based on this solution, the ergotropy can be given as,

$$\begin{aligned} \mathcal{E} [\hat{\rho}, \hat{H}] &= \text{tr} \left\{ \hat{\rho} \hat{H} \right\} - \text{tr} \left\{ \hat{U}_{\min} \hat{\rho} \hat{U}_{\min}^\dagger \hat{H} \right\} \\ &= \text{tr} \left\{ \hat{\rho} \hat{H} \right\} - \sum_k P_k E_k. \end{aligned} \quad (1.4.47)$$

1.5. Metrology

Metrology is the scientific field of measurements that is, for example, concerned with the achievable precision in sensing. In this section, we discuss two different metrological tasks. On the one hand, parameter estimation, where the task is to infer the value of an unknown parameter from observed outcome statistics. On the other hand, state discrimination, where the task is to distinguish between two probability distributions or quantum states. In both cases, we first apply the classical theory to the probability distributions of the quantum measurement statistics for a certain measurement. Then we proceed by introducing the quantum analogue by optimizing over the quantum measurement.

For parameter estimation, we introduce the notion of *Fisher information* that characterizes the sensitivity with respect to the unknown parameter. The distance between two probability distributions, on the other hand, can be expressed by the *trace distance* that we use for state discrimination.

The Fisher information and trace distance can both be applied to the quantum case by the same procedure: We first note that the classical notion can be applied to the outcome statistics after performing a POVM measurement. Then, in order to define a measurement-independent object, the *quantum Fisher information* and (*quantum*) *trace distance* are defined as the maximization over all possible POVMs.

Parameter estimation in the classical and quantum context are discussed in Subsections 1.5.1 and 1.5.2. A criterion for when classical and quantum Fisher information are the same is presented in Subsection 1.5.3. Finally, the classical and quantum trace distance are shown in Subsections 1.5.4 and 1.5.5.

1.5.1. Classical parameter estimation

In the field of parameter estimation, we are challenged with the following task: Consider a system that, depending on an unknown parameter ϑ , creates a random output ξ . Assume we know the *likelihood function* $P(\xi|\vartheta)$, which is the parameter-dependent probability density of the observation, from our physical model. Based on an observation ξ , we want to estimate the parameter ϑ . To be more precise, we want to find a function

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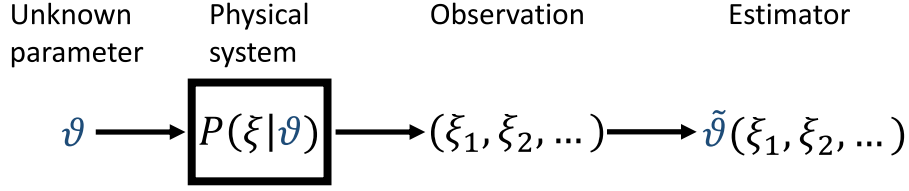


Figure 1.3.: Flow chart of a classical parameter estimation problem: An unknown parameter ϑ influences a physical system. The random and parameter dependent behavior of the system is encoded in the known likelihood function $P(\xi|\vartheta)$. Based on random observations from the system, an estimator $\tilde{\vartheta}(\xi_1, \xi_2, \dots)$ for the actual parameter ϑ is calculated.

$\tilde{\vartheta}(\xi)$, called *estimator*, that minimizes the error $|\tilde{\vartheta}(\xi) - \vartheta|$ in some sense, specified later. The whole setting is depicted in Fig. 1.3.

Given a chosen estimator, we define the average estimate,

$$\mathbb{E}_{\vartheta}[\tilde{\vartheta}] = \int d\xi \tilde{\vartheta}(\xi) P(\xi|\vartheta). \quad (1.5.1)$$

To quantify the error of the estimate, we consider its deviation from the true value of the parameter and split it into two parts,

$$\tilde{\vartheta}(\xi) - \vartheta = \left(\tilde{\vartheta}(\xi) - \mathbb{E}_{\vartheta}[\tilde{\vartheta}] \right) + \left(\mathbb{E}_{\vartheta}[\tilde{\vartheta}] - \vartheta \right). \quad (1.5.2)$$

The first part describes the deviation of the estimator from its mean, while the second part describes the deviation from the mean estimate to the true value of the parameter ϑ . If we assume our estimator to be *unbiased*, meaning that $\mathbb{E}_{\vartheta}[\tilde{\vartheta}] = \vartheta$, the second term in (1.5.2) vanishes. Hence, the error of the estimator is characterized by its deviation from the mean. The standard measure for such a deviation is the variance,

$$\mathbb{V}_{\vartheta}[\tilde{\vartheta}] = \int d\xi \left(\tilde{\vartheta}(\xi) - \vartheta \right)^2 P(\xi|\vartheta). \quad (1.5.3)$$

The variance can be bounded from below for any (unbiased) estimator in terms of the so-called Fisher information(FI) [107],

$$\mathcal{I}(\vartheta) = \int d\xi \frac{(\partial P(\xi|\vartheta))^2}{P(\xi|\vartheta)}, \quad (1.5.4)$$

where ∂ denotes the derivative with respect to ϑ . The FI can be seen as the sensitivity of the outcome with respect to a small change of the parameter, and appears in the *Cramér–Rao bound* [108, 109],

$$\mathbb{V}_{\vartheta}[\tilde{\vartheta}] \geq \frac{1}{\mathcal{I}(\vartheta)}. \quad (1.5.5)$$

This shows that the error of any unbiased estimator can be bounded by a function of

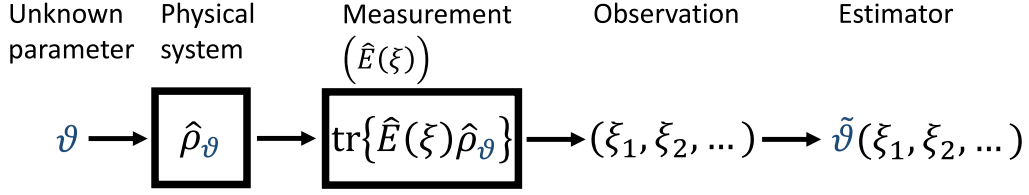


Figure 1.4.: Flow chart of a quantum parameter estimation problem: An unknown parameter ϑ is encoded in the system state $\hat{\rho}_\vartheta$. If the POVM $\hat{E}(\xi)$ is measured on the system, each observation is sampled according to the likelihood $P(\xi|\vartheta) = \text{tr}\{\hat{E}(\xi)\hat{\rho}_\vartheta\}$. Based on the observations $\xi_1, \xi_2, \dots$, the value of the parameter is estimated by $\tilde{\vartheta}(\xi_1, \xi_2, \dots)$

the likelihood function $P(\xi | \vartheta)$, without specifying the estimator any further. In [110], Fisher himself proved that a specific estimator, called *maximum likelihood estimator*, reaches the bound asymptotically. Fortunately, we can improve the Cramér–Rao bound by repeating the experiment: Consider N observations $\xi_1, \xi_2, \dots, \xi_N$, independently drawn from the likelihood function $P(\xi | \vartheta)$. Then, the joint likelihood function of the N observations becomes the product of the individual likelihood functions,

$$P_N(\xi_1, \xi_2, \dots, \xi_N | \vartheta) = \prod_{j=1}^N P(\xi_j | \vartheta). \quad (1.5.6)$$

A straightforward calculation shows that the FI of $P_N(\xi_1, \xi_2, \dots, \xi_N | \vartheta)$, $\mathcal{I}_N(\vartheta)$, is N times the single-observation FI,

$$\mathcal{I}_N(\vartheta) = N\mathcal{I}(\vartheta). \quad (1.5.7)$$

Hence, for unbiased estimators based on N independent observations, the *Cramér–Rao bound* scales inversely proportional to N ,

$$\mathbb{V}_\vartheta[\tilde{\vartheta}] \geq \frac{1}{N\mathcal{I}(\vartheta)}, \quad (1.5.8)$$

which corresponds to the well-known shot-noise limit for the counting of N independent ‘clicks’ in a detector. For this reason, the FI is often interpreted as the amount of information about the parameter ϑ that is carried by a single observation.

Up to now, we have assumed a fixed, and not yet specified, measurement procedure underlying the likelihood function. In the following, we will consider quantum measurements and allow them to vary.

1.5.2. Quantum parameter estimation

The results of classical estimation theory, explained in Subsection 1.5.1, can be directly applied to the outcome statistics of a quantum measurement. However in quantum mechanics, we have the additional freedom of choosing the measurement basis, see Fig. 1.3.

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The setting is now as follows: The quantum system is prepared in some parameter-dependent state $\hat{\rho}_\vartheta$, and we choose to interrogate it by means of a POVM $\left(\hat{E}(\xi)\right)$. The likelihood of obtaining an outcome ξ is then

$$P(\xi|\vartheta) = \text{tr} \left\{ \hat{E}(\xi) \hat{\rho}_\vartheta \right\}. \quad (1.5.9)$$

We could now evaluate the corresponding FI (1.5.4) and obtain the Cramér–Rao bound (1.5.5), which depends strongly on the chosen measurement. For a more fundamental bound, we can maximize the Fisher information over the measurements to obtain the so-called *quantum Fisher information* (QFI) [111],

$$\mathcal{J}(\vartheta) = \max_{(\hat{E})} \mathcal{I}(\vartheta). \quad (1.5.10)$$

Per definition, the Cramér–Rao bound still holds for this quantity,

$$\mathbb{V}_\vartheta[\tilde{\vartheta}] \geq \frac{1}{N\mathcal{J}(\vartheta)}. \quad (1.5.11)$$

Fortunately, there is a way to circumvent the maximization (1.5.10) and to calculate the QFI directly from $\hat{\rho}_\vartheta$ [112]: In order to do so, one needs to find the *symmetric logarithmic derivative* (SLD) $\hat{L}_\vartheta$, implicitly defined through a Lyapunov equation,

$$\partial \hat{\rho}_\vartheta = \frac{1}{2} \left\{ \hat{L}_\vartheta, \hat{\rho}_\vartheta \right\}, \quad (1.5.12)$$

where we introduced the abbreviation $d/d\vartheta = \partial$. The QFI is then simply given by,

$$\mathcal{J}(\vartheta) = \text{tr} \left\{ \hat{\rho}_\vartheta \hat{L}_\vartheta^2 \right\}. \quad (1.5.13)$$

In general, finding the SLD is a difficult numerical task. In this thesis however, we will be mostly concerned with the QFI for pure states $\hat{\rho}_\vartheta = |\psi_\vartheta\rangle \langle \psi_\vartheta|$, for which a simple explicit expression can be derived: We recall that pure states fulfill $\hat{\rho}_\vartheta^2 = \hat{\rho}_\vartheta$ and apply the product rule,

$$\partial \hat{\rho}_\vartheta = \partial \hat{\rho}_\vartheta^2 = \hat{\rho}_\vartheta \partial \hat{\rho}_\vartheta + (\partial \hat{\rho}_\vartheta) \hat{\rho}_\vartheta = \frac{1}{2} \{2\partial \hat{\rho}_\vartheta, \hat{\rho}_\vartheta\}. \quad (1.5.14)$$

By comparison with (1.5.12), we find the SLD,

$$\hat{L}_\vartheta = 2\partial \hat{\rho}_\vartheta = 2(|\partial \psi_\vartheta\rangle \langle \psi_\vartheta| + |\psi_\vartheta\rangle \langle \partial \psi_\vartheta|). \quad (1.5.15)$$

From the SLD, the QFI is found in a straightforward calculation,

$$\mathcal{J}(\vartheta) = 4 \left(\langle \partial \psi_\vartheta | \partial \psi_\vartheta \rangle - |\langle \psi_\vartheta | \partial \psi_\vartheta \rangle|^2 \right). \quad (1.5.16)$$

Here, we define $|\partial \psi_\vartheta\rangle = \frac{d}{d\vartheta} |\psi_\vartheta\rangle$.

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For comparison, the Fisher information for a given projective measurement basis $(|\xi\rangle)$ with likelihood $P(\xi|\vartheta) = |\langle\xi|\psi_\vartheta\rangle|^2$, reads as

$$\mathcal{I}(\vartheta) = 4 \int d\xi \frac{\text{Re} \{ \langle\psi_\vartheta|\xi\rangle \langle\xi|\partial\psi_\vartheta\rangle \}^2}{|\langle\xi|\psi_\vartheta\rangle|^2}. \quad (1.5.17)$$

We conclude with a specific scenario, called *phase estimation*. Here, the evolution of the pure state is generated by a self-adjoint operator $\hat{G}$,

$$|\psi_\vartheta\rangle = e^{-i\vartheta\hat{G}} |\psi_0\rangle. \quad (1.5.18)$$

In this case, the SLD (1.5.15) is found to be the commutator

$$\hat{L}_\vartheta = 2i \left[\hat{G}, |\psi_\vartheta\rangle \langle\psi_\vartheta| \right] \quad (1.5.19)$$

and (1.5.16) simplifies to the ϑ -independent formula,

$$\mathcal{J} = 4 \left(\langle\psi_0| \hat{G}^2 |\psi_0\rangle - (\langle\psi_0| \hat{G} |\psi_0\rangle)^2 \right) = 4\mathbb{V} \left[\hat{G} \right]_{\psi_0}, \quad (1.5.20)$$

with the variance of the operator, $\mathbb{V}[\hat{G}]_{\psi_0}$.

In Chapter 4 we will compare the FI in several bases with the QFI in order to find the optimal measurement basis for a scattering scenario in electron microscopes.

1.5.3. Optimality criterion

By definition, the optimal sensing precision is achieved in a basis, in which the QFI saturates the CFI. A criterion for this optimality of a given basis can be found in [113]. For better understanding, we show this insightful derivation, using the present notation: Say, we want to measure our system in the continuous basis $(|\xi\rangle)_\xi$. Then the system state $|\psi_\vartheta\rangle$ can be expanded in this basis as

$$|\psi_\vartheta\rangle = \int d\xi \psi(\xi|\vartheta) |\xi\rangle = \int d\xi \sqrt{p(\xi|\vartheta)} e^{i\varphi(\xi|\vartheta)} |\xi\rangle, \quad (1.5.21)$$

making use of the polar decomposition of the complex numbers $\psi(\xi|\vartheta)$. Notice that $p(\xi|\vartheta)$ is already the likelihood function that enters the FI for this measurement. With expression (1.5.21) at hand, we can proceed to calculate the QFI (1.5.16). Taking the derivative of the state with respect to the parameter, we find,

$$|\partial\psi_\vartheta\rangle = \int d\xi \left(\frac{\partial p(\xi|\vartheta)}{2p(\xi|\vartheta)} + i\partial\varphi(\xi|\vartheta) \right) \psi(\xi|\vartheta) |\xi\rangle. \quad (1.5.22)$$

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We further calculate,

$$\begin{aligned}
\langle \psi_\vartheta | \partial \psi_\vartheta \rangle &= \int d\xi \left(\frac{\partial p(\xi|\vartheta)}{2p(\xi|\vartheta)} + i\partial\varphi(\xi|\vartheta) \right) p(\xi|\vartheta) \\
&= \frac{1}{2} \frac{\partial}{\partial\vartheta} \int d\xi p(\xi|\vartheta) + i \int d\xi p(\xi|\vartheta) \partial\varphi(\xi|\vartheta) \\
&= i\mathbb{E}_\vartheta [\partial\varphi],
\end{aligned} \tag{1.5.23}$$

where the first term vanishes after exchanging derivative and integral and the second term can be rephrased as the expectation value of the derivative of the phase. We also find,

$$\begin{aligned}
\langle \partial \psi_\vartheta | \partial \psi_\vartheta \rangle &= \int d\xi \left(\frac{\partial p(\xi|\vartheta)}{2p(\xi|\vartheta)} - i\partial\varphi(\xi|\vartheta) \right) \left(\frac{\partial p(\xi|\vartheta)}{2p(\xi|\vartheta)} + i\partial\varphi(\xi|\vartheta) \right) p(\xi|\vartheta) \\
&= \frac{1}{4} \int d\xi \frac{(\partial p(\xi|\vartheta))^2}{p(\xi|\vartheta)} + \int d\xi p(\xi|\vartheta) (\partial\varphi(\xi|\vartheta))^2 \\
&= \frac{1}{4} \mathcal{I}(\vartheta) + \mathbb{E}_\vartheta [(\partial\varphi)^2],
\end{aligned} \tag{1.5.24}$$

which splits into the FI for the likelihood $p(\xi|\vartheta)$ and the second moment of the derivative of the phases. Altogether, we find a connection between quantum and classical Fisher information,

$$\mathcal{J}(\vartheta) = \mathcal{I}(\vartheta) + 4\mathbb{V}_\vartheta [\partial\varphi]. \tag{1.5.25}$$

Note that the left hand side of (1.5.25) does not depend on the measurement basis, while both contributions on the right hand side do. In other words, for any basis, the QFI decomposes into a part encoded in the amplitudes (the CFI) and a part encoded in the phases. Since the measurement is agnostic to the phase information, we want the phase term to be minimized. In the basis where it vanishes, the CFI consequently saturates the QFI.

By noting $e^{2i\varphi(\xi|\vartheta)} = \psi(\xi|\vartheta)/\psi(\xi|\vartheta)^*$, one can directly derive,

$$\partial\varphi(\xi|\vartheta) = \frac{\text{Im} \{ \psi(\xi|\vartheta)^* \partial \psi(\xi|\vartheta) \}}{|\psi(\xi|\vartheta)|^2} = \frac{\text{Im} \{ \langle \psi_\vartheta | \xi \rangle \langle \xi | \partial \psi_\vartheta \rangle \}}{|\langle \xi | \psi_\vartheta \rangle|^2}. \tag{1.5.26}$$

The variance of a random variable is zero if and only if it is constant. Here, this is equivalent to $\partial\varphi$ being independent of ξ , i.e. $\partial\varphi(\xi|\vartheta) = c(\vartheta)$. We can determine this constant by taking the expectation value on both sides,

$$c(\vartheta) = \mathbb{E}_\vartheta [\partial\varphi] = \text{Im} \{ \langle \psi_\vartheta | \partial \psi_\vartheta \rangle \}, \tag{1.5.27}$$

where the second equality is found in (1.5.26). This results in a useful optimality criterion: If and only if

$$\text{Im} \{ \langle \psi_\vartheta | \xi \rangle \langle \xi | \partial \psi_\vartheta \rangle \} = |\langle \xi | \psi_\vartheta \rangle|^2 \text{Im} \{ \langle \psi_\vartheta | \partial \psi_\vartheta \rangle \} \tag{1.5.28}$$

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is fulfilled, the measurement in the $(|\xi\rangle)$ -basis saturates the QFI.

An equivalent, more compact version, e.g. used in [11], can be directly derived by using the $\mathbb{R}$ -linearity of the imaginary part,

$$\langle \psi_\vartheta | \xi \rangle \langle \xi | \hat{Q} | \partial \psi_\vartheta \rangle \in \mathbb{R}, \quad (1.5.29)$$

where $\hat{Q} = \mathbb{1} - |\psi_\vartheta\rangle \langle \psi_\vartheta|$ is the projector onto the orthogonal subspace of $|\psi_\vartheta\rangle$.

We conclude with the remark that $\langle \xi | \psi_\vartheta \rangle$ and $\langle \xi | \partial \psi_\vartheta \rangle \in \mathbb{R}$ for all ξ trivially implies (1.5.28) and thus constitutes a sufficient condition for optimality.

1.5.4. Classical state discrimination

Consider an experiment that can run in two different configurations, for example a coin that is either biased or not. If the system is in configuration A , our observations ξ are distributed with respect to the probability density $P_A(\xi)$. In the other configuration B , outcomes are produced with respect to $P_B(\xi)$. Provided the experiment is with equal probability in any of the two configurations, how well can we infer the configuration from a single observation ξ ?

Unless the distributions have non-overlapping support, this question cannot be answered with certainty. However, we can maximize the success probability, P_{suc} . To this end, we fix a set X and announce configuration A , if $\xi \in X$. On the other hand, we announce configuration B , if $\xi \in X^c$. For this strategy, we can derive the success probability,

$$P_{\text{suc}}(X) = \frac{1}{2} \int_X d\xi P_A(\xi) + \frac{1}{2} \int_{X^c} d\xi P_B(\xi) = \frac{1}{2} \left(1 + \int_X d\xi [P_A(\xi) - P_B(\xi)] \right), \quad (1.5.30)$$

where we used the rule for complementary probabilities in the second step. We define the *total variation distance*, L_1 -distance, or just *classical trace distance* as the second term of the success probability maximized over the set X ,

$$D_C(P_A, P_B) = \sup_X \left| \int_X d\xi [P_A(\xi) - P_B(\xi)] \right|. \quad (1.5.31)$$

Fortunately, this optimization is explicitly solved by the set $X = \{\xi | P(\xi) \geq Q(\xi)\}$. Put differently, given ξ , the best strategy is to announce the configuration for which the probability to observe ξ is higher. From this optimal set, one can also see the alternative explicit definition

$$D_C(P_A, P_B) = \frac{1}{2} \int d\xi |P_A(\xi) - P_B(\xi)|. \quad (1.5.32)$$

Using (1.5.31) in the definition of the success probability (1.5.30), yields the optimal success probability,

$$P_{\text{suc}} = \frac{1}{2} (1 + D_C(P_A, P_B)), \quad (1.5.33)$$

which gives an operational meaning to the trace distance. A detailed discussion of the matter can be found in [70].

1.5.5. Quantum state discrimination

In quantum state discrimination, we obtain one of the states $\hat{\rho}$, $\hat{\sigma}$ with equal probability and try to infer which one we obtained. In case we are constrained to measure a specific POVM, say $(\hat{E}(\xi))$, the outcomes are distributed by either $P(\xi) = \text{tr} \left\{ \hat{E}(\xi) \hat{\rho} \right\}$ or $Q(\xi) = \text{tr} \left\{ \hat{E}(\xi) \hat{\sigma} \right\}$. Then, we can apply the classical theory from Subsection 1.5.4.

In fact, one can define the (quantum) trace distance as the classical trace distance maximized over all POVMs,

$$D_Q(\hat{\rho}, \hat{\sigma}) = \sup_{(\hat{E})} D_C(P, Q). \quad (1.5.34)$$

Due to the dichotomic nature of the problem, it is sufficient to consider the POVM $(\hat{F}, \mathbb{1} - \hat{F})$ and optimize over $\hat{F}$,

$$D_Q(\hat{\rho}, \hat{\sigma}) = \sup_{\mathbb{1} \geq \hat{F} \geq 0} \text{tr} \left\{ \hat{F} (\hat{\rho} - \hat{\sigma}) \right\}. \quad (1.5.35)$$

Analogous to the classical case, the optimal measurement $\hat{F}$ is the projector onto the positive eigenspace of $\hat{\rho} - \hat{\sigma}$. This yields the explicit formula,

$$D_Q(\hat{\rho}, \hat{\sigma}) = \frac{1}{2} \text{tr} \left\{ |\hat{\rho} - \hat{\sigma}| \right\}. \quad (1.5.36)$$

For pure states, $|\psi\rangle$ and $|\phi\rangle$, the formula simplifies,

$$D_Q(|\psi\rangle, |\phi\rangle) = \sqrt{1 - |\langle\psi|\phi\rangle|^2}. \quad (1.5.37)$$

The equivalence of all these definitions can be found in [70].

We conclude with the remark that the operational meaning as a notion of success probability is still valid in the quantum case,

$$p_{\text{suc}} = \frac{1}{2} (1 + D_Q(\hat{\rho}, \hat{\sigma})). \quad (1.5.38)$$

2. Otto cycles with a quantum planar rotor

This Chapter is based on the publication [A].

2.1. Introduction

In the field of quantum thermodynamics, discussed in Section 1.4, the study of quantum analogues of classical thermal machine models such as the Carnot cycle [20–24] and the Otto cycle [25–31] is one of the main concerns. Instead of a classical macroscopic working medium such as a gas, these machines employ quantum systems of varying complexity. These systems undergo a controlled sequence of strokes that alternate between thermal contact with hot and cold reservoirs and time-dependent modulation of the system’s Hamiltonian, effectively mimicking the (model-extrinsic) motion of a “piston.” Here, we focus on the Otto cycle, which admits two useful modes of operation: An engine mode that outputs work at the expense of heat from the hot reservoir and a refrigerator that extracts heat from the cold reservoir at the expense of work.

The quantum systems acting as working media range from small finite-dimensional systems [38, 114, 115] to many-body systems [116–118] and infinite-dimensional continuous-variable systems [29, 119, 120]. Experimental proof-of-principle realizations of the Otto cycle were performed with trapped ions [33, 34], nano beams [37], nitrogen vacancy centers [36], spins with nuclear magnetic resonance techniques [38], optomechanical systems [41], quantum gases [43] and proposed for nanomechanical resonators [121], circuit QED [122], and quantum dots [123].

Continuous-variable working media often represent motional degrees of freedom with a classical analogue, such as the position and momentum of a particle or, in our case, the orientation and angular momentum of a rigid body. These working media are suitable to study quantum effects on the machine performance by admitting a direct comparison between the classical and the quantum version of the studied machine. In contrast, many authors consider the quantum-classical comparison to be a comparison of machine models with and without coherence [124, 125]. While the advantages of quantum machines are often highlighted in specific cases [126–130], the performance of a machine model can in general both improve and deteriorate due to quantization of the working medium.

The harmonic oscillator is, due its mathematical well understood behavior, an exhaustively studied working medium. Surprisingly, it can be shown that for an Otto cycle with a harmonic oscillator working medium, the parameter regimes correspond-

ing to different operation modes (e.g., engine and refrigerator) are identical in both the classical and quantum cases. To be more precise, a homogeneous scaling of the energy levels with respect to the work parameter λ that represents the periodic piston modulation of the harmonic frequency, $E_n(\lambda) = \lambda \hbar \omega (n + 1/2)$, implies classical operation modes [131]. Furthermore, this holds true for any Otto cycle, where the energy spectrum scales proportionally with the work parameter, $E_n(\lambda) \propto \lambda^k$.

In this work, we study the quantum planar rotor as a mechanical working medium for the quantum Otto cycle. This complements previous theoretical works employing the rotor as an autonomous piston degree of freedom for engines [132–136]. The quantization of angular momentum is known to lead to genuinely non-classical phenomena in the free evolution of a single planar rotor [137] as well as in the dynamics of coherently interlocked rotors [138]. Rotor-based machines could be demonstrated experimentally on the basis of molecular rotors, whose dynamics are nowadays routinely observed and controlled with help of tailored laser pulses [139, 140]. A platform that could realize rotor-like dynamics is circuit QED [141], where the Josephson phase corresponds to the rotor angle. Finally, state-of-the-art experiments in levitated optomechanics with rigid nanorotors are steadily approaching the quantum regime [142–146].

The theoretical model of a planar rotor subjected to an externally modulated potential in the four-stroke Otto cycle is formulated in Section 2.2. Classical and quantum predictions are then examined for two physically motivated examples, illustrated in Fig. 2.2 and Fig. 2.6. The first example, presented in Section 2.3, considers a rotating electric dipole subject to an electric field of varying strength in the plane of rotation. Assuming ideal quenches of the electric field and full thermalization in between, we show that the so defined Otto cycle always performs worse in the quantum case. In the second example that we present in Section 2.4, we consider a magnetic dipole subject to a magnetic field of varying strength perpendicular to the rotation plane. Here, we show that the classical machine does not allow for an engine nor for a refrigerator operation mode. The quantum machine, on the other hand, shows both operation modes, when the cold stroke of the Otto cycle operates in the low-excitation regime. Hence, this rotor-based machine has a genuine advantage when run with a quantum planar rotor. A conclusion of our findings is presented in Section 2.5.

2.2. Theoretical model

A planar rotor is a dynamical degree of freedom described by a single angular coordinate $\alpha \in [0, 2\pi)$ and its conjugate angular momentum L_z . Physically, it represents, for example, a particle on a ring in the xy -plane, the phase variable in a superconducting Josephson loop, and the orientation of a rigid rotor on a fixed plane of rotation. We will first introduce the notation and theoretical description of a classical and quantum planar rotor, before briefly reviewing the ideal four-stroke Otto cycle.

2.2.1. Classical and quantum planar rotor

In the classical case, the canonical variables (α, L_z) of the planar rotor behave analogous to the position and momentum of linear motion in one dimension. Physical systems are characterized by Hamilton (functions) $H(\alpha, L_z)$ that obey a strict 2π -periodicity in α and ensemble states are described by phase-space probability densities $P(\alpha, L_z)$. The Hamiltonians, we will consider in this work, are of the general form,

$$H = H(\alpha, L_z) = \frac{(L_z - I\omega)^2}{2I} + V(\alpha), \quad (2.2.1)$$

where I is the moment of inertia, $V(\alpha) = V(\alpha + 2\pi)$ is a periodic potential, and $I\omega$ is an angular momentum displacement. The latter can be viewed as a boost with respect to a rotating frame at angular frequency ω .

In thermal equilibrium at temperature T , we assume such a planar rotor to be in a Gibbs state (see Subsection 1.4.2),

$$P(\alpha, L_z) = \frac{1}{Z} e^{-H(\alpha, L_z)/k_B T}, \quad (2.2.2)$$

with the partition function,

$$\begin{aligned} Z &= \int_0^{2\pi} d\alpha \int_{-\infty}^{\infty} dL_z e^{-H(\alpha, L_z)/k_B T} \\ &= \sqrt{2\pi I k_B T} \int_0^{2\pi} d\alpha e^{-V(\alpha)/k_B T}. \end{aligned} \quad (2.2.3)$$

Quantum mechanically, linear and rotational motion exhibit fundamental differences. In the case of a quantum rotor, the state components in the rotational basis, $\psi(\alpha) = \langle \alpha | \psi \rangle$, form a 2π -periodic wavefunction. This periodicity implies the quantization of angular momentum in integer multiples of $\hbar$. Thus, the angular momentum operator can be expressed in the orthonormal basis of angular momentum eigenstates $(|m\rangle)_{m \in \mathbb{Z}}$,

$$\hat{L}_z = \sum_{m \in \mathbb{Z}} \hbar m |m\rangle \langle m|. \quad (2.2.4)$$

The components in the orientational and the angular momentum basis are connected via the Fourier transform,

$$\langle \alpha | m \rangle = e^{im\alpha} / \sqrt{2\pi}. \quad (2.2.5)$$

In order to ensure the preservation of wavefunction periodicity, we define the conjugate angle operator by the unitary momentum displacement operators $\exp(ik\hat{\alpha})$, which obey periodicity for $k \in \mathbb{Z}$ and act like angular momentum kicks in the angular momentum basis,

$$\exp(ik\hat{\alpha}) |m\rangle = |m + k\rangle. \quad (2.2.6)$$

2. Otto cycles with a quantum planar rotor

The basis of orientation states $|\alpha\rangle$ is consistently obtained as,

$$|\alpha\rangle = \sum_{m \in \mathbb{Z}} \frac{e^{-im\alpha}}{\sqrt{2\pi}} |m\rangle. \quad (2.2.7)$$

They form an improper continuous basis with the orthogonality property $\langle\alpha_1|\alpha_2\rangle = \delta(\alpha_1 - \alpha_2 \bmod 2\pi)$. As eigenstates of the displacement operators they are automatically eigenstates of any periodic function of $\hat{\alpha}$ by virtue of the Fourier transform; e.g., $V(\hat{\alpha})|\alpha\rangle = V(\alpha)|\alpha\rangle$.

The commutation relation between $\hat{L}_z$ and any periodic function $f(\hat{\alpha})$ can be expressed as,

$$[f(\hat{\alpha}), \hat{L}_z] = i\hbar f'(\hat{\alpha}), \quad \forall \text{ } 2\pi\text{-periodic } f(\alpha). \quad (2.2.8)$$

This constitutes the canonical commutation relation (1.2.1) for the planar rotor. In the quantum case, the generic Hamiltonian (2.2.1) will have a discrete spectrum of energy eigenvalues E_n . The thermal state is then represented by the density matrix,

$$\hat{\gamma} = \exp(-\hat{H}/k_B T)/Z, \quad (2.2.9)$$

with the partition function,

$$Z = \text{tr}\{\exp(-\hat{H}/k_B T)\}. \quad (2.2.10)$$

For convenience in the following analytical and numerical calculations, we introduce a natural energy scale,

$$E = \frac{\hbar^2}{I}, \quad (2.2.11)$$

which corresponds to the energy of the lowest excited rotor state. In the comparison of classical and quantum rotors, we only expect notable differences for thermal energies that do not exceed this scale too far.

2.2.2. The Otto cycle

The Otto cycle, which can be formulated in both classical and quantum settings, is among the most widely studied and instructive models of thermal machines [29, 147]. We proceed with the description of the quantum version and introduce the classical counterpart later. The cycle consists of a quantum system with a control parameter-dependent Hamiltonian $\hat{H}(\lambda)$ operating between one hot and one cold heat bath with respective temperatures $T_h > T_c$. The parameter, an abstraction from the piston motion in a classical machine, varies between the extreme values λ_h and λ_c . The temperature T and the control parameter λ comprise the independent thermodynamic state variable of the system.

The idealized Otto cycle, sketched in the phase diagram spanned by λ and T in Fig. 2.1, consists of four discrete strokes. On the one hand, two isothermal work strokes

2. Otto cycles with a quantum planar rotor

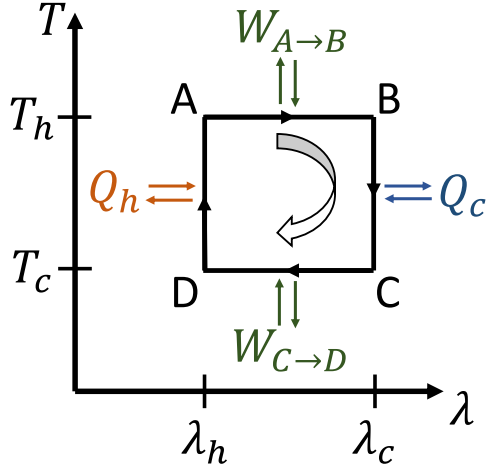


Figure 2.1.: Evolution of the state variables temperature T and control parameter λ within the four-stroke Otto cycle with a generic working medium. The horizontal lines correspond to an isothermal change, where the control parameter is varied from λ_h to λ_c , or vice versa. The two vertical lines represent “isochoric” thermalization of the working medium, where heat is exchanged with a hot reservoir at temperature T_h (left) and a cold reservoir at T_c at a fixed value of the control parameter λ . Together with the specific choice of Hamiltonian $\hat{H}(\lambda)$, the four extremal parameters λ_c , λ_h , T_c , and T_h fully characterize an Otto cycle.

($A \rightarrow B$ and $C \rightarrow D$) in which the control parameter is varied between λ_h and λ_c while the system is isolated. On the other hand, two “isochoric” thermalization strokes ($B \rightarrow C$ and $D \rightarrow A$), where the system is coupled to either the hot (T_h) or the cold (T_c) reservoir at constant control parameter $\lambda \in \{\lambda_c, \lambda_h\}$. We assume idealized, infinitely slow thermalization strokes that result in the two respective Gibbs states,

$$\hat{\gamma}_j = \frac{1}{Z(\lambda_j, T_j)} \exp \left[-\frac{\hat{H}(\lambda_j)}{k_B T_j} \right], \quad j \in \{h, c\}. \quad (2.2.12)$$

On the contrary, we assume fast, quasi-instantaneously changes of the work parameter $\lambda_h \leftrightarrow \lambda_c$ for the work strokes, keeping the initial thermal populations of the system’s energy levels constant.¹

In a general stroke, the change in the system energy is the sum of the work and heat flows, as stated by the first law of thermodynamics discussed in Subsection 1.4.1. For the Otto cycle, however, the energy change of any stroke can be fully attributed to either work or heat. We thus identify the mean energy change in each thermalization stroke

¹In the magnetic dipole setting, an adiabatically slow ramp between λ_h and λ_c is equivalent to an instantaneous quench, because the energy basis remains unchanged.

2. Otto cycles with a quantum planar rotor

with fixed parameter λ as the respective net heat input,

$$\begin{aligned} Q_c &= \langle H_c \rangle_c - \langle H_c \rangle_h = \text{tr}\{\hat{H}(\lambda_c)(\hat{\gamma}_c - \hat{\gamma}_h)\} \\ Q_h &= \langle H_h \rangle_h - \langle H_h \rangle_c = \text{tr}\{\hat{H}(\lambda_h)(\hat{\gamma}_h - \hat{\gamma}_c)\}. \end{aligned} \quad (2.2.13)$$

The mean energy at the control parameter value λ_i in thermal equilibrium at temperature T_j is abbreviated as $\langle H_i \rangle_j = \text{tr}\{\hat{H}(\lambda_i)\hat{\gamma}_j\}$. These four mean energy values fully characterize the operation mode and the performance, e.g. work output and efficiency, of the ideal Otto cycle.

In the two isochoric strokes, the change in mean energy corresponds to a change in the control parameter and is thus identified as work,

$$\begin{aligned} W_{A \rightarrow B} &= \langle \hat{H}_c \rangle_h - \langle \hat{H}_h \rangle_h = \text{tr}\left\{\left[\hat{H}(\lambda_c) - \hat{H}(\lambda_h)\right]\hat{\gamma}_h\right\} \\ W_{C \rightarrow D} &= \langle \hat{H}_c \rangle_h - \langle \hat{H}_h \rangle_h = \text{tr}\left\{\left[\hat{H}(\lambda_h) - \hat{H}(\lambda_c)\right]\hat{\gamma}_c\right\}. \end{aligned} \quad (2.2.14)$$

Applying the first law over a complete cycle requires that the net total work output is given by,

$$W = W_{A \rightarrow B} + W_{C \rightarrow D} = -(Q_c + Q_h). \quad (2.2.15)$$

Three operation modes can be distinguished for such an ideal Otto cycle. When the cycle yields a net work output, i.e. $W < 0$, it is called *engine* with efficiency $\eta = |W|/Q_h$. A *refrigerator*, on the other hand, draws heat from the cold reservoir, $Q_c > 0$. In any other case, the cycle neither produces a work output nor reverses the heat flow. Then we consider it to be useless and call it *heater*.

Throughout this section, we consider a classical or quantum planar rotor, described by a Hamiltonian of the form (2.2.1), as the working medium. In both of the following case studies, the control parameter λ represents the strength of an external electric or magnetic field, leading to a change in the potential strength V or the boost frequency ω . For the classical treatment, we use the phase space descriptions (2.2.2) and (2.2.3), instead of the respective Gibbs states (2.2.12), partition functions and averages.

2.2.3. Fluctuations of work

For macroscopic working media, the inherently random thermodynamic quantities are well represented by their mean value since fluctuations are negligible. This changes for working media of microscopic size, as we discuss in Subsection 1.4.3. In order to quantify the fluctuations of work, we employ the concept of the operator of work. Recall from Subsection 1.4.3 that, given a parametric Hamiltonian $\hat{H}(\lambda(t))$ that generates the unitary time evolution $\hat{U}(t)$, the operator of work is defined as,

$$\hat{W}_{0 \rightarrow t} = \hat{U}(t)^\dagger \hat{H}(\lambda(t)) \hat{U}(t) - \hat{H}(\lambda(0)). \quad (2.2.16)$$

In the following case studies, the Hamiltonian (2.2.1) can always be brought into the generic form $\hat{H} = \hat{H}_0 + \lambda \hat{V}$ and the work strokes are idealized to be quasi-instantaneous

2. Otto cycles with a quantum planar rotor

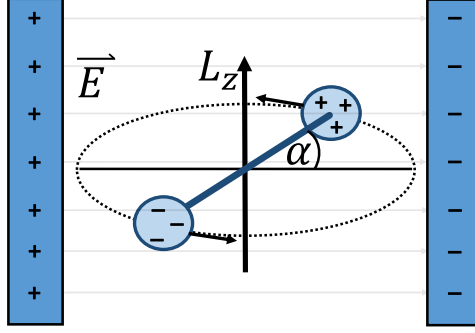


Figure 2.2.: Sketch of a physical setting, where a quantum planar rotor serves as the working medium in an Otto cycle: The rotor possesses an electric dipole moment and is subject to an externally controlled electric field parallel to the rotation plane.

quenches. This allows to explicitly state the operator of work for the two isothermal strokes from points A to B and from C to D in Fig. 2.1,

$$\hat{W}_{A \rightarrow B} = (\lambda_c - \lambda_h) \hat{V} = -\hat{W}_{C \rightarrow D}. \quad (2.2.17)$$

Since the measurements of the two operators are performed with respect to different initial states $\hat{\gamma}_h$ and $\hat{\gamma}_c$, the respective outcome statistics are independent from each other and we can derive the total per-cycle work,

$$\mathbb{V}[W] = (\lambda_c - \lambda_h)^2 (\mathbb{V}_h[V] + \mathbb{V}_c[V]), \quad (2.2.18)$$

with $\mathbb{V}_h[V] = \langle V^2 \rangle_h - \langle V \rangle_h^2$; see App. B.1.1 for details. We will use the *scaled variance*,

$$\frac{\mathbb{V}[W]}{\langle W \rangle^2} = \frac{\mathbb{V}_h[V] + \mathbb{V}_c[V]}{(\langle V \rangle_c - \langle V \rangle_h)^2}, \quad (2.2.19)$$

in order to quantify the the relative amount of work fluctuations. Equations (2.2.18) and (2.2.19) also hold in the classical case, in which quantum expectation values with respect to (2.2.12) are replaced by classical phase space averages with respect to (2.2.2).

2.3. Electric dipole machine

The first setting, we investigate, is depicted in Fig. 2.2: A dumbbell carrying opposite charges at its tips rotates in the xy -plane under the influence of an externally controlled homogeneous electric field directed along the x -axis, such as that produced by a capacitor with variable voltage. The electric field strength $\mathbf{E}_\lambda = E_\lambda \mathbf{e}_x$ is parametrized by the control parameter λ . The charge configuration of the dumbbell induces an electric dipole moment, oriented along the symmetry axis of this rotor $\mathbf{d} = d(\mathbf{e}_x \cos \alpha + \mathbf{e}_y \sin \alpha)$, yielding the potential energy $V(\alpha) = -\mathbf{E}_\lambda \cdot \mathbf{d} = -dE_\lambda \cos \alpha$. Parameterizing the interaction

2. Otto cycles with a quantum planar rotor

such that $dE_\lambda = E\lambda/2$, we find, up to an additive constant, the Hamiltonian

$$H(\lambda) = \frac{L_z^2}{2I} + E\lambda \sin^2\left(\frac{\alpha}{2}\right) = E \left[\frac{L_z^2}{2\hbar^2} + \lambda \sin^2\left(\frac{\alpha}{2}\right) \right]. \quad (2.3.1)$$

Note that this system coincides with mathematical pendulum, which can be approximated by a harmonic oscillator of frequency $\omega_{\text{eff}} = \sqrt{E\lambda/2I}$ in the limit of low excitations and $\lambda \gg 1$. Therefore, the rotor regime $\lambda \sim 1$ will be in the focus of our studies.

2.3.1. Classical description

In the classical regime, the required mean energies $\langle H_i \rangle_j$ can be derived analytically. To this end, we derive the partition function (2.2.3) of the corresponding Gibbs state with respect to the Hamiltonian (2.3.1).

$$Z(\lambda, T) = \sqrt{2\pi I k_B T} 2\pi e^{-E\lambda/2k_B T} I_0\left(\frac{E\lambda}{2k_B T}\right). \quad (2.3.2)$$

The average energies can now be expressed as derivatives of the logarithm of the partition function

$$\begin{aligned} \langle H_i \rangle_j &= \int dL_z \int d\alpha H(\lambda_i) \frac{\exp[-H(\lambda_j)/k_B T_j]}{Z(\lambda_j, T_j)} \\ &= k_B T_j \left[\frac{1}{2} - \frac{\lambda_i}{\lambda_j} \frac{\partial \ln Z(\lambda_j, T_j)}{\partial \lambda_j} \right] \\ &= \frac{k_B T_j}{2} + \frac{E\lambda_i}{2} \left[1 - \frac{I_1(x_j)}{I_0(x_j)} \right], \end{aligned} \quad (2.3.3)$$

with $x_j = E\lambda_j/(2k_B T_j)$ and I_n modified Bessel functions. The heat and work inputs per cycle are given by differences of the energies and read

$$Q_c = \frac{k_B(T_c - T_h)}{2} + \frac{E\lambda_c}{2} \left[\frac{I_1(x_h)}{I_0(x_h)} - \frac{I_1(x_c)}{I_0(x_c)} \right], \quad (2.3.4)$$

$$Q_h = \frac{k_B(T_h - T_c)}{2} + \frac{E\lambda_h}{2} \left[\frac{I_1(x_c)}{I_0(x_c)} - \frac{I_1(x_h)}{I_0(x_h)} \right], \quad (2.3.5)$$

$$W = \frac{E(\lambda_h - \lambda_c)}{2} \left[\frac{I_1(x_h)}{I_0(x_h)} - \frac{I_1(x_c)}{I_0(x_c)} \right]. \quad (2.3.6)$$

The modes of operation are already encoded in the signs of these expressions. In order to characterize the regimes, we note that the function I_1/I_0 is strictly monotonously increasing and hence, inequalities between function values also hold between the respective arguments. First, we focus on the engine regime, where $W < 0$. This is achieved by exactly one of the two factors in (2.3.6) being negative: Either the first factor is positive ($\lambda_h > \lambda_c$) and the second one is negative ($x_c > x_h$), or vice versa. The second case, however, implies $T_c > T_h$ leading to a contradiction. The remaining case can be summarized

2. Otto cycles with a quantum planar rotor

as

$$W < 0 \quad \Longleftrightarrow \quad \frac{T_h}{T_c} > \frac{\lambda_h}{\lambda_c} > 1. \quad (2.3.7)$$

The refrigeration regime, on the other hand, can not be simplified so strongly. We find the implicit characterization

$$Q_c > 0 \quad \Longleftrightarrow \quad \frac{I_1(x_h)}{I_0(x_h)} - \frac{I_1(x_c)}{I_0(x_c)} > \frac{k_B(T_h - T_c)}{E\lambda_c}. \quad (2.3.8)$$

We will now study the work output and cold reservoir heat exchange of such an engine in comparison to the quantum case.

2.3.2. Quantum-classical comparison

We now consider the quantum version of the machine. Unfortunately, an analytical expression for the mean energies $\langle H_i \rangle_j$ is not longer known and numerical methods have to be employed. To this end, we use the quantum optics package for the Julia programming language [148].

The output of the classical(a) and quantum(b) version of the electric dipole machine is presented in Fig. 2.3 for an exemplary parameter setting. While the cold-stroke parameters are fixed at the moderate values $\lambda_c = 1$ and $k_B T_c = E$, the hot-stroke parameters λ_h and T_h are varied. The red- and blue-shaded contour enclosed by the dashed lines correspond to the engine and refrigeration mode of operation, respectively. The quantity of the work output $-W$ and heat transfer to the cold bath Q_c are shown in progressively darker shades. In both cases, the behavior predicted by the classical condition (2.3.7) is observed: Work production occurs when the temperature ratio is greater than the ratio of the work parameters. The more these parameters (and hence the temperatures) differ, the greater the absolute work output. The refrigeration regime is found in the regime of strong coupling parameters $\lambda_h \gg 1$, i.e. strongly confined pendulum motion. Altogether, the behavior of the quantum case is very similar to the classical one, with a small decrease in size of operation regimes and output.

The differences become more pronounced in Fig. 2.4, where the cold bath temperature is reduced to $k_B T_c = 0.05E$, close to the ground state. The refrigeration mode is now no longer visible in the quantum case shown in Fig. 2.4 (b). In general, the quantum machine's disadvantage increases in both the operating regime and output. This agrees with the intuition that quantum Gibbs states of localized or trapped motion typically occupy more phase-space area than classical Gibbs states of the same temperature, and the discrepancy grows at lower temperatures. Hence, from a classical perspective, the quantum machine appears to operate at a lower temperature bias and thus more poorly.

Another confirmation of this intuition can be drawn from the consistently higher work fluctuations in the quantum case. Inserting the control Hamiltonian $\hat{V} = E \sin^2(\hat{\alpha}/2)$

2. Otto cycles with a quantum planar rotor

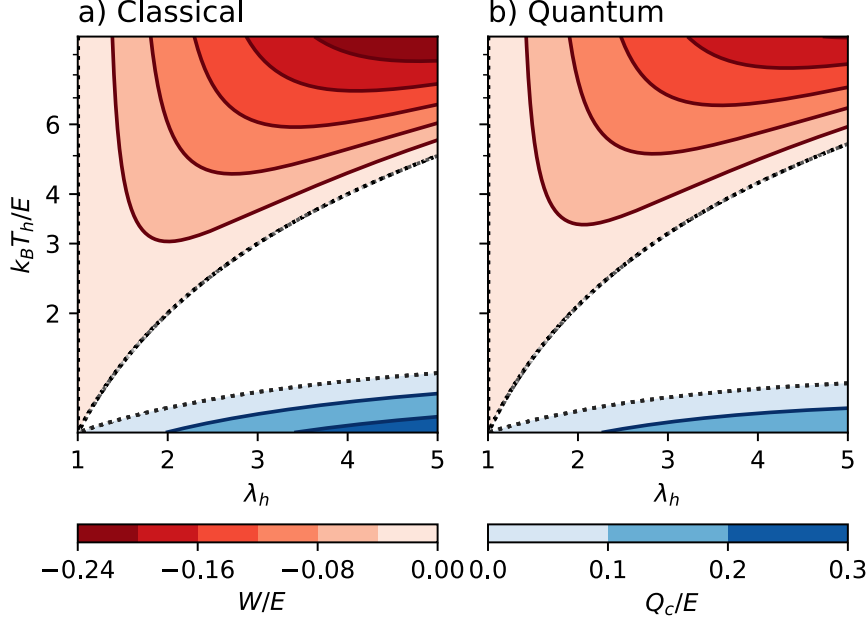


Figure 2.3.: Energy output of an Otto cycle for the electric dipole machine setting with (a) a classical and (b) a quantum planar rotor. We plot against the hot-stroke control parameter λ_h and temperature T_h , distinguishing between two operation modes marked by the dotted lines: Engine operation with work output $W < 0$ (red shades) and refrigeration with heat output $Q_c > 0$ (blue). The cold-stroke parameters are fixed at $k_B T_c = E$ and $\lambda_c = 1$.

into (2.2.19), we obtain the scaled work variance,

$$\frac{\mathbb{V}[W]}{\langle W \rangle^2} = \frac{\mathbb{V}_h[\sin^2(\alpha/2)] + \mathbb{V}_c[\sin^2(\alpha/2)]}{(\langle \sin^2(\alpha/2) \rangle_h - \langle \sin^2(\alpha/2) \rangle_c)^2}. \quad (2.3.9)$$

Classically, this can even be derived analytically, as shown in Appendix B.1.2, while the quantum version is calculated numerically. For both cases, the square root of the scaled variance is presented for the same parameter regime as in Fig. 2.4 in Fig. 2.5. We see that the quantum version (b) possesses consistently higher fluctuations than its classical counterpart (a). A regime, where $\sqrt{\mathbb{V}[W]} \leq \langle W \rangle$, can only be observed classically. Quantum-mechanically, the work is always dominated by fluctuations.

That quantum mechanics can also be advantageous in rotor based otto cycles, is highlighted in the next case study.

2.4. Magnetic dipole machine

The second setting, we consider, is depicted in Fig. 2.6: A dumbbell carrying similar charges at its tips rotates in the xy -plane under the influence of an externally controlled

2. Otto cycles with a quantum planar rotor

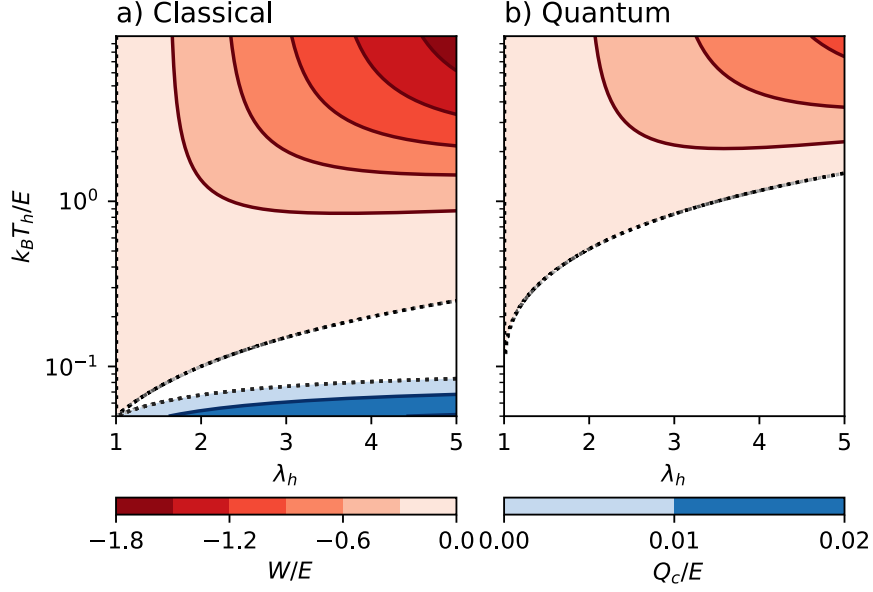


Figure 2.4.: Energy output of (a) the classical and (b) the quantum electric dipole machine, plotted in the same manner as in Fig. 2.3 as a function of the hot-stroke control parameter λ_h and temperature T_h . Here, we set the cold-stroke temperature to a lower value, $k_B T_c = 0.05E$.

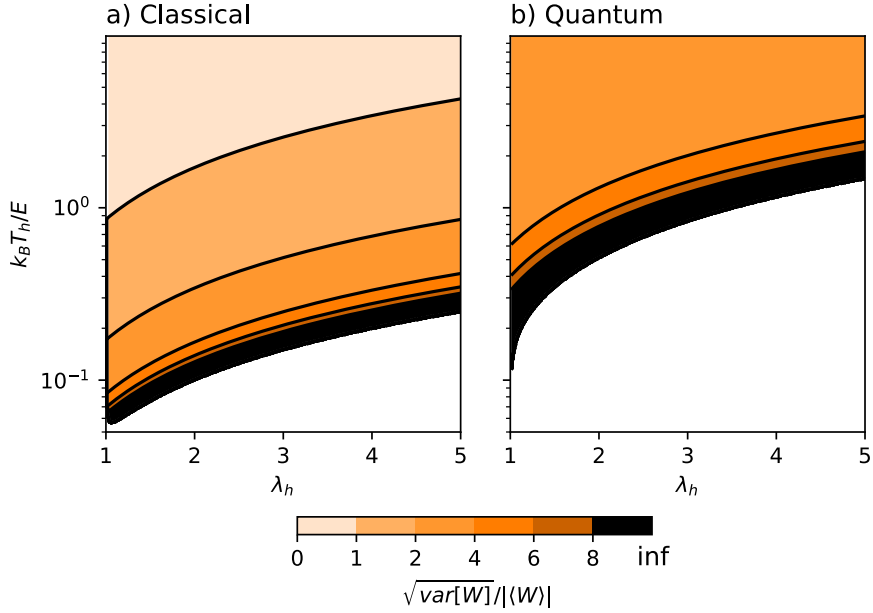


Figure 2.5.: Square root of the scaled work variance for (a) the classical and (b) the quantum electric dipole machine, plotted against against the hot-stroke control parameter λ_h and temperature T_h for the same settings as in Fig. 2.4. Unshaded regions correspond to no work output ($\langle W \rangle \geq 0$), and the black shades represent a diverging scaled work variance.

2. Otto cycles with a quantum planar rotor

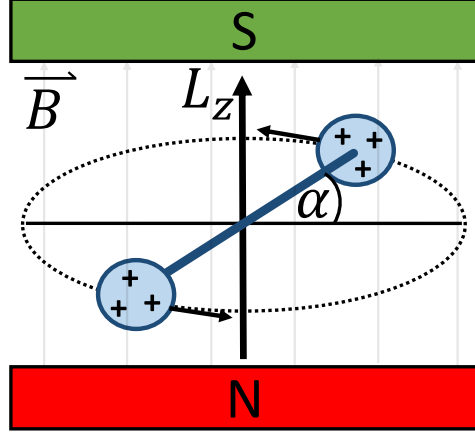


Figure 2.6.: Sketch of a physical setting, where a quantum planar rotor serves as the working medium in an Otto cycle: The rotor possesses a magnetic dipole moment and is subjected to an externally controlled magnetic field, perpendicular to the rotation plane.

homogeneous magnetic field perpendicular to the plane, $\mathbf{B}_\lambda = B_\lambda \mathbf{e}_z$. The rotation of the dumbbell creates a circular current which is associated to a magnetic moment proportional to the angular momentum, $\boldsymbol{\mu} = \mu(L_z/\hbar)\mathbf{e}_z$. The potential energy of the magnetic moment in the external field is $V = -\boldsymbol{\mu} \cdot \mathbf{B}_\lambda = -\omega_\lambda L_z$, where we defined the controlled Larmor frequency $\omega_\lambda = \mu B_\lambda/\hbar \equiv \lambda\hbar/I$. The total Hamiltonian reads

$$H(\lambda) = \frac{L_z^2}{2I} - \omega_\lambda L_z = E \left[\frac{L_z^2}{2\hbar^2} - \lambda \frac{L_z}{\hbar} \right], \quad (2.4.1)$$

where $\hbar\lambda$ determines the net angular momentum displacement that minimizes the energy in the field.

From (2.4.1), we find the relevant mean energies, the total per-cycle work (2.2.15), and the cold-bath heat input Q_c in (2.2.13), holding for the classical and the quantum case

$$\langle H_i \rangle_j = \frac{\langle L_z^2 \rangle_j}{2I} - \frac{E\lambda_i}{\hbar} \langle L_z \rangle_j, \quad (2.4.2)$$

$$W = E(\lambda_h - \lambda_c) \frac{\langle L_z \rangle_h - \langle L_z \rangle_c}{\hbar}, \quad (2.4.3)$$

$$Q_c = \frac{\langle L_z^2 \rangle_c - \langle L_z^2 \rangle_h}{2I} - E\lambda_c \frac{\langle L_z \rangle_c - \langle L_z \rangle_h}{\hbar}. \quad (2.4.4)$$

2.4.1. Classical no-go result

We will now see that a classical planar rotor with a magnetic dipole setup cannot operate as an Otto engine or a refrigerator, no matter the choice of temperatures or control parameters. After completing the square in (2.4.1), the classical partition function is

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straightforwardly derived to be

$$Z(\lambda, T) = Z(0, T)e^{E\lambda^2/2k_B T} = 2\pi\hbar\sqrt{\frac{2\pi k_B T}{E}} e^{E\lambda^2/2k_B T}. \quad (2.4.5)$$

Taking derivatives of (2.4.5) we find all desired expectation values: For the classical angular momentum, we find

$$\langle L_z \rangle_j = \hbar \frac{k_B T_j}{E} \frac{\partial}{\partial \lambda_j} \ln Z(\lambda_j, T_j) = \hbar \lambda_j. \quad (2.4.6)$$

The mean energies become

$$\begin{aligned} \langle H_i \rangle_j &= k_B T_j \left[T_j \frac{\partial}{\partial T_j} + (\lambda_j - \lambda_i) \frac{\partial}{\partial \lambda_j} \right] \ln Z(\lambda_j, T_j) \\ &= \frac{k_B T_j}{2} + \frac{E \lambda_j}{2} (\lambda_j - 2\lambda_i). \end{aligned} \quad (2.4.7)$$

The resulting work (2.4.3) and cold-bath heat (2.4.4) are

$$W = E(\lambda_h - \lambda_c)^2 > 0, \quad (2.4.8)$$

$$Q_c = -\frac{k_B(T_h - T_c)}{2} - \frac{E}{2}(\lambda_h - \lambda_c)^2 < 0, \quad (2.4.9)$$

which precludes any useful operation of the Otto cycle.

2.4.2. Quantum machine operation

We will now see that, given a cold stroke sufficiently close to the ground state, an engine and a refrigerator mode are enabled in the quantum version of the magnetic dipole engine. The performance, characterized by the mean energies $\langle H_i \rangle_j$, is given by derivatives of the partition function, as in the first line of (2.4.7). Equations (2.4.2) and (2.4.4) also hold, but with the expectation values taken over the quantum Gibbs state at T_h and T_c . For the quantum case, the partition function is a discrete sum of the quantized angular momentum levels. By using the Poisson summation rule [149], which states that we can sum over the Fourier transform instead of the function itself, we find the quantum partition function as a product of the classical partition (2.4.5) function

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(in units of Plank's action quantum) and a Jacobi theta function [150]

$$\begin{aligned}
Z(\lambda, T) &= \sum_{m=-\infty}^{+\infty} e^{-Em(m-2\lambda)/2k_B T} \\
&= \sqrt{\frac{2\pi k_B T}{E}} e^{E\lambda^2/2k_B T} \sum_{\nu=-\infty}^{+\infty} e^{-2\pi\nu(i\lambda + \pi\nu k_B T/E)} \\
&= \sqrt{\frac{2\pi k_B T}{E}} e^{E\lambda^2/2k_B T} \vartheta_3\left(-\pi\lambda, e^{-2\pi^2 k_B T/E}\right). \tag{2.4.10}
\end{aligned}$$

With the partition function at hand, the quantum expectation value of angular momentum can be derived. We split it into $\langle L_z \rangle =: \hbar(\lambda + \epsilon)$, where $\hbar\lambda$ is the classical mean value and ϵ is the quantum correction. Invoking an identity for the derivative of the Jacobi theta function, we derive an explicit Fourier expansion of the deviation [150],

$$\begin{aligned}
\epsilon &= \frac{\langle L_z \rangle}{\hbar} - \lambda = \frac{k_B T}{E} \frac{\partial \ln Z(\lambda, T)}{\partial \lambda} - \lambda \\
&= \sum_{n=1}^{\infty} (-)^n \frac{2\pi k_B T/E}{\sinh(2\pi^2 n k_B T/E)} \sin(2\pi n \lambda). \tag{2.4.11}
\end{aligned}$$

The resulting average momentum $\langle L_z \rangle$ as a function λ is depicted in Fig. 2.7 for different equilibrium temperatures. For integer or half-integer values of λ , classical and quantum expectation value coincide for any T . Independent of λ , the quantum value approaches the classical value for increasing temperatures. Already at $k_B T = 0.5E$ (green line), quantum and classical value are visibly not distinguishable. In the limit of small temperatures $k_B T \ll E$ (blue curve), on the other hand, the coefficients of the Fourier series of the quantum correction ϵ (2.4.11) converge to $(-)^n/\pi n$, which corresponds to a triangular saw-tooth pattern. Added to the linear function $\lambda \mapsto \hbar\lambda$, this yields a stepwise increase in $\langle L_z \rangle$ at every half-integer λ .

Qualitatively, the behavior of $\lambda \mapsto \langle L_z \rangle$ can be well understood by physical arguments: Recall that the system Hamiltonian is diagonal in the angular momentum basis $\{|m\rangle\}$ with the energy eigenvalues $E_m = E(m - \lambda)^2/2 - E\lambda^2/2$, lying on a parabola. This parabola has reflection symmetry around $m = \lambda$. Note however, that this symmetry is, in general, not manifested in the spectrum due to the discrete nature of the indices $m \in \mathbb{Z}$. Two exceptions are integer ($\mathbb{Z}$) and half-integer ($\mathbb{Z} + \frac{1}{2}$) values of λ : For $\lambda \in \mathbb{Z}$, there is a unique ground state $m = \lambda$ minimizing the energy and degenerate energy doublets at $m = \lambda \pm n$ with $n \in \mathbb{N}$. In thermal equilibrium, the probability of finding the system in either state of a doublet is equal and thus the contribution of any doublet to the mean angular momentum averages to $\hbar\lambda$. This yields a mean angular momentum of $\langle L_z \rangle = \hbar\lambda$ for any temperature T . For $\lambda \in \mathbb{Z} + \frac{1}{2}$, the degenerate energy levels are of the form $m = \lambda \pm (n - \frac{1}{2})$ with $n \in \mathbb{N}$, where the ground state is now degenerate, too. Again, the contribution from any doublet averages to $\hbar\lambda$ and we find $\langle L_z \rangle = \hbar\lambda$ for any temperature T . Hence, deviations from the classical behavior can only be observed

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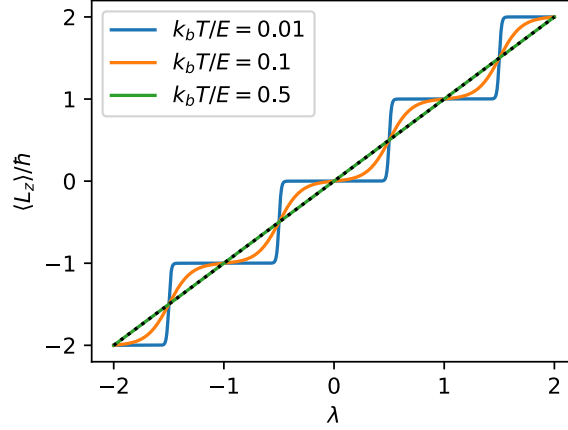


Figure 2.7.: Mean angular momentum of a quantum planar rotor in Gibbs states of three different temperatures, with respect to the magnetic dipole Hamiltonian (2.4.1) of varying momentum displacement λ . The dotted line marks the classical mean value $\langle L_z \rangle = \hbar \lambda$, achieved at high temperatures.

for $\lambda \bmod 1/2 \neq 0$. In the limit of small temperatures, the Gibbs state approaches the pure state at minimal energy, the ground state. Here, the ground state is given by $|\lfloor \lambda \rfloor\rangle$, where $\lfloor \lambda \rfloor$ is the integer closest to λ . Conclusively, $\langle L_z \rangle = \hbar(\lambda + \epsilon) \approx \hbar \lfloor \lambda \rfloor$ and thus $|\epsilon| < |\lambda - \lfloor \lambda \rfloor| < 1/2$. In summary, we explained the following behavior

$$\langle L_z \rangle \approx \begin{cases} \hbar \lfloor \lambda \rfloor & \text{for } T \rightarrow 0 \text{ and } \lambda \notin \mathbb{Z} + \frac{1}{2} \\ \hbar \lambda & \text{for } \lambda \in \mathbb{Z} + \frac{1}{2} \\ \hbar \lambda & \text{for } T \rightarrow \infty. \end{cases} \quad (2.4.12)$$

This behavior of the average angular momentum is crucial for the impact of quantization on the machine operation modes. W.l.o.g. we can restrict ourselves to the case $\lambda_c > \lambda_h$ since the opposite case is treated analogously and does not show qualitatively different behavior. The per-cycle work (2.4.3) can be rewritten in terms of the quantum corrections ϵ_c, ϵ_h

$$W = E(\lambda_c - \lambda_h)^2 \left(1 + \frac{\epsilon_c - \epsilon_h}{\lambda_c - \lambda_h} \right), \quad (2.4.13)$$

where a negative sign implies the engine regime. Since the triangle inequality implies $|\epsilon_c \pm \epsilon_h| \leq |\epsilon_c| + |\epsilon_h| = 1$, the control parameters have to fulfill the necessary condition $\lambda_c - \lambda_h < 1$. In addition, we find integer offsets of both parameters $\lambda_{h,c} \rightarrow \lambda_{h,c} + m$ to leave the work invariant because the quantum corrections are periodic, as can be seen from (2.4.11). Summarizing, we can restrict ourselves to study the engine regime for the parameters $1 \geq \lambda_c \geq \lambda_h \geq 0$.

In the high temperature regime, i.e. $k_B T_{h,c} > E$, the quantum corrections vanish and we find, analogous to the classical machine, no work output. For small temperatures, i.e. in the deep quantum regime $k_B T_{h,c} \ll E$, we obtain $W \approx E(\lambda_c - \lambda_h)(\lfloor \lambda_c \rfloor - \lfloor \lambda_h \rfloor)$

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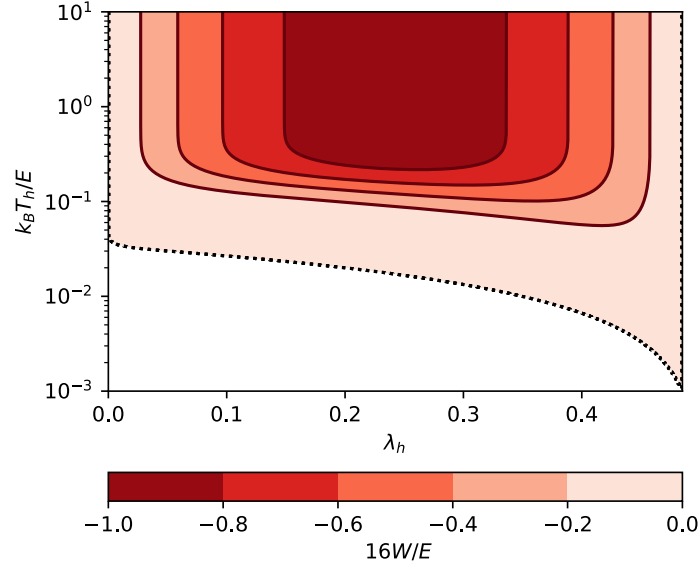


Figure 2.8.: Energy output of an Otto cycle for the magnetic dipole machine setting with a quantum planar rotor. We plot against the hot-stroke control parameter λ_h and temperature T_h , distinguishing between two operation modes marked by the dotted lines: engine operation with work output $W < 0$ (red shades) and refrigeration with heat output $Q_c > 0$ (blue). The cold-stroke parameters are fixed at $k_B T_c = 0.001E$ and $\lambda_c = 0.485$. The work output is normalized with respect to theoretically predicted optimum $W = -E/16$, see Equation (2.4.14).

by using (2.4.12) in (2.4.3). Due to the monotonicity of rounding, this expression fulfills $W > 0$ and hence does not allow an engine operation.

In the remaining regime of comparably low T_c and comparably high T_h , the machine is found to admit an appreciable work output: In this case, we find the work output $W \approx E(\lambda_c - \lambda_h)([\lambda_c] - \lambda_h)$, which becomes negative if and only if $\lambda_c > \lambda_h > [\lambda_c]$. This, on the other hand, requires $\lambda_{c,h} < 0$ in order to achieve $[\lambda_c] = 0$. The work output becomes

$$\frac{W}{E} \approx -(\lambda_c - \lambda_h) \lambda_h = \left(\lambda_h - \frac{\lambda_c}{2} \right)^2 - \frac{\lambda_c^2}{4} > -\frac{1}{16}, \quad (2.4.14)$$

where we completed the square to find the optimal value: Setting $\lambda_h = \lambda_c/2$ results in the maximum work output of $E\lambda_c^2/4$ per cycle for a given $\lambda_c \in (0, 1/2)$. Summarizing, the maximal work output $E/16$ is given for λ_c close to (but slightly smaller) than $1/2$, $\lambda_h = \lambda_c/2$, $T_c \rightarrow 0$, and T_h sufficiently big.²

In Fig. 2.8, we present the work output in the optimal engine regime as a function of the hot-stroke parameters λ_h and T_h for fixed $\lambda_c = 0.485$ and $k_B T_c = 0.001E$. As predicted, the optimal work, close to $W \approx -E/16$, is achieved at $\lambda_h = \lambda_c/2 \approx 0.25$ and for temperatures $k_B T_h > 0.2E$.

²In the alternative case $\lambda_c < \lambda_h$, the maximum work output in (2.4.14) is achievable by setting $\lambda_h = (1 + \lambda_c)/2$, in the asymptotic limit of $\lambda_c \rightarrow 1/2$ from above, vanishing T_c and high T_h .

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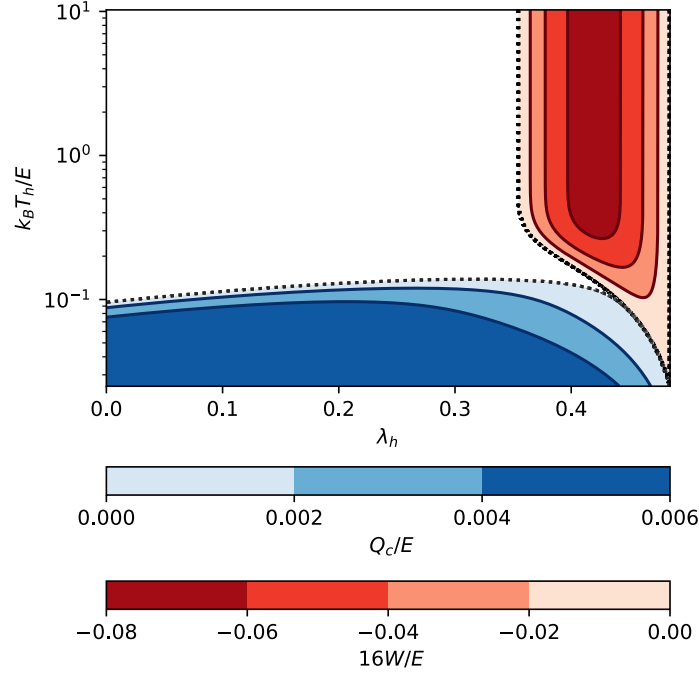


Figure 2.9.: Energy output of the magnetic dipole machine, plotted in the same manner as Fig. 2.8 against the hot-stroke parameter λ_h and the temperature T_h . The cold-stroke parameters are fixed at $k_B T_c = 0.025E$ and $\lambda_c = 0.485$.

An increase of the cold bath temperature closes the engine operation window and nullifies the work output. An illustration of this is given in Fig. 2.9, where we set $k_B T_c = 0.025E$. On the other hand, the machine can also operate as a refrigerator, provided the hot bath temperature is sufficiently small, i.e. $k_B T_h < 0.1E$.

All things considered, we have demonstrated that the magnetic dipole machine supports useful operation modes in the low-temperature regime, due to angular momentum quantization. This regime, however, is dominated by fluctuations, as indicated by the scaled work variance (2.2.19). With the identification $\hat{V} = -E\hat{L}_z/\hbar$ it reads

$$\frac{\mathbb{V}[W]}{\langle W \rangle^2} = \frac{\mathbb{V}_h[L_z] + \mathbb{V}_c[L_z]}{(\langle L_z \rangle_h - \langle L_z \rangle_c)^2}. \quad (2.4.15)$$

By exploiting the particular parameter choices in the relevant operation regime, this expression can be further simplified: For temperatures of almost zero T_c and high T_h , we find $\mathbb{V}_c[L_z] \approx 0$, $\langle L_z \rangle_c \approx 0$, $\langle L_z \rangle_h \approx \hbar\lambda_h$, and $\mathbb{V}_h[L_z] \approx \hbar^2 k_B T_h/E$, where the latter expression is the classical variance calculated in (2.4.7) because we concluded that $k_B T_h \gtrsim 0.5E$ is already sufficient for the classical limit to be valid. This yields

$$\frac{\mathbb{V}[W]}{\langle W \rangle^2} \approx \frac{\mathbb{V}_h[L_z]}{\hbar^2 \lambda_h^2} = \frac{1}{\lambda_h^2} \frac{k_B T_h}{E} \geq 2. \quad (2.4.16)$$

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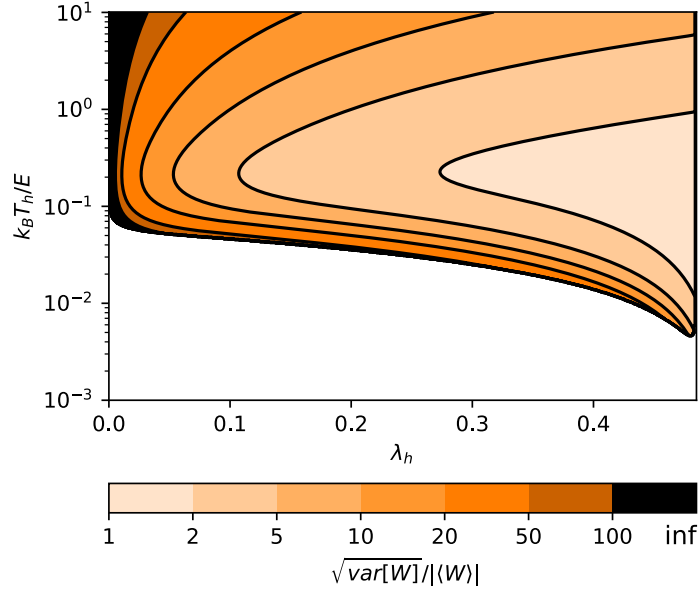


Figure 2.10.: Square root of the scaled work variance for the quantum magnetic dipole machine, plotted against the hot-stroke control parameter λ_h and temperature T_h for the same settings as in Fig. 2.8. The unshaded region corresponds to no work output, and the black region is where the scaled variance diverges.

The lower bound follows from the necessary condition for engine operation $\lambda_h < \lambda_c \in (0, 1/2)$. Hence, the work fluctuations always exceed the mean value. This gets even worse for an increasing hot-bath temperature. This derivation is well confirmed by Fig. 2.10, which shows the square root of the scaled variance (2.2.19) for the same parameters as in Fig. 2.8.

2.5. Conclusion

Our work explores two physically motivated realizations of an Otto cycle with a planar rotor as the working medium, emphasizing how quantization leads to differences in operational regimes and performance. The first realization consists of a rotating electric dipole subject to an electric field of controlled strength parallel to the rotation plane. Thus, the system is equivalent to a mathematical pendulum. We found that the quantized rotor had consistently lower energy output and smaller engine and refrigerator operation windows at low temperatures.

In the second realization, we considered a charged rotor that generates a magnetic moment proportional to its angular momentum subject to a controlled external magnetic field. While we could show that the classical machine does not admit any useful operation mode, we could locate and characterize an engine operation mode for the quantum version. In this operation mode, the rotor state cycles between a deeply quantum cold temperature regime of almost no excitations and a quasi-classical hot regime of arbitrary

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ily high excitations. Hence, the presented model constitutes a genuine quantum thermal machine, distinct from previously studied models based on finite-dimensional systems with only a few levels.

Possible experimental platforms to demonstrate the discussed Otto machine models and their quantum features could be molecular rotors, levitated nanorotors, or with Josephson loops in circuit QED. The magnetic dipole engine, for example, could be demonstrated with a charged nanorotor electrically slowed and aligned in a Paul trap [143]. One could then apply a switchable magnetic field to implement the Otto cycle in a proof of principle experiment. Other mechanical degrees of freedom with a non-homogeneous spectrum could be investigated as working medium for similar genuine quantum machines in future theoretical work.

3. Thermodynamics in a scattering environment

With his famous equation in 1872, Ludwig Boltzmann provided a derivation of gas dynamics based on a statistical treatment of microscopic collisions [151]. Over a century later, Dümcke used similar arguments and quantum scattering theory in order to derive the evolution of a finite-dimensional quantum system subjected to a quantum gas [55]. Follow-up works in the years thereafter were mainly focused on the effect on motional degrees of freedom and their diffusion and decoherence [152–156], leading to the formulation of the quantum version of the linear Boltzmann equation and its extension to internal degrees of freedom [157, 158].

The birth of quantum thermodynamics can be traced back to a seminal work in 1959 proposing a thermodynamic interpretation of the 3-level maser [76]. The field emerged together with the theory of open quantum systems and the ability to model the effect of thermal baths on quantum particles [159]. A versatile operational approach to constructing models for open quantum dynamics, is provided by the framework of repeated interactions [51]: the system is evolved by means of repeated unitary interactions with a sequence of ancilla systems in discrete time steps. The computational advantage of this approach, however, comes at the price of an algorithmic and sometimes even artificial treatment of physics. For example, we will show in Subsection 3.1.1 that a system qubit, repeatedly interacting with other ancilla qubits via the so-called Flip-Flop interaction, only thermalizes when the ancilla qubits possess the same energy gap as the system. Put differently, the environment has to be tailored to the system of interest in order to observe physically reasonable dynamics. This is also reflected in [15], where the author explains how the laws of thermodynamics have to be understood in the context of the framework of repeated interactions.

The desire to reconcile collision models and physical reality lead to the recent rediscovery and extension to internal degrees of freedom of scattering based collision models for gas environments in a thermodynamic context, that were discussed in great detail so far restricted to the one-dimensional case [53, 160–163]. Unfortunately, the one-dimensional nature of these treatments misses some crucial features of a realistic three-dimensional environment: For example, the statistical distribution of momentum in the gas needs to be modified to the so-called effusion distribution [54].

In this work, we combine three-dimensional scattering theory with structured gas particles in order to derive a thermodynamic consistent master equation: First, we briefly introduce repeated interaction models and illustrate their shortcomings using the example of the Flip-Flop interaction in Section 3.1. Next, we derive a scattering-based master equation for internal degrees of freedom in Section 3.2. The consistency of this

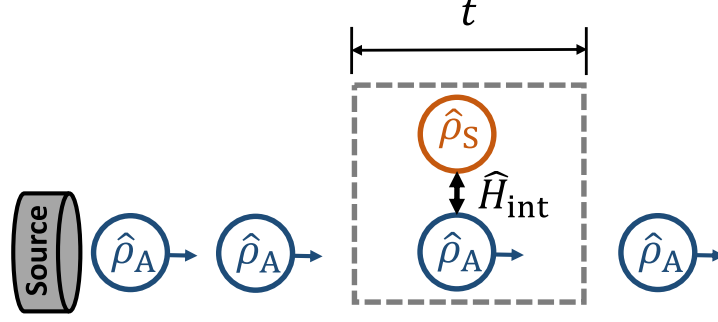


Figure 3.1.: Sketch of a basic repeated interaction model: The system S in state $\hat{\rho}_S$ interacts sequentially with ancilla systems in state $\hat{\rho}_A$. They co-evolve with respect to the interaction Hamiltonian $\hat{H}_{\text{int}}$ for a time t . After the interaction the ancilla is discarded.

master equation with the laws of thermodynamics is investigated in Section 3.3. Finally, we discuss several examples assuming a Gaussian interaction potential in Section 3.4. In particular, we explore the possibility of work production in a nonequilibrium setting in which the motional and internal degrees of freedom of the gas are thermal with respect to different temperatures.

The entire chapter is based on publication [B].

3.1. Repeated interaction models and their limitations

In this section, we introduce the framework of *repeated interactions*, a widely used collisional approach to model open systems in quantum thermodynamics [15]. A broad introduction into this field can be found in [51].

The basic scheme is sketched in Fig. 3.1: the time evolution of the system is generated by discrete operations in which the system S is allowed to interact with a sequence of ancillas A in some prepared states. System and ancilla evolve freely according to the free Hamiltonians $\hat{H}_S$ and $\hat{H}_A$. Over a time interval t , an interaction Hamiltonian $\hat{H}_{\text{int}}$ is activated and builds up correlations between S and A . After the interaction, the ancilla is traced out. This interaction is referred to as a 'collision' in these models, even though it is controlled by an externally imposed time dependence. These collisions can be summarized by a quantum channel,

$$\Lambda[\hat{\rho}_S] = \text{tr}_A \left\{ e^{-it(\hat{H}_S + \hat{H}_A + \hat{H}_{\text{int}})/\hbar} (\hat{\rho}_S \otimes \hat{\rho}_A) e^{it(\hat{H}_S + \hat{H}_A + \hat{H}_{\text{int}})/\hbar} \right\}. \quad (3.1.1)$$

With this channel at hand, the time evolution can now be thought of as a step-wise process, where identical operators are concatenated to a Markov sequence, parameters are changed in time, or collisions are applied randomly alternating with the free evolution [51].

3. Thermodynamics in a scattering environment

In a thermodynamic context, the ancilla states are typically assumed to be in a thermal state $\hat{\gamma}_A$ at some temperature T ,

$$\hat{\gamma}_A = \exp\left(-\frac{\hat{H}_A}{k_B T}\right) / Z_A. \quad (3.1.2)$$

If, additionally, the interaction is energy conserving in the sense that $[\hat{H}_S + \hat{H}_A, \hat{H}_{\text{int}}] = 0$, the channel is called a *thermal operation*, as formally defined in [164]. In quantum thermodynamics, thermal operations serve as building block in a more mathematical, resource theoretic approach [3]. They are used to model the interaction with thermal reservoirs. This is mainly due to the built-in energy conservation and the fact that they properly describe thermalization: The Gibbs state γ_S with respect to $\hat{H}_S$ and temperature T is a fix point of the channel Λ , i.e.,

$$\Lambda[\hat{\gamma}_S] = \hat{\gamma}_S. \quad (3.1.3)$$

This is due to the fact that,

$$\hat{\gamma}_S \otimes \hat{\gamma}_A \propto \exp\left(-\frac{\hat{H}_S + \hat{H}_A}{k_B T}\right), \quad (3.1.4)$$

which then commutes with the time evolution in (3.1.1).

The major advantage of the presented framework lies in its algorithmic structure, well suited for numerical calculations: While typical thermal environments are much larger than the system under consideration, repeated interactions only require the treatment of the unitary two-body interaction between system S and an adequate subsystem A of the environment. When the two-body interaction is solved, i.e. Λ is known, the full open system evolution can be found via iteration. Especially for finite dimensional systems this can be conveniently done numerically. In fact, the use of repeated thermal operations can be regarded as the quantum version of heuristic rate equations describing thermally induced state transitions in stochastic thermodynamics. There, thermalization is ensured by the condition of local detailed balance [165].

On the other hand, this algorithmic treatment does not always properly reflect an actual physical situation we aim to describe. This can lead to unexpected and even unphysical behavior, as we will exemplify and discuss in the Subsection 3.1.1.

3.1.1. The Flip-Flop model

In the most elementary setting, we consider a system qubit interacting with a sequence of identically prepared ancilla qubits. Here, the simplest non-trivial interaction is arguably the flip-flop model in which the qubits exchange excitations via the Hamiltonian,

$$\hat{H}_S = \frac{\hbar\omega_S}{2}\hat{\sigma}_z^{(S)}, \quad \hat{H}_A = \frac{\hbar\omega_A}{2}\hat{\sigma}_z^{(A)} \quad (3.1.5)$$

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and the interaction by the exchange of excitations

$$\hat{H}_{\text{int}} = \hbar g_0 \left(\hat{\sigma}_+^{(\text{S})} \hat{\sigma}_-^{(\text{A})} + \hat{\sigma}_-^{(\text{S})} \hat{\sigma}_+^{(\text{A})} \right). \quad (3.1.6)$$

Note that $\hat{H}_{\text{S}} + \hat{H}_{\text{A}}$ is diagonal in the $\sigma_z^{(\text{S})} \otimes \sigma_z^{(\text{A})}$ -eigenbasis and $\text{span}(|10\rangle, |01\rangle)$ is an invariant two-dimensional subspace of $\hat{H}_{\text{int}}$. This allows us to introduce new Pauli operators on the interacting subspace

$$\hat{S}_x = |01\rangle \langle 10| + |10\rangle \langle 01| \quad (3.1.7)$$

$$\hat{S}_y = i |01\rangle \langle 10| - i |10\rangle \langle 01| \quad (3.1.8)$$

$$\hat{S}_z = |10\rangle \langle 10| - |01\rangle \langle 01| \quad (3.1.9)$$

and on the remaining states

$$\hat{F}_z = |11\rangle \langle 11| - |00\rangle \langle 00|. \quad (3.1.10)$$

Since the $(\hat{S}_j)_j$ and $\hat{F}_z$ act on two different orthogonal subspaces, we trivially find the commutator $[\hat{S}_j, \hat{F}_z] = 0$. In the new operators, the Hamiltonian becomes

$$\hat{H} = \hat{H}_{\text{S}} + \hat{H}_{\text{A}} + \hat{H}_{\text{int}} = \hbar \bar{\omega} \hat{F}_z + \hbar \left(\delta \hat{S}_z + g_0 \hat{S}_x \right), \quad (3.1.11)$$

where $\bar{\omega} = (\omega_1 + \omega_2)/2$ is the mean frequency and $\delta = \omega_1 - \omega_2$ is the detuning. By introducing the effective frequency $\Omega = \sqrt{g_0^2 + \delta^2}$ and the normalized vector $\mathbf{n} = g_0/\Omega \mathbf{e}_x + \delta/\Omega \mathbf{e}_z$, we find the even shorter expression,

$$\hat{H} = \hbar \bar{\omega} \hat{F}_z + \hbar \Omega \mathbf{n} \cdot \hat{\mathbf{S}}. \quad (3.1.12)$$

After an interaction time t , the ancilla degree of freedom, initially assumed to be in a thermal state $\hat{\rho}_{\text{th}}(p) = (1-p) |1\rangle \langle 1| + p |0\rangle \langle 0|$, is traced out, and we obtain the channel

$$\Lambda[\hat{\rho}_{\text{S}}] = \text{tr}_{\text{A}} \left\{ \hat{U}(t) (\hat{\rho}_{\text{S}} \otimes \hat{\rho}_{\text{th}}(p)) \hat{U}^\dagger(t) \right\}. \quad (3.1.13)$$

The effect of this channel can also be expressed in terms of transformation of the Bloch vector $\mathbf{r}$,

$$\mathbf{r} \xrightarrow{\Lambda} \begin{pmatrix} 0 \\ 0 \\ 1 - 2p \end{pmatrix} + |A(t)| R(\mathbf{e}_z, \vartheta(t)) \begin{pmatrix} r_x \\ r_y \\ -|A(t)|([1 - 2p] - r_z) \end{pmatrix}, \quad (3.1.14)$$

where $A(t) = \cos(\Omega t) + i(\delta/\Omega) \sin(\Omega t)$ and $R(\mathbf{e}_z, \vartheta(t))$ is a rotation around the z -axis. The derivation can be found in Appendix C.1.1. Note that $\mathbf{e}_z(1 - 2p) = \mathbf{r}_{\text{th}}(p)$ is the

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Bloch vector of the thermal state $\hat{\rho}_{\text{th}}(p)$ and

$$|A(t)|^2 = 1 - \left(\frac{g_0}{\Omega}\right)^2 \sin^2(\Omega t) \leq 1. \quad (3.1.15)$$

This allows us to make a statement about the evolution of the Bloch vector towards the thermal state. The distance of the system Bloch vector to the thermal Bloch vector reduces upon application of the channel,

$$\begin{aligned} |\mathbf{r}' - \mathbf{r}_{\text{th}}(p)| &= |A(t)| \left| R(\mathbf{e}_z, \vartheta(t)) \begin{pmatrix} r_x \\ r_y \\ -|A(t)|([1 - 2p] - r_z) \end{pmatrix} \right| \\ &\leq |A(t)| \left| \begin{pmatrix} r_x \\ r_y \\ r_z \end{pmatrix} - \begin{pmatrix} 0 \\ 0 \\ 1 - 2p \end{pmatrix} \right| = |A(t)| |\mathbf{r} - \mathbf{r}_{\text{th}}(p)|. \end{aligned} \quad (3.1.16)$$

Here we used that the Euclidean distance does not change under rotations and increased the magnitude of the z -component by neglecting $A(t)$ in the vector. The Euclidean distance of the Bloch vectors is equivalent to the trace distance (1.5.34) of the qubit states which then reads,

$$D_Q(\Lambda[\hat{\rho}_S], \hat{\rho}_{\text{th}}(p)) \leq |A(t)| D_Q(\hat{\rho}_S, \hat{\rho}_{\text{th}}(p)). \quad (3.1.17)$$

This shows that the interaction with the ancilla qubit is a contraction with the thermal state as the fix point. If the interaction time t is chosen such that $\cos(\Omega t) \neq 1$ the map is strictly contractive in the sense that the distance between the system state and the thermal state $\hat{\rho}_{\text{th}}(p)$ decreases with every step. The N -fold application of Λ then yields

$$\Lambda^{\circ N}[\hat{\rho}_S] = \underbrace{\Lambda \circ \dots \circ \Lambda}_{N \text{ times}}[\hat{\rho}_S] \xrightarrow{N \rightarrow \infty} \hat{\rho}_{\text{th}}(p). \quad (3.1.18)$$

On first sight, this seems to be the desired thermalization behavior, but notice that $\hat{\rho}_{\text{th}}(p)$ is the thermal state of the ancilla systems. If the system has the same frequency as the ancillae, i.e. detuning $\delta = 0$, then this state is also the correct thermal state of the system. However, if system and ancillae are detuned, the system will converge to the wrong thermal state. Put differently, the system will thermalize properly if and only if system and ancillae are on resonance, $\delta = 0$. The argument extends straightforwardly to general multilevel systems. Correct thermalization is only possible if the transition energies of the ancillae are resonant with the system transitions [52].

We have seen that a qubit, subject to the Flip-Flop collision model, thermalizes only properly if the ancilla qubits are in resonance. This makes evident that the environment has to be tailored to the system of interest. On the other hand, a sufficiently large physical environment of uncontrolled gas particles with motional and internal states in thermal equilibrium should act as a thermal reservoir and thermalize the system regardless of whether the resonances match or not.

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The limitation of repeated interaction models and their discrepancy with respect to physical collisions comes from the properties of thermal operations in the beginning of this section: By definition, a thermal operation has to fulfill energy conservation in the form of

$$\left[\hat{H}_S + \hat{H}_A, \hat{H}_{\text{int}} \right] = 0. \quad (3.1.19)$$

For the Flip-Flop model, however, the commutator reads as,

$$\left[\hat{H}_S + \hat{H}_A, \hat{H}_{\text{int}} \right] = \frac{\hbar^2 g_0}{2} (\omega_S - \omega_A) \left(\hat{\sigma}_+^{(S)} \hat{\sigma}_-^{(A)} - \hat{\sigma}_-^{(S)} \hat{\sigma}_+^{(A)} \right). \quad (3.1.20)$$

The only non-trivial way to make the right hand side vanish is to have the frequencies be on resonance, $\omega_S = \omega_A$. Hence, the property that makes the Flip Flop model a thermal operation is the same property that we found manually in order to achieve proper thermalization. This shows that, in general, the energy conservation condition (3.1.19) is a necessary condition for thermalization.

A careful examination of why energy conservation must be enforced manually offers significant insight: although $\hat{H}_S$, $\hat{H}_A$, and $\hat{H}_{\text{int}}$ are explicitly time independent, the full Hamiltonian is not, because the interaction is only switched on for a time t . Thus mathematically speaking, the Hamiltonian beyond the interaction window $[t + t_0, t_0]$ at time τ reads,

$$\hat{H}(\tau) = \hat{H}_S + \hat{H}_A + \Theta(\tau - t_0) \Theta(t + t_0 - \tau) \hat{H}_{\text{int}}, \quad (3.1.21)$$

and will, in general, not conserve energy. Physically, we can think of an external controller who varies certain control fields in order to switch the interaction on and off. The change in the control fields itself can then add energy to the system, unless (3.1.19) is fulfilled.

This demonstrates that the idealizations in repeated interaction models are too strong in some cases. In the following we will tackle the problem by using scattering theory for a more physical treatment of the collision: the inclusion of motional degrees of freedom and a position-dependent interaction allows for the replacement of the external controller while keeping the time-independence of the full Hamiltonian. This type of collision model is the focus of the remainder of the chapter.

3.2. Master equation for the evolution in a dilute gas environment

In contrast to the repeated interaction models, we here promote the ancilla system interacting with our system S to a gas particle that possesses motional (M) and internal degrees of freedom (A). The timing of the controlled interactions is replaced by the random motion of the particles. The goal of this section is to derive a master equation for the effective dynamics of a general system S subjected to such a gas.

Several of such microscopic derivations can be found in the literature [55, 156, 166]. Although the resulting master equations share the same form apart from proportion-

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ality factors, the derivations themselves and their underlying assumptions can differ significantly. Before we go through our detailed derivation in the next subsections, we summarize the crucial derivation steps in the remainder of the current subsection. This does not only serve as a summary of the following subsections but also provides some insight in the approach of different authors. For the derivation, we follow along the lines of Dümcke [55], Diosi [166], and Hornberger [156] before we extend the framework to ancillae with internal degrees of freedom. The derivation occurs in four steps, denoted in the following list:

- 1) Reduction of the many-body description: We use the BBGKY-hierarchy to reduce the dynamics to an effective two-body evolution (Detailed discussion in Subsection 3.2.2). The derivation by Dümcke is also based on this hierarchy. Diosi starts already from a two-body scattering interaction. A more stochastic approach is found in Hornbergers work, where a positive operator determines the probability to scatter with a single gas particle.
- 2) Coarse-grained time evolution: We describe the evolution on time scales longer then the duration of the average collision. The instantaneous change due to a collision is then well described by scattering theory. (Detailed discussion in Subsection 3.2.3).
- 3) Trace over the motional degrees of freedom: The idealized scattering description results in the appearance of delta functions evaluated at zero. This is a fundamental problem that we solve with a regularization procedure. (Subsection 3.2.4). his Problem is solved by Dümcke and Diosi by the same regularization procedure, while Hornberger applies an analysis of wavepackets. A comparison of the two approaches is given in [167].
- 4) Trace over the internal degrees of freedom (Subsection 3.2.5).

A detailed discussion of the steps is presented in the following subsections.

3.2.1. Thermal state of a single gas particle

The thermal state of a system with Hamiltonian $\hat{H}$ at temperature T is given by the following unit-trace operator $\hat{\gamma} = \exp(-\hat{H}/(k_B T))/Z$, as discussed in Subsection 1.4.2. However, the thermal state of a free gas particle with Hamiltonian,

$$\hat{H}_{\text{kin}} = \frac{\hat{\mathbf{p}}^2}{2m}, \quad (3.2.1)$$

has to be uniformly distributed in position space. This makes it impossible to properly normalize the thermal state with respect to $\hat{H}_{\text{kin}}$ ¹, which mathematically manifest in a singular partition function Z .

¹Note that it is, for example, impossible to have an equal distribution on $\mathbb{N}$ or $\mathbb{R}$

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This singularity can be avoided by restricting the particles position to a large box with equally long sides of length L . Assuming periodic boundary conditions yields an eigenbasis of proper momentum eigenstates $|\mathbf{n}\rangle$ with $\mathbf{n} \in \mathbb{Z}^3$ and the momenta,

$$\mathbf{p}_{\mathbf{n}} = \left(\frac{2\pi\hbar}{L} \right) \mathbf{n}. \quad (3.2.2)$$

In this basis, the partition function is properly defined and reads,

$$Z_{\text{free}} = \sum_{\mathbf{n} \in \mathbb{Z}^3} \langle \mathbf{n} | \exp \left(-\frac{1}{2} \left(\frac{\mathbf{p}}{p_{\text{th}}} \right)^2 \right) | \mathbf{n} \rangle = \left(\sum_{n \in \mathbb{Z}} \exp \left(-\frac{1}{2} \left(\frac{p_n}{p_{\text{th}}} \right)^2 \right) \right)^3, \quad (3.2.3)$$

where we used the factorization of the exponential function in order to make the summation one-dimensional in the last step. In the limit of asymptotically large L , the spacing between momentum eigenvalues becomes small and the partition function can be approximated by an integral,

$$\sum_{n \in \mathbb{Z}} \exp \left(-\frac{1}{2} \left(\frac{p_n}{p_{\text{th}}} \right)^2 \right) \approx \frac{L}{2\pi\hbar} \int_{-\infty}^{\infty} dp \exp \left(-\frac{1}{2} \left(\frac{p}{p_{\text{th}}} \right)^2 \right) = \frac{L}{2\pi\hbar} \sqrt{2\pi} p_{\text{th}}. \quad (3.2.4)$$

Altogether, we obtain a thermal state of a free particle,

$$\hat{\gamma}_{\text{free}} = \left(\frac{2\pi\hbar}{L} \right)^3 \mu(\hat{\mathbf{p}}), \quad (3.2.5)$$

where $\mu(\mathbf{p})$ is a Gaussian with zero mean and a standard deviation of p_{th} .

3.2.2. Reduction of the many-body problem

The $(N+1)$ -body interaction of system and gas is practically unsolvable. We approach this problem by deriving a hierarchy of equations, where the equation for the k -body problem requires the solution of the $k+1$ -body problem. In the low density limit, we then truncate the hierarchy and reduce the evolution of the system to a closed version of the two-body problem. A similar derivation can be found in [168].

In order to derive the hierarchy, we go into the interaction picture. With respect to this frame, the time evolution of the reduced system state reads as,

$$\begin{aligned} i\hbar \frac{d\hat{\rho}_{\text{S}}^{(\text{I})}(t)}{dt} &= \text{tr}_{1,\dots,N} \left\{ \left[\hat{V}^{(\text{I})}(t), \hat{\rho}_{\text{S},1,\dots,N}^{(\text{I})}(t) \right] \right\} = \sum_{j=1}^N \text{tr}_{1,\dots,N} \left\{ \left[\hat{V}_j^{(\text{I})}(t), \hat{\rho}_{\text{S},1,\dots,N}^{(\text{I})}(t) \right] \right\} \\ &= \sum_{j=1}^N \text{tr}_j \left\{ \left[\hat{V}_j^{(\text{I})}(t), \hat{\rho}_{\text{S},j}^{(\text{I})}(t) \right] \right\} = N \text{tr}_1 \left\{ \left[\hat{V}_1^{(\text{I})}(t), \hat{\rho}_{\text{S},1}^{(\text{I})}(t) \right] \right\}, \end{aligned} \quad (3.2.6)$$

where, in the first step, we used the linearity of the trace to expand in the two-body

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terms of the interaction. In the second step, we noted that only the j -th ancilla space is affected non-trivially by the two-body interaction $\hat{V}_j^{(I)}(t)$ such that the partial trace on $N - 1$ spaces could already be performed on the state. In the final step, we assume that, on average, our systems is affected by any gas particle in the same fashion. Hence, we can replace the sum by N times the expression with a single two-body term. Note that the equation is not closed since the evolution of the two-body state requires the three-body state, as we will see in the following.

The two-body state, evolves according to

$$\begin{aligned} i\hbar \frac{d\hat{\rho}_{S,1}^{(I)}(t)}{dt} &= \text{tr}_{2,\dots,N} \left\{ \left[\hat{V}^{(I)}(t), \hat{\rho}_{S,1,\dots,N}^{(I)}(t) \right] \right\} = \left[\hat{V}_1^{(I)}(t), \hat{\rho}_{S1}^{(I)}(t) \right] + \sum_{j=2}^N \text{tr}_j \left\{ \left[\hat{V}_j^{(I)}, \hat{\rho}_{S,1,j} \right] \right\} \\ &= \left[\hat{V}_1^{(I)}(t), \hat{\rho}_{S1}^{(I)}(t) \right] + (N-1) \text{tr}_2 \left\{ \left[\hat{V}_2^{(I)}, \hat{\rho}_{S,1,2}^{(I)} \right] \right\}, \end{aligned} \quad (3.2.7)$$

where the argumentation is similar to (3.2.6). A comparison of Equations (3.2.6) and (3.2.7) reveals the first two entries in a chain of equations of motion, where the evolution of the k -body correlation $\hat{\rho}_{S,1,\dots,k-1}^{(I)}$ is given in terms of the $k+1$ -body correlations $\hat{\rho}_{S,1,\dots,k}^{(I)}$. This chain of equations is known as the *BBGKY-hierarchy*² [77]. The full BBGKY hierarchy is an expedient way to express the exact $(N+1)$ -body problem, because it admits an approximate treatment by means of truncation in the regime of low density:

The second term in the last line of Equation (3.2.7) introduces a three-body interaction between system and two gas particles. Since the gas particles do not interact directly, their interaction is mediated by the system S . We will now study the scaling of this term and show that it can be neglected: If we assume the state to be roughly $\hat{\rho}_{S,1,2}^{(I)} \approx \hat{\rho}_{S,1}^{(I)} \otimes \hat{\gamma}_2$, we get a $1/L^3$ scaling from the thermal state, which yields the particle density $n = N/L^3$ as a prefactor. Hence, in the limit of a dilute gas, the second term in Equation (3.2.7) is suppressed. To be more precise, let us assume a characteristic length scale R of the interaction operators $\hat{V}_j$. Under the assumption that $N(R/L)^3 \ll 1$, it becomes very unlikely that more than one gas particle is in the interaction volume $\propto R^3$ around the system. Conclusively, the interactions between gas particles, mediated by the system, are negligible. The remaining term in Equation (3.2.7) and Equation (3.2.6) can be rephrased as the closed system of equations,

$$\frac{d\hat{\rho}_S^{(I)}(t)}{dt} = N \text{tr}_1 \left\{ \frac{d\hat{\rho}_{S,1}^{(I)}(t)}{dt} \right\} \quad (3.2.8)$$

$$\frac{d\hat{\rho}_{S,1}^{(I)}(t)}{dt} = -\frac{i}{\hbar} \left[\hat{V}_1^{(I)}(t), \hat{\rho}_{S,1}^{(I)}(t) \right] \quad (3.2.9)$$

This shows that the combined effect of the non-interacting scattering environment can be expressed as the effect of a single gas particle multiplied by the number of particles³.

²Named after Bogoliubov, Born, Green, Kirkwood and Yvon.

³Note that, especially in older literature [166], $N\hat{\gamma}_{\text{free}}$ is directly chosen as the "state" of the gas.

3.2.3. Scattering description

In Subsection 3.2.2, we learned that the N -body dynamics of our system S in a thermal gas can be reduced to the dynamics of the system with a single gas particle. However, note that the joint evolution of system and the representative gas particle over long periods of time will drive the gas representative out of equilibrium, which is a contradiction to the assumption of a thermal gas. This problem is circumvented by “resetting” the gas particle as a Gibbs state after short periods of time. This can consistently be implemented by the scattering approach that we will establish in this subsection.

In order to better describe the two-body problem, we slightly change the notation: The gas particle consists of a motional degree of freedom we denote with M and an internal degree of freedom, we call ancilla A . Since the effect of the environment is now effectively represented by the coupled dynamics of the system and this single gas particle, we call the motional and ancilla degree of freedom together "environment" $E = (A, M)$. The Hamiltonian becomes,

$$\hat{H} = \hat{h}_S + \hat{h}_A + \frac{\hat{\mathbf{p}}^2}{2m} + \hat{V}, \quad (3.2.10)$$

with the internal Hamiltonians $\hat{h}_{S/A}$, the kinetic energy of the gas particle $\hat{\mathbf{p}}^2/(2m)$ and the interaction Hamiltonian $\hat{V}$. The relevant equations from Subsection 3.2.2 are,

$$\frac{d\hat{\rho}_S^{(I)}(t)}{dt} = N \operatorname{tr}_E \left\{ \frac{d\hat{\rho}_{S,E}^{(I)}(t)}{dt} \right\} \quad (3.2.11)$$

$$\frac{d\hat{\rho}_{S,E}^{(I)}(t)}{dt} = \left[\hat{V}^{(I)}(t), \hat{\rho}_{SE}^{(I)}(t) \right]. \quad (3.2.12)$$

Integrating (3.2.11) on both sides from t to $t + \Delta t$ yields,

$$\hat{\rho}_S^{(I)}(t + \Delta t) - \hat{\rho}_S^{(I)}(t) = N \operatorname{tr}_E \left\{ \hat{\rho}_{SE}^{(I)}(t + \Delta t) - \hat{\rho}_{SE}^{(I)}(t) \right\} \quad (3.2.13)$$

The evolution of the combined state, (3.2.12), can be formally solved by the unitary evolution

$$\hat{\rho}_{SE}^{(I)}(t + \Delta t) = \hat{U}^{(I)}(t + \Delta t, t) \hat{\rho}_{SE}^{(I)}(t) \hat{U}^{(I)}(t + \Delta t, t), \quad (3.2.14)$$

as given by the time-ordered exponential

$$\hat{U}^{(I)}(t + \Delta t, t) = \mathcal{T} \left\{ \exp \left(-\frac{i}{\hbar} \int_t^{t+\Delta t} dt' \hat{V}^{(I)}(t') \right) \right\}. \quad (3.2.15)$$

We will now simplify the unitary by employing scattering theory. In order to do so, we assume the time scales of the free evolution t_S and of the scattering process t_E to be separated by several orders of magnitude, see discussion in Subsection 3.2.6. Then, our

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time resolution Δt can be chosen in such a way that,

$$t_S \gg \Delta t \gg t_E. \quad (3.2.16)$$

Since the interaction with the environment happens on much smaller time scales than Δt , we can assume that the interaction is completed within Δt , which allows us to expand the limits of integration,

$$\hat{U}^{(I)}(t + \Delta t, t) \approx \mathcal{T} \left\{ \exp \left(-\frac{i}{\hbar} \int_{-\infty}^{\infty} dt' \hat{V}^{(I)}(t') \right) \right\} = \hat{S}. \quad (3.2.17)$$

This is a crucial step in the derivation: The interaction with the environment is reduced to a single operator which simplifies the calculations significantly. However, we have to keep in mind that the infinite interaction time is just an approximation. We will later see that this will influence our treatment of unavoidable divergences. The evolution over time Δt now becomes,

$$\hat{\rho}_S^{(I)}(t + \Delta t) = \hat{\rho}_S^{(I)}(t) + N \operatorname{tr}_E \left\{ \hat{S} \hat{\rho}_{SE}(t) \hat{S}^\dagger - \hat{\rho}_{SE}(t) \right\}. \quad (3.2.18)$$

This can be brought into Lindblad form by using the usual decomposition $\hat{S} = \mathbb{1} + i\hat{T}$ of the scattering operator,

$$\hat{S} \hat{\rho}_{SE} \hat{S}^\dagger = (\mathbb{1} + i\hat{T}) \hat{\rho}_{SE} (\mathbb{1} + i\hat{T})^\dagger = \hat{\rho}_{SE} = \hat{\rho}_{SE} + \hat{T} \hat{\rho}_{SE} \hat{T}^\dagger + i (\hat{T} \hat{\rho}_{SE} - \hat{\rho}_{SE} \hat{T}^\dagger) \quad (3.2.19)$$

and then the optical theorem to see,

$$\begin{aligned} (\hat{T} \hat{\rho}_{SE} - \hat{\rho}_{SE} \hat{T}^\dagger) &= (\operatorname{Re} \{ \hat{T} \} + i\hat{T}^\dagger \hat{T}) \hat{\rho}_{SE} - \hat{\rho}_{SE} (\operatorname{Re} \{ \hat{T} \} - i\hat{T}^\dagger \hat{T}) \\ &= [\operatorname{Re} \{ \hat{T} \}, \hat{\rho}_{SE}] + i \{ \hat{T}^\dagger \hat{T}, \hat{\rho}_{SE} \}. \end{aligned} \quad (3.2.20)$$

Combining (3.2.19) and (3.2.20), we get,

$$\hat{S} \hat{\rho}_{SE} \hat{S}^\dagger - \hat{\rho}_{SE} = i [\operatorname{Re} \{ \hat{T} \}, \hat{\rho}_{SE}] + \mathcal{D} [\hat{T}] \hat{\rho}_{SE} = \mathcal{L} [\hat{T}] \hat{\rho}_{SE}, \quad (3.2.21)$$

where the superoperator $\mathcal{L}$ consists of the Hermitean part $i [\operatorname{Re} \{ \hat{T} \}, \cdot]$ and the dissipator $\mathcal{D} [\hat{T}] = \hat{T} \cdot \hat{T}^\dagger - \{ \hat{T}^\dagger \hat{T}, \cdot \} / 2$. With this, the equation of motion for the system degree of freedom becomes,

$$\hat{\rho}_S^{(I)}(t + \Delta t) - \hat{\rho}_S^{(I)}(t) = N \operatorname{tr}_E \{ \mathcal{L} \hat{\rho}_{SE}(t) \}. \quad (3.2.22)$$

Under the assumption that system and environment de-correlate sufficiently fast and that interaction picture and Schrödinger picture coincide at time t , we can approximate

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on the right hand side,

$$\hat{\rho}_{\text{SE}}^{(\text{I})}(t) \approx \hat{\rho}_{\text{S}}(t) \otimes \hat{\gamma}_{\text{E}}. \quad (3.2.23)$$

The two states on the right hand side are now in the Schrödinger picture. Finally, we can transform the left hand side back into Schrödinger picture and expand in Δt , since $\Delta t \ll t_{\text{s}}$. Neglecting terms of order $(\Delta t/t_{\text{free}})^2$ and higher and also neglecting a mixed term of free evolution and scattering yields

$$\hat{\rho}_{\text{S}}(t + \Delta t) \approx \hat{\rho}_{\text{S}}(t) - \frac{i}{\hbar} [\hat{h}_{\text{S}}, \hat{\rho}_{\text{S}}] \Delta t + N \text{tr}_{\text{E}} \left\{ \mathcal{L}[\hat{T}] (\hat{\rho}_{\text{S}}(t) \otimes \hat{\gamma}_{\text{E}}) \right\}. \quad (3.2.24)$$

From this equation for the coarse-grained evolution of $\hat{\rho}_{\text{S}}(t)$ over a time Δt , we will proceed in the following subsections to derive a master equation.

3.2.4. Trace over motion

The remaining step in our derivation is to bring (3.2.24) into Lindblad form by performing the trace in $\text{tr}_{\text{E}} \left\{ \mathcal{L}[\hat{T}] (\hat{\rho}_{\text{S}}(t) \otimes \hat{\gamma}_{\text{E}}) \right\}$. The environment consists of two parts $E = (A, M)$. We will begin with the motional degree of freedom M. For this step, we combine system S and ancilla A by $\hat{\rho}_{\text{S}} \otimes \hat{\gamma}_{\text{E}} = \hat{\rho}_{\text{SA}} \otimes \hat{\gamma}_{\text{M}}$ and label the energy eigenstates and eigenvectors with greek indices.

We start by taking the partial trace of the dissipator in the improper momentum basis and inserting the explicit form (3.2.5) of the motional Gibbs state,

$$\begin{aligned} & N \text{tr}_{\text{M}} \left\{ \mathcal{D}[\hat{T}] (\hat{\rho}_{\text{SA}}(t) \otimes \hat{\gamma}_{\text{M}}) \right\} \\ &= (2\pi\hbar)^3 \frac{N}{L^3} \int d^3\mathbf{q} \langle \mathbf{q} | \left(\hat{T} \hat{\rho}_{\text{SA}}(t) \otimes \mu(\hat{\mathbf{p}}) \hat{T}^\dagger - \frac{1}{2} \left\{ \hat{T}^\dagger \hat{T}, \hat{\rho}_{\text{SA}}(t) \otimes \mu(\hat{\mathbf{p}}) \right\} \right) | \mathbf{q} \rangle \\ &= (2\pi\hbar)^3 n \int d^3\mathbf{p} \mu(\mathbf{p}) \int d^3\mathbf{q} \mathcal{D} \left[\langle \mathbf{q} | \hat{T} | \mathbf{p} \rangle \right] \hat{\rho}_{\text{SA}}. \end{aligned} \quad (3.2.25)$$

Here, we introduce the particle density, $n = N/L^3$ in the third line. Expressing the T -operator by scattering amplitudes, as introduced in Subsection 1.2.4, we can write,

$$\langle \mathbf{q} | \hat{T} | \mathbf{p} \rangle = \frac{1}{2\pi\hbar m} \sum_{\nu, \kappa} \delta^{(1)}(E_\nu(\mathbf{q}) - E_\kappa(\mathbf{p})) f_{\nu\kappa}(\mathbf{q}, \mathbf{p}) |\nu\rangle \langle \kappa|, \quad (3.2.26)$$

where $E_\nu(\mathbf{q}) = \mathbf{q}^2/(2m) + \epsilon_\nu$ denotes the free energy associated with the state $|\mathbf{q}, \nu\rangle$. In principle, (3.2.25) is already a dissipator in Lindblad form. Carrying out the remaining integrals and the delta functions in the operators (3.2.26) will reveal the internal transitions induced by the dissipator. First, observe that each term of $\mathcal{D} \left[\langle \mathbf{q} | \hat{T} | \mathbf{p} \rangle \right] \hat{\rho}_{\text{SA}}$

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contains two delta functions. These can be phrased as

$$\begin{aligned}
& \delta^{(1)}(E_{\nu_2}(\mathbf{q}) - E_{\kappa_2}(\mathbf{p})) \delta^{(1)}(E_{\nu_1}(\mathbf{q}) - E_{\kappa_1}(\mathbf{p})) \\
&= \delta^{(1)}(\Delta_{\nu_1\kappa_1} - \Delta_{\nu_2\kappa_2}) \delta^{(1)}(E_{\nu_1}(\mathbf{q}) - E_{\kappa_1}(\mathbf{p})) \\
&= \frac{\Delta t}{2\pi\hbar} \delta^{(0)}(\Delta_{\nu_1\kappa_1} - \Delta_{\nu_2\kappa_2}) \frac{m}{|\mathbf{q}|} \delta^{(1)}\left(|\mathbf{q}| - \sqrt{\mathbf{p}^2 - 2m\Delta_{\nu_1\kappa_1}}\right), \tag{3.2.27}
\end{aligned}$$

where we used the second delta function to replace $\frac{\mathbf{q}^2}{2m} = \frac{\mathbf{p}^2}{2m} - \Delta_{\nu_1\kappa_1}$ with the internal energy difference $\Delta_{\nu\kappa} = \epsilon_\nu - \epsilon_\kappa$ in the first delta function and introduced $\delta^{(0)}$ for the Kronecker delta of non-integers. Note that $\delta^{(1)}(\Delta_{\nu_1\kappa_1} - \Delta_{\nu_2\kappa_2})$ is a delta function of values from a discrete set: If the argument is non-zero, the whole expression is zero, which coincides with the Kronecker delta. But, if $\Delta_{\nu_1\kappa_1} - \Delta_{\nu_2\kappa_2} = 0$, we obtain $\delta^{(1)}(0)$, which in general is ill-defined. In Subsection 1.2.6, we showed that this expression can be regularized in this case and we find $\delta^{(1)}(0) = \Delta t/(2\pi\hbar)$ and thus the scaled Kronecker delta,

$$\delta^{(1)}(\Delta_{\nu_1\kappa_1} - \Delta_{\nu_2\kappa_2}) = \frac{\Delta t}{2\pi\hbar} \delta^{(0)}(\Delta_{\nu_1\kappa_1} - \Delta_{\nu_2\kappa_2}). \tag{3.2.28}$$

Finally, we use a distributional identity from Appendix C.2.1 to rewrite the second delta function. This completes the calculation of (3.2.27).

Let us now insert (3.2.27) into (3.2.25) and carry out the integral over $\mathbf{q}$,

$$\begin{aligned}
& \int d^3\mathbf{q} \langle \mathbf{q} | \hat{T} | \mathbf{p} \rangle \hat{\rho}_{\text{SA}} \langle \mathbf{p} | \hat{T}^\dagger | \mathbf{q} \rangle = \frac{1}{(2\pi\hbar)^3} \frac{\Delta t}{m} \int d\Omega \sum_{\substack{\Delta_{\nu_1\kappa_1} = \Delta_{\nu_2\kappa_2} \\ \nu_1, \kappa_1, \nu_2, \kappa_2}} \\
& P_{\nu_1\kappa_1}(\mathbf{p}, \Omega) f_{\nu_1\kappa_1}(\mathbf{P}_{\nu_1\kappa_1}(\mathbf{p}, \Omega), \mathbf{p}) |\nu_1\rangle \langle \kappa_1| \hat{\rho}_{\text{SA}} |\kappa_2\rangle \langle \nu_2| f_{\nu_2\kappa_2}^*(\mathbf{P}_{\nu_2\kappa_2}(\mathbf{p}, \Omega), \mathbf{p}) \\
&= \frac{1}{(2\pi\hbar)^3} \frac{p\Delta t}{m} \int d\Omega \sum_E \left(\sum_{\nu_1, \kappa_1}^{\Delta_{\nu_1, \kappa_1} = E} \sqrt{\frac{P_{\nu_1\kappa_1}(\mathbf{p}, \Omega)}{p}} f_{\nu_1\kappa_1}(\mathbf{P}_{\nu_1\kappa_1}(\mathbf{p}, \Omega), \mathbf{p}) |\nu_1\rangle \langle \kappa_1| \right) \\
& \hat{\rho}_{\text{SA}} \left(\sum_{\nu_2, \kappa_2}^{\Delta_{\nu_2, \kappa_2} = E} \sqrt{\frac{P_{\nu_2\kappa_2}(\mathbf{p}, \Omega)}{p}} f_{\nu_2\kappa_2}(\mathbf{P}_{\nu_2\kappa_2}(\mathbf{p}, \Omega), \mathbf{p}) |\nu_2\rangle \langle \kappa_2| \right)^\dagger \\
&= \frac{1}{(2\pi\hbar)^3} \frac{p\Delta t}{m} \int d\Omega \sum_E \hat{L}_E(\mathbf{p}, \Omega) \hat{\rho}_{\text{SA}} \hat{L}_E^\dagger(\mathbf{p}, \Omega), \tag{3.2.29}
\end{aligned}$$

where, we split the condition $\Delta_{\nu_1, \kappa_1} = \Delta_{\nu_2, \kappa_2}$ by using

$$\begin{aligned}
& \sum_{E \in \mathbb{R}} \delta^{(0)}(\Delta_{\nu_1, \kappa_1} - E) \delta^{(0)}(\Delta_{\nu_2, \kappa_2} - E) = \delta^{(0)}(\Delta_{\nu_2, \kappa_2} - \Delta_{\nu_1, \kappa_1}) \sum_E \delta^{(0)}(\Delta_{\nu_1, \kappa_1} - E) \\
&= \delta^{(0)}(\Delta_{\nu_2, \kappa_2} - \Delta_{\nu_1, \kappa_1}), \tag{3.2.30}
\end{aligned}$$

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and multiplied the expression by p/p . Finally, we introduced the Lindblad operator

$$\hat{L}_E(\mathbf{p}, \Omega) = \sum_{\nu, \kappa}^{\Delta_{\nu, \kappa} = E} \sqrt{\frac{P_{\nu\kappa}(\mathbf{p}, \Omega)}{p}} f_{\nu\kappa}(\mathbf{P}_{\nu\kappa}(\mathbf{p}, \Omega), \mathbf{p}) |\nu\rangle \langle \kappa|. \quad (3.2.31)$$

With this we have simplified the first term of the dissipator $\mathcal{D}[\hat{T}]$. The remaining terms are simplified in the same manner, as done in App. C.2.2,

$$\int d^3\mathbf{q} \left\{ \langle \mathbf{q} | \hat{T} | \mathbf{p} \rangle, \hat{\rho}_{\text{SA}} \right\} = \frac{1}{(2\pi\hbar)^3} \frac{p\Delta t}{m} \int d\Omega \sum_E \left\{ \hat{L}_E^\dagger(\mathbf{p}, \Omega) \hat{L}_E(\mathbf{p}, \Omega), \hat{\rho}_{\text{SA}} \right\}. \quad (3.2.32)$$

In total, we are left with

$$\begin{aligned} & N \text{tr}_M \left\{ \mathcal{D}[\hat{T}] (\hat{\rho}_{\text{SA}}(t) \otimes \hat{\gamma}_M) \right\} \\ &= \Delta t \int d^3\mathbf{p} \mu(\mathbf{p}) |\mathbf{j}(\mathbf{p})| \int d\Omega \sum_E \mathcal{D} \left[\hat{L}_E(\mathbf{p}, \Omega) \right] \hat{\rho}_{\text{SA}}, \end{aligned} \quad (3.2.33)$$

with $\mathbf{j}(\mathbf{p}) = n\mathbf{p}/m$ the particle flux density.

Besides the dissipator, we also obtain a contribution to the Hamiltonian that originates from the term

$$\begin{aligned} & N \text{tr}_M \left\{ i \left[\text{Re} \left\{ \hat{T} \right\}, \hat{\rho}_{\text{SA}} \otimes \hat{\gamma}_M \right] \right\} \\ &= i(2\pi\hbar)^3 \frac{N}{L^3} \int d^3\mathbf{p} \langle \mathbf{p} | \left[\text{Re}\{\hat{T}\}, \hat{\rho}_{\text{SA}} \otimes \mu(\hat{\mathbf{p}}) \right] | \mathbf{p} \rangle \\ &= i(2\pi\hbar)^3 n \int d^3\mathbf{p} \mu(\mathbf{p}) \left[\langle \mathbf{p} | \text{Re}\{\hat{T}\} | \mathbf{p} \rangle, \hat{\rho}_{\text{SA}} \right], \end{aligned} \quad (3.2.34)$$

which only involves the diagonal momentum matrix elements of the T -operator. They read as,

$$\begin{aligned} \langle \mathbf{p} | \hat{T} | \mathbf{p} \rangle &= \frac{1}{2\pi\hbar m} \sum_{\nu, \kappa} \delta^{(1)}(\Delta_{\nu\kappa}) f_{\nu\kappa}(\mathbf{p}, \mathbf{p}) |\nu\rangle \langle \kappa| \\ &= \frac{\Delta t}{(2\pi\hbar)^2 m} \sum_{\nu, \kappa} \delta^{(0)}(\Delta_{\nu\kappa}) f_{\nu\kappa}(\mathbf{p}, \mathbf{p}) |\nu\rangle \langle \kappa|. \end{aligned} \quad (3.2.35)$$

Once again, we made use of the delta regularization replacing the delta function by a rescaled Kronecker delta, $\delta^{(1)}(\Delta_{\nu\kappa}) = \Delta t / (2\pi\hbar) \delta^{(0)}(\Delta_{\nu\kappa})$, in the same fashion as in (3.2.28). The real part of the T -operator matrix elements reads,

$$\langle \mathbf{p} | \text{Re}\{\hat{T}\} | \mathbf{p} \rangle = \frac{\Delta t}{(2\pi\hbar)^2 m} \sum_{\nu, \kappa}^{\epsilon_\nu = \epsilon_\kappa} \frac{1}{2} (f_{\nu\kappa}(\mathbf{p}, \mathbf{p}) + f_{\kappa\nu}^*(\mathbf{p}, \mathbf{p})) |\nu\rangle \langle \kappa|. \quad (3.2.36)$$

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Hence, we can identify the effective Hamiltonian,

$$\hat{h}_{\text{eff,SA}} = -\frac{2\pi\hbar^2}{m}n \int d^3\mathbf{p} \mu(\mathbf{p}) \sum_{\nu,\kappa}^{\epsilon_\nu=\epsilon_\kappa} \frac{1}{2} (f_{\nu,\kappa}(\mathbf{p}, \mathbf{p}) + f_{\kappa,\nu}^*(\mathbf{p}, \mathbf{p})) |\nu\rangle \langle \kappa|. \quad (3.2.37)$$

Putting everything together, the coarse-grained evolution of System and Ancilla over the time interval $[t, t + \Delta t]$ is,

$$\begin{aligned} N \text{tr}_M \left\{ \mathcal{L}[\hat{T}] (\hat{\rho}_{\text{SA}}(t) \otimes \hat{\gamma}_M) \right\} = & -\frac{i}{\hbar} [\hat{h}_{\text{eff}}, \hat{\rho}_{\text{SA}}] \Delta t \\ & + \int d^3\mathbf{p} \mu(\mathbf{p}) |\mathbf{j}(\mathbf{p})| \int d\Omega \sum_E \mathcal{D} [\hat{L}_E(\mathbf{p}, \Omega)] \hat{\rho}_{\text{SA}} \Delta t. \end{aligned} \quad (3.2.38)$$

3.2.5. Trace over the ancilla and discussion of the master equation

The last subsection concluded with an expression for the joint evolution of system and ancilla $\hat{\rho}_{\text{SA}}(t + \Delta t) - \hat{\rho}_{\text{SA}}(t)$ in a time interval $[t, t + \Delta t]$ due to the motional degree of freedom of the gas. Now, we will trace out the ancilla. To this end, we assume $\hat{\rho}_{\text{SA}}(t) \approx \hat{\rho}_S(t) \otimes \hat{\gamma}_A$ at the beginning of the time step. This is fulfilled because we consider a new gas particle in any time step Δt .

In order to better distinguish indices referring to the system and indices referring to the ancilla, we introduce the following notation: greek letters denoting combined states of system and ancilla are to be understood as multi indices $\nu = (n_S, n_A)$, meaning $|\nu\rangle = |n_S, n_A\rangle = |n_S\rangle_S |n_A\rangle_A$. To further improve readability, we will use system indices as subscripts and ancilla indices as superscripts, e.g.

$$\begin{aligned} \epsilon_\nu &= \epsilon_{n_S} + \epsilon^{n_A} \\ f_{\nu\kappa}(\mathbf{p}, \mathbf{q}) &= f_{n_S, k_S}^{n_A, k_A}(\mathbf{p}, \mathbf{q}) \\ \mathbf{P}_{\nu\kappa}(\Omega, \mathbf{p}) &= \mathbf{P}_{n_S, k_S}^{n_A, k_A}(\Omega, \mathbf{p}) \\ &\vdots \end{aligned} \quad (3.2.39)$$

and so on. We define the populations of the ancilla Gibbs state to be p^{n_A} such that $\langle n_A | \hat{\gamma}_A | k_A \rangle = p^{n_A} \delta_{n_A, k_A}$. The straightforward but tedious process of taking the partial trace over the ancilla degrees of freedom is presented in Appendix C.2.3. After adding the free Hamiltonian $\hat{h}_S$ and dividing by Δt , the resulting master equation becomes,

$$\begin{aligned} \frac{d}{dt} \hat{\rho}_S \approx & \frac{\hat{\rho}_S(t + \Delta t) - \hat{\rho}_S(t)}{\Delta t} \approx -\frac{i}{\hbar} [\hat{h}_S + \hat{h}_{\text{eff}}, \hat{\rho}_S(t)] \\ & + \sum_{n_A, k_A} p^{k_A} \int d^3\mathbf{p} \mu(\mathbf{p}) |\mathbf{j}(\mathbf{p})| \int d\Omega \sum_E \mathcal{D} [\hat{L}_E^{n_A, k_A}(\mathbf{p}, \Omega)] \hat{\rho}_S(t), \end{aligned} \quad (3.2.40)$$

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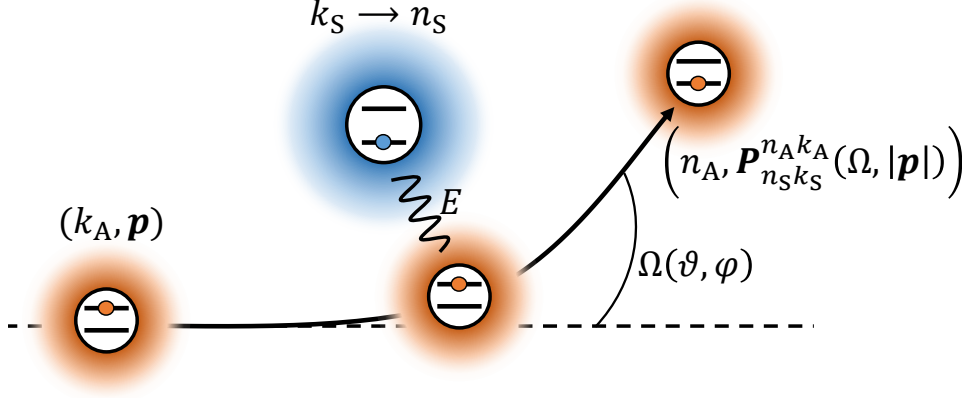


Figure 3.2.: Sketch of a system-ancilla scattering event and relevant parameters. Here we focus on a transition of the system degrees of freedom, $k_S \rightarrow n_S$, induced by the Lindblad operator $\hat{L}_E^{n_A, k_A}(\Omega, \mathbf{p})$: Consider the two-body interaction between system (blue) and gas particle/environment (orange). Initially, the incoming gas particle has momentum $\mathbf{p}$ and is in the internal state labeled by k_A . In the interaction, only transitions that change the internal energy of the system by $\epsilon_{n_S} - \epsilon_{k_S} = E$ are considered by this specific Lindblad operator. Finally, the gas particle leaves in the direction specified by the solid angle Ω and in the internal state n_A . Note that the length of the final momentum is already determined by energy conservation.

where,

$$\hat{h}_{\text{eff}} = -\frac{2\pi\hbar^2}{m}n \sum_{k_A} p^{k_A} \int d^3\mathbf{p} \mu(\mathbf{p}) \sum_{n_S, k_S}^{\epsilon_{n_S}=\epsilon_{k_S}} \frac{1}{2} \left(f_{n_S, k_S}^{k_A, k_A}(\mathbf{p}, \mathbf{p}) + f_{k_S, n_S}^{k_A, k_A^*}(\mathbf{p}, \mathbf{p}) \right) |n_S\rangle \langle k_S| \quad (3.2.41)$$

$$\hat{L}_E^{n_A, k_A}(\mathbf{p}, \Omega) = \sum_{n_S, k_S}^{\Delta_{n_S, k_S}=E} \sqrt{\frac{P_{n_S, k_S}^{n_A, k_A}(p)}{p}} f_{n_S, k_S}^{n_A, k_A} \left(\mathbf{P}_{n_S, k_S}^{n_A, k_A}(\Omega, p), \mathbf{p} \right) |n_S\rangle \langle k_S|. \quad (3.2.42)$$

Recall that Δt in (3.2.40) must be greater than the duration of a scattering event but sufficiently small with respect to system time scales to justify approximating the time derivative in (3.2.40) by the finite difference. While treating the difference quotient as a derivative is a reasonable approximation, dynamics on shorter time scales than the scattering events are not resolved properly.

Before we conclude this subsection, we shortly discuss the master equation. The five indices labeling the Lindblad operators are depicted in Fig. 3.2: Recall that the interaction with the environment reduces to the two-body interaction between the system and a gas particle. This interaction is described by scattering theory, where the initial state of the gas particle is described by the incoming momentum $\mathbf{p}$ and internal state k_A . The post scattering state of the gas particle, on the other hand, is given in terms of the direction Ω in which the particle leaves with respect to the incoming direction and

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the internal state n_A . Finally, E describes the type of transitions that can occur: The Lindblad operator $\hat{L}_E^{n_A, k_A}(\Omega, \mathbf{p})$ induces only those transitions $n_S \leftarrow k_S$ of the system that fulfill $E = \epsilon_{n_S} - \epsilon_{k_S}$. A comparison with Subsection 1.2.4 shows that the strength with which the interaction occurs is linked to the differential cross section,

$$\hat{L}_E^{n_A, k_A}(\Omega, \mathbf{p}) = \sum_{n_S, k_S}^{\Delta_{n_S, k_S} = E} \sqrt{\frac{d\sigma_{n_S, k_S}^{n_A, k_A}(\mathbf{p})}{d\Omega}} |n_S\rangle \langle k_S|, \quad (3.2.43)$$

where the square root has to be treated with caution since the scattering amplitude is in general complex. For the intuitive understanding of the master equation, however, (3.2.43) is very helpful. The differential cross section is a measure of how likely a transition $(n_S, n_A, \Omega) \leftarrow (k_S, k_A, \mathbf{p})$ occurs. The greater this quantity, the stronger the corresponding transition in our system degrees of freedom.

For the sake of readability, we will from now on write,

$$\frac{d}{dt} \hat{\rho}_S(t) = -\frac{i}{\hbar} [\hat{h}_S + \hat{h}_{\text{eff}}, \hat{\rho}_S(t)] + \mathcal{C} \hat{\rho}_S(t), \quad (3.2.44)$$

where $\mathcal{C}$ denotes the full dissipator from (3.2.40).

3.2.6. Discussion of time scales

In the preceding derivation of a the low density master equation, the assumption of “fast scattering events” was crucial. In fact, we had to assume that there is a coarse-grained time scale Δt that is much larger than the scattering time and much shorter than the free evolution. Here, we will estimate the time scales and give the assumption a clear meaning.

Given a characteristic length R of the interaction potential, we approximate the scattering time t_E by the time a thermal gas particle, with momentum $p_{\text{th}} = \sqrt{mk_B T}$, needs to cross the interaction zone,

$$t_E = 2R \sqrt{\frac{m}{k_B T}}. \quad (3.2.45)$$

With the maximal frequency of the system ω_S , the accumulated phase of the system should be negligible over the time period of a scattering event, $\omega_S t_E \ll 1$. In terms of the characteristic energy of the interaction,

$$E_R = \frac{1}{2m} \left(\frac{\hbar}{R} \right)^2, \quad (3.2.46)$$

the condition can be rephrased as,

$$\hbar \omega_S \ll \sqrt{E_R k_B T}. \quad (3.2.47)$$

Under this condition the scattering theory can be applied, as shown in Subsection 3.2.3.

3.3. Thermodynamic consistency

With the master equation (3.2.44) describing the effect of a scattering environment at hand, we discuss its applicability to thermodynamic problems. To this end, we first consider the laws of thermodynamics, presented in Subsection 1.4.1, discuss their precise meaning in this microscopic dynamical setting, and demonstrate their validity.

In Subsection 3.3.1, we show that the zeroth law applies in the sense that the Gibbs state representing thermal equilibrium with the environment is a fix point of the master equation and that the master equation fulfills the third law, i.e. the unattainability principle. Next in Subsection 3.3.2, we identify the rate of heat flow per unit time from the environment and show that energy conservation holds. We also prove that the second law holds in the form of the Clausius inequality.

3.3.1. The zeroth and the third law

The zeroth law of thermodynamics states that being in thermal equilibrium is an equivalence relation, as explained in Subsection 1.4.1. This implies that, provided the system equilibrates, the system should end up in a Gibbs state at the temperature of the gas. Hence,

$$\hat{\rho}_S(t) \xrightarrow[t \rightarrow \infty]{} \frac{1}{Z} \exp \left(-\frac{\hat{h}_S}{k_B T} \right) = \hat{\gamma}_S. \quad (3.3.1)$$

In practice, it strongly depends on the interaction $\hat{V}$ whether the system thermalizes or not. For the case that $\hat{h}_S$ is non-degenerate, however, we can show that $\hat{\gamma}_S$ is a steady state of the open system evolution: A non-degenerate Hamiltonian simplifies the calculation because $\epsilon_{n_S} = \epsilon_{k_S}$ is equivalent to $n_S = k_S$. Due to this fact, the effective Hamiltonian becomes diagonal,

$$\hat{h}_{\text{eff}} = -\frac{2\pi\hbar^2}{m} n \sum_{k_A} p^{k_A} \int d^3\mathbf{p} \mu(\mathbf{p}) \sum_{k_S} \text{Re} \left\{ f_{k_S, k_S}^{k_A, k_A}(\mathbf{p}, \mathbf{p}) \right\} |k_S\rangle \langle k_S|, \quad (3.3.2)$$

which only shifts the existing energy levels but does not introduce transitions between energy levels. Similar conclusions can be drawn for the dissipator, for which direct calculation yields,

$$\langle n_{S1} | \mathcal{D} \left[\hat{L}_E^{n_A k_A} \right] (|k_S\rangle \langle k_S|) | n_{S2} \rangle \propto \delta_{n_{S1} n_{S2}} \quad (3.3.3)$$

$$\langle n_S | \mathcal{D} \left[\hat{L}_E^{n_A k_A} \right] (|k_{S1}\rangle \langle k_{S2}|) | n_S \rangle \propto \delta_{k_{S1} k_{S2}}. \quad (3.3.4)$$

Hence, transitions $|k_S\rangle \langle k_S| \longleftrightarrow |n_{S1}\rangle \langle n_{S2}|$ occur if and only if $n_{S1} = n_{S2}$. Conclusively, the diagonal elements evolve independently from the non-diagonal elements and vice versa.

Now, assuming a diagonal state $\hat{\rho}_S(t) = \sum_{k_S} p_{k_S}(t) |k_S\rangle \langle k_S|$, we find a classical master

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equation for the energy level populations,

$$\dot{p}_{n_S}(t) = \langle n_S | \mathcal{C}(\hat{\rho}_S) | n_S \rangle = \sum_{k_S} \underbrace{\langle n_S | \mathcal{C}(|k_S\rangle \langle k_S|) | n_S \rangle}_{=M_{n_S k_S}} p_{k_S}(t), \quad (3.3.5)$$

where the unitary evolution does not contribute because $\hat{\rho}_S$ commutes with $\hat{h}_S$ and $\hat{h}_{\text{eff}}$, and we defined the transition matrix

$$\begin{aligned} M_{n_S k_S} &= \sum_{n_A, k_A} p^{k_A} \int d^3 \mathbf{p} \mu(\mathbf{p}) |\mathbf{j}(\mathbf{p})| \int d\Omega \sum_E \langle n_S | \mathcal{D} \left[\hat{L}_E^{n_A k_A}(\mathbf{p}, \Omega) \right] (|k_S\rangle \langle k_S|) | n_S \rangle. \end{aligned} \quad (3.3.6)$$

This matrix can be expressed in terms of the differential cross section by noting

$$\begin{aligned} &\sum_E \langle n_S | \mathcal{D} \left[\hat{L}_E^{n_A k_A}(\mathbf{p}, \Omega) \right] (|k_S\rangle \langle k_S|) | n_S \rangle \\ &= \sum_E \left(\left| \langle n_S | \hat{L}_E^{n_A k_A}(\mathbf{p}, \Omega) | k_S \rangle \right|^2 - \delta_{n_S k_S} \sum_{l_S} \left| \langle l_S | \hat{L}_E^{n_A k_A}(\mathbf{p}, \Omega) | n_S \rangle \right|^2 \right) \\ &= \frac{d\sigma_{n_S k_S}^{n_A k_A}(\Omega, \mathbf{p})}{d\Omega} - \delta_{n_S k_S} \sum_{l_S} \frac{d\sigma_{l_S n_S}^{n_A k_A}(\Omega, \mathbf{p})}{d\Omega}. \end{aligned} \quad (3.3.7)$$

Due to the linearity of integration, we can integrate term by term and find

$$\begin{aligned} &\sum_{n_A, k_A} p^{k_A} \int d^3 \mathbf{p} \mu(\mathbf{p}) |\mathbf{j}(\mathbf{p})| \int d\Omega \frac{d\sigma_{n_S k_S}^{n_A k_A}(\Omega, \mathbf{p})}{d\Omega} \\ &= \sum_{n_A, k_A} p^{k_A} \int d^3 \mathbf{p} \mu(\mathbf{p}) |\mathbf{j}(\mathbf{p})| \sigma_{n_S k_S}^{n_A k_A}(\mathbf{p}) = \sum_{n_A, k_A} p^{k_A} \mathbb{E} \left[|\mathbf{j}| \sigma_{n_S k_S}^{n_A k_A} \right] \\ &= \gamma_{n_S k_S}, \end{aligned} \quad (3.3.8)$$

where we rewrote the momentum integration as an expectation value with respect to $\mu(\mathbf{p})$, and defined these rates as $\gamma_{n_S n_S}$. This reveals the typical probability-preserving form of a Markov chain,

$$\dot{p}_{n_S}(t) = \sum_{k_S} M_{n_S k_S} p_{k_S}(t) = \sum_{k_S} (\gamma_{n_S k_S} p_{k_S}(t) - \gamma_{k_S n_S} p_{n_S}(t)), \quad (3.3.9)$$

in which the first term describes the flow of probability from all states k_S into the state of consideration n_S and the second term describes the total flow out of state n_S .

In order to show the local detailed balance property of the rates $\gamma_{n_S k_S}$, we assume system and gas to fulfill micro reversibility (see Subsection 1.2.4). On the level of scattering amplitudes, this symmetry reads $f_{n_S, k_S}^{n_A, k_A}(\mathbf{q}, \mathbf{p}) = f_{k_S, n_S}^{k_A, n_A}(-\mathbf{p}, -\mathbf{q})$. We employ

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this by expressing the rates by the scattering amplitudes,

$$\begin{aligned}
\gamma_{n_S k_S} &= n \sum_{n_A k_A} p^{k_A} \int d^3 \mathbf{q} \int d^3 \mathbf{p} \mu(\mathbf{p}) \delta^{(1)} \left(\frac{\mathbf{q}^2}{2m} - \left[\frac{\mathbf{p}^2}{2m} - \Delta_{n_S, k_S}^{n_A, k_A} \right] \right) |f_{n_S, k_S}^{n_A, k_A}(\mathbf{q}, \mathbf{p})|^2 \\
&= n \sum_{n_A k_A} p^{k_A} \int d^3 \mathbf{q} \int d^3 \mathbf{p} \mu(\mathbf{p}) \delta^{(1)} \left(\frac{\mathbf{p}^2}{2m} - \left[\frac{\mathbf{q}^2}{2m} - \Delta_{k_S, n_S}^{k_A, n_A} \right] \right) |f_{k_S, n_S}^{k_A, n_A}(\mathbf{p}, \mathbf{q})|^2 \\
&= n \sum_{n_A k_A} e^{\beta \Delta_{n_A, k_A}^{n_A, k_A}} p^{n_A} \int d^3 \mathbf{q} \int d^3 \mathbf{p} \mu(\mathbf{q}) e^{\beta \Delta_{k_S, n_S}^{k_A, n_A}} \\
&\quad \delta^{(1)} \left(\frac{\mathbf{p}^2}{2m} - \left[\frac{\mathbf{q}^2}{2m} - \Delta_{k_S, n_S}^{k_A, n_A} \right] \right) |f_{k_S, n_S}^{k_A, n_A}(\mathbf{p}, \mathbf{q})|^2 \\
&= e^{\beta \Delta_{k_S, n_S}} \gamma_{k_S n_S},
\end{aligned} \tag{3.3.10}$$

where we first use micro-reversibility to flip the order of the system and ancilla level indices. Since $\mathbf{p}$ and $\mathbf{q}$ are integrated over all directions, the extra signs can be absorbed. In the same step, we also used $\Delta_{n_S, k_S}^{n_A, k_A} = -\Delta_{k_S, n_S}^{k_A, n_A}$ to rewrite the delta function. Second, we used $p^{k_A}/p^{n_A} = e^{\beta \Delta_{n_A, k_A}^{n_A, k_A}}$ and the delta function to replace the argument of the Maxwell-Boltzmann distribution $\mu(\mathbf{p})$ of the gas,

$$\mu(\mathbf{p}) \delta^{(1)} \left(\frac{\mathbf{p}^2}{2m} - \left[\frac{\mathbf{q}^2}{2m} - \Delta_{k_S, n_S}^{k_A, n_A} \right] \right) = e^{\beta \Delta_{k_S, n_S}^{k_A, n_A}} \mu(\mathbf{q}) \delta^{(1)} \left(\frac{\mathbf{p}^2}{2m} - \left[\frac{\mathbf{q}^2}{2m} - \Delta_{k_S, n_S}^{k_A, n_A} \right] \right).$$

In the third step we noted $e^{\beta \Delta_{n_A, k_A}^{n_A, k_A}} e^{\beta \Delta_{k_S, n_S}^{k_A, n_A}} = e^{\beta \Delta_{k_S, n_S}}$ and recognized the formula for the rate in the opposite direction. This shows that the classical master equation for the populations of a mixed state fulfills the local detailed balance condition

$$\gamma_{n_S k_S} e^{-\beta \epsilon_{k_S}} = \gamma_{k_S n_S} e^{-\beta \epsilon_{n_S}}. \tag{3.3.11}$$

If we assume our system to be in the thermal state $\hat{\gamma}_S = \exp(-\beta \hat{h}_S)/Z$, which has the populations $p_k \propto e^{-\beta \epsilon_k}$, the evolution in this state according to (3.3.9) is

$$\dot{p}_n \propto \sum_k \underbrace{(\gamma_{nk} e^{-\beta \epsilon_k} - \gamma_{kn} e^{-\beta \epsilon_n})}_{=0} = 0. \tag{3.3.12}$$

Here, local detailed balance (3.3.11) ensures that the in-flow and out-flow of probability cancel for each pair of energy levels individually. Hence, the populations of the Gibbs state do not change under the master equation. On operator level, this reads,

$$\mathcal{C} \hat{\gamma}_S = 0. \tag{3.3.13}$$

In summary, we have derived the rates for the classical master equation, describing the evolution of the populations of a diagonal state, shown that the rates fulfill the local detailed balance condition if system and gas fulfill micro reversibility. This shows

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that the Gibbs state with respect to $\hat{h}_S$ and the gas temperature T is a steady state of the dynamics⁴. As discussed above, this shows compatibility with the zeroth law of thermodynamics.

From this result, we can also conclude that the master equation is consistent with the third law of thermodynamics: In the form of the unattainability principle, the third law states that the system can not be cooled down to absolute zero in a finite amount of time. If we now wanted the system to thermalize at $T = 0$ via the master equation, we would necessarily have to reduce the temperature of the gas to absolute zero as well. Because the system evolves dynamically in response to the gas, its temperature decreases more slowly. Therefore, the system will not reach absolute zero in finite time unless the gas does too. Hence, as long as we consider boundary conditions of the gas in accordance with the third law, the master equation is consistent with the third law, as well.

3.3.2. The first and second law

The first and second law state limitations for possible changes in energy. We will now show that the thermal gas environment is compatible with these two concepts.

From our microscopic description it follows immediately that every collision is energy preserving because the scattering operator $\hat{S}$ commutes with the free Hamiltonian of system, ancilla, and motional degree of freedom. This already shows,

$$\Delta E_S + \Delta E_A + \Delta E_M = 0, \quad (3.3.14)$$

where

$$\Delta E_{S/A} = \text{tr} \left\{ \hat{h}_{S/A} \left(\hat{S} \hat{\rho} \hat{S}^\dagger - \hat{\rho} \right) \right\}, \quad (3.3.15)$$

$$\Delta E_M = \text{tr} \left\{ \frac{\hat{\mathbf{p}}^2}{2m} \left(\hat{S} \hat{\rho} \hat{S}^\dagger - \hat{\rho} \right) \right\}. \quad (3.3.16)$$

It is now tempting to see ancilla and motional degree of freedom as two individual thermal baths and define the heats $Q_{A/M}$ as the negative change of the corresponding energies $-\Delta E_{A/M}$. This definition, however, ignores the energy flow between ancilla and motion. Specifically in this case, any collision consists of a three-body interaction, which makes the distinction between heat from the ancilla and heat from the motion impossible. Hence, we have to consider a single thermal environment consisting of ancilla and motion. Defining the internal energy $U_S = \text{tr} \left\{ \hat{h}_S \hat{\rho} \right\}$ and $Q = -(\Delta E_A + \Delta E_M)$ we obtain the first law of thermodynamics,

$$\Delta U_S = Q \quad (3.3.17)$$

for every single scattering event. On the level of the master equation, we find the

⁴Note that the Gibbs state does not include the “Lambd-shift”-term, $\hat{h}_{\text{eff}}$.

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continuous version,

$$\frac{d}{dt}U_S(t) = \frac{d}{dt} \left\{ \hat{h}_S \hat{\rho}_S(t) \right\} = \text{tr} \left\{ \hat{h}_S \mathcal{C} \hat{\rho}_S(t) \right\} = \dot{Q}(t), \quad (3.3.18)$$

where we used the master equation (3.2.44) and $[\hat{h}_{\text{eff}}, \hat{h}_S] = 0^5$.

We will now show that the second law holds in the form of a Clausius inequality for the rates of entropy change and heat flow,

$$\frac{d}{dt} \mathcal{S} [\hat{\rho}_S] \geq \beta \dot{Q}. \quad (3.3.19)$$

Here, we employ the von Neumann entropy, introduced in Subsection 1.4.2,

$$\mathcal{S} [\hat{\rho}_S] = -\text{tr} \{ \hat{\rho}_S \log (\hat{\rho}_S) \}. \quad (3.3.20)$$

With the given preliminaries, we can directly use results from [14, 169] in order to show the statement (3.3.19). For the sake of completeness, however, let us repeat the arguments here. As a prerequisite, we need the *relative entropy* between two states $\hat{\rho}$ and $\hat{\sigma}$

$$\mathcal{S} [\hat{\rho} || \hat{\sigma}] = \text{tr} \{ \hat{\rho} (\log (\hat{\rho}) - \log (\hat{\sigma})) \} = -\mathcal{S} [\hat{\rho}] - \text{tr} \{ \hat{\rho} \log (\hat{\sigma}) \}, \quad (3.3.21)$$

which is contractive in the sense that,

$$\mathcal{S} [\Lambda[\hat{\rho}] || \Lambda[\hat{\sigma}]] \leq \mathcal{S} [\hat{\rho} || \hat{\sigma}] \quad (3.3.22)$$

for any channel Λ [170]. Now let $\Lambda_{\Delta t}$ denote the channel that evolves our system state over a time Δt with respect to the master equation, i.e. $\Lambda_{\Delta t}[\hat{\rho}_S(t)] = \hat{\rho}_S(t + \Delta t)$. Using the fact that the Gibbs state $\hat{\rho}_S$ is a steady state of the channel, i.e. $\Lambda_{\Delta t}[\hat{\rho}_S] = \hat{\rho}_S$, the contractivity yields

$$\mathcal{S} [\hat{\rho}_S(t + \Delta t) || \hat{\rho}_S] \leq \mathcal{S} [\hat{\rho}_S(t) || \hat{\rho}_S] \quad (3.3.23)$$

and hence in the limit $\Delta t \rightarrow 0$,

$$\frac{d}{dt} \mathcal{S} [\hat{\rho}_S(t) || \hat{\rho}_S] \leq 0. \quad (3.3.24)$$

From this and the definition (3.3.21), it follows that,

$$\frac{d}{dt} \mathcal{S} [\hat{\rho}_S] \geq -\text{tr} \{ (\mathcal{C} \hat{\rho}_S) \log (\hat{\rho}_S) \}, \quad (3.3.25)$$

where we expressed the derivative of $\hat{\rho}_S$ by the master equation (3.2.44) and used that

⁵Notice that this is true even in the degenerate case: $\hat{h}_S \hat{h}_{\text{eff}} = \sum_{j,k}^{\epsilon_k = \epsilon_l} \epsilon_j e_{j,k} |j\rangle \langle k| = \sum_{j,k}^{\epsilon_k = \epsilon_l} e_{j,k} \epsilon_k |j\rangle \langle k| = \hat{h}_{\text{eff}} \hat{h}_S$, where $e_{j,k} = \langle j | \hat{h}_{\text{eff}} | k \rangle$.

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$[\hat{h}_S + \hat{h}_{\text{eff}}, \hat{\gamma}_S] = 0$. Finally, we notice that

$$-\log(\hat{\gamma}_S) = \beta \hat{h}_S + \log(Z) \quad (3.3.26)$$

and plug it into (3.3.25). This yields the desired result,

$$\frac{d}{dt} \mathcal{S}[\hat{\rho}_S] \geq \underbrace{\beta \text{tr} \{ \hat{h}_S \mathcal{C} \hat{\rho}_S \}}_{=\dot{Q}} + \log(Z) \underbrace{\text{tr} \{ \mathcal{C} \hat{\rho}_S \}}_{=0}, \quad (3.3.27)$$

since $\mathcal{C}$ generates trace preserving dynamics. The master equation is thus consistent with the first- and second law of thermodynamics.

3.4. Case studies in the Born high-energy limit

The central element of the derived master equation (3.2.40) are the scattering amplitudes, which depend sensitively on the system-gas interaction. In order to explore possible scattering amplitudes and by that possible system-gas dynamics, we now present a specific interaction with the spatial profile of a Gaussian and calculate the scattering amplitudes in the Born high-energy limit.

A simple way to construct interactions between system and gas is to combine an operator $\hat{v}$, acting on the internal degrees of freedom of S and A with a position dependent potential acting on the motional degrees of freedom M. The convenient choice of a Gaussian potential yields the general interaction Hamiltonian,

$$\hat{V} = V_0 \hat{v} \otimes \exp \left(-\frac{1}{2} \left(\frac{\hat{\mathbf{x}}}{R} \right)^2 \right). \quad (3.4.1)$$

In the limit of highly energetic, i.e. hot, gas we can employ the Born approximation. In first order Born approximation and with Greek multi index notation the scattering amplitudes read as,

$$\begin{aligned} f_{\nu\kappa}^{(1)}(\mathbf{p}_2, \mathbf{p}_1) &= -(2\pi)^2 \hbar m \langle \mathbf{p}_2, \nu | \hat{V} | \mathbf{p}_1, \kappa \rangle \\ &= -\sqrt{\frac{\pi}{2}} R \frac{V_0}{E_R} \langle \nu | \hat{v} | \kappa \rangle \exp \left(-\frac{1}{2} \frac{(\mathbf{p}_2 - \mathbf{p}_1)^2}{p_R^2} \right). \end{aligned} \quad (3.4.2)$$

Here we performed a Fourier transform and introduced the characteristic momentum $p_R = \hbar/R$ and energy $E_R = p_R^2/(2m)$ of the potential.

Recalling the general form of the Lindblad operators (3.2.42) we plug in the scattering

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amplitude,

$$\hat{L}_E^{n_A, k_A}(\Omega, \mathbf{p}) = -\sqrt{\frac{\pi}{2}} R \frac{V_0}{E_R} \sum_{n_S, k_S}^{\Delta_{n_S, k_S}=E} \sqrt{\frac{P_{n_S, k_S}^{n_A, k_A}(p)}{p}} \exp\left(-\frac{(\mathbf{P}_{n_S, k_S}^{n_A, k_A}(\Omega, p_0) - \mathbf{p})^2}{2p_R}\right) \langle n_S, n_A | \hat{v} | k_S, k_A \rangle | n_S \rangle \langle k_S|. \quad (3.4.3)$$

Fortunately, it is possible to evaluate the integrals with respect to Ω and $\mathbf{p}$ in the master equation explicitly, drastically reducing the complexity of the equation. An explicit calculation in Appendix C.2.4 shows,

$$\int d^3\mathbf{p} \mu(\mathbf{p}) |\mathbf{j}(\mathbf{p})| \int d\Omega \sum_E \mathcal{D} \left[\hat{L}_E^{n_A, k_A}(\mathbf{p}_0, \Omega) \right] \hat{\rho}_S(t) = \Gamma \sum_E \mathcal{D} \left[\hat{J}_E^{n_A, k_A} \right] \hat{\rho}_S(t), \quad (3.4.4)$$

with new Lindblad operators,

$$\hat{J}_E^{n_A, k_A} = \sum_{k_S, n_S}^{\Delta_{n_S, k_S}=E} \sqrt{I\left(\frac{E_R}{k_B T}, \frac{\Delta_{n_S, k_S}^{n_A, k_A}}{E_R}\right)} \langle n_S, n_A | \hat{v} | k_S, k_A \rangle | n_S \rangle \langle k_S|. \quad (3.4.5)$$

The five-dimensional integrals over Ω and $\mathbf{p}$ are now expressed in terms of the rate Γ and an one-dimensional parametric integral I ,

$$\Gamma = \sqrt{2\pi^3} (nR^3) \left(\frac{V_0}{E_R}\right)^2 \left(\frac{p_R}{\sqrt{m k_B T}}\right)^3 \frac{E_R}{\hbar} \quad (3.4.6)$$

$$I(\alpha, s) = e^s \int_{\max\{0, s\}}^{\infty} dz e^{-(2+\alpha)z} \sinh(2\sqrt{z}\sqrt{z-s}). \quad (3.4.7)$$

The integrals in the effective Hamiltonian can also be solved and yield,

$$\hat{h}_{\text{eff}} = \sqrt{2\pi^3} (nR^3) V_0 \sum_{k_A} p^{k_A} \sum_{n_S, k_S} \langle n_S, k_A | \hat{v} | k_S, k_A \rangle | n_S \rangle \langle k_S|. \quad (3.4.8)$$

The parametric integral $I(\alpha, s)$ encodes the key properties of the dissipator: local detailed balance in the case of thermal ancillas as well as a dependence of the dissipation rate on the internal resonances and motional temperature. Its behavior over a wide range of parameters can be seen in Fig. 3.3. It is shown that $I(\alpha, s)$, which is proportional to the corresponding transition rate in the Lindblad master equation, decays much faster for positive s than for negative s . This can also be understood from the formulas, where we find,

$$I(\alpha, -s) = e^{\alpha s} I(\alpha, s), \quad (3.4.9)$$

as derived in Observation 2 in Appendix C.2.5. Hence, $I(\alpha, |s|) = e^{-\alpha|s|} I(\alpha, -|s|)$, i.e., the function decays exponentially faster with an increasingly positive s than with an increasingly negative s . Physically, this can be understood, too: in the scattering

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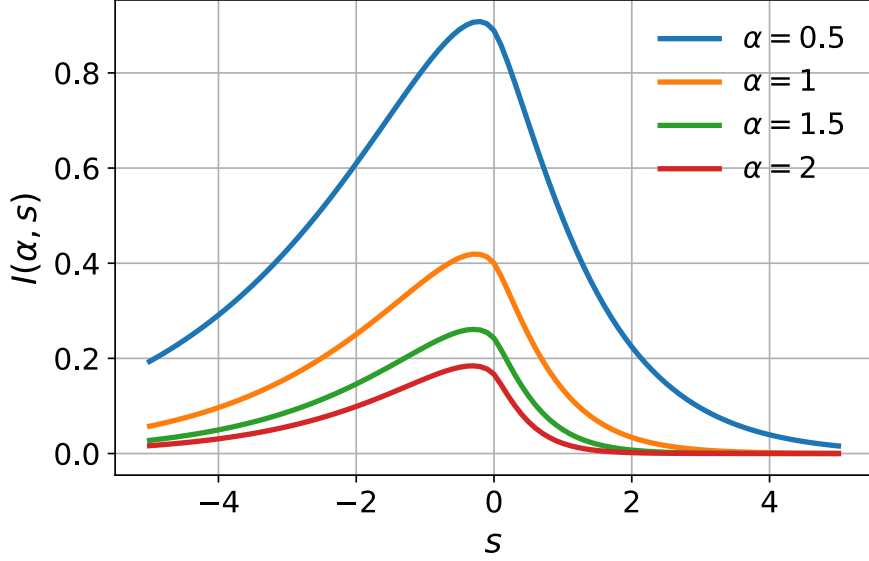


Figure 3.3.: Plot of the integral $I(\alpha, s)$ versus the normalized energy gap s for different values of the characteristic energy of the potential over the thermal energy α . For positive s the curves are exponentially suppressed and decay much faster than for negative s . The maximum is found in all cases for negative s close to the origin.

process, s is the energy that is transferred from the motional degree of freedom to system and ancilla, normalized to the characteristic energy of the potential. So for positive s , the probability of the gas particle to have a sufficiently high kinetic energy to induce an excitation falls off exponentially, as can be seen from $\log(\mu(\mathbf{p})) \propto -E_{\text{kin}}$. Negative s , on the other hand, correspond to an internal de-excitation that contributes to the kinetic energy. The maximum of $s \mapsto I(\alpha, s)$ is always found close to $s \approx 0$ slightly shifted towards negative values.

In summary, when the interaction operator takes the form of (3.4.1) and the first-order Born approximation is applicable, the analysis simplifies significantly. In such cases, we can use reduced forms of both the effective Hamiltonian and the Lindblad operators. The main features governing the open system dynamics depend on the energy gaps of the free Hamiltonian $\hat{h}_S$ and the operator $\hat{v}$.

In order to exemplify the derivation, we discuss systems with different structures of the energy levels in the following subsections. Besides deriving the specific master equation in each case, we also discuss the possibilities when motion and ancilla are thermal with respect to different temperatures.

The internal interactions $\hat{v}$ in these subsections are always motivated by physical scenarios. However, for the sake of simplicity and symbolic solutions, we choose to work with a Gaussian shape of the interaction potential. A more realistic spatial dependence would certainly affect the dissipation rates appearing in the master equation, but we expect the same qualitative behavior as with our Gaussian model.

We start with the simplest case of a qubit system in Subsection 3.4.1. Second, we ex-

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amine a large spin in Subsection 3.4.2. Then, we consider a harmonic oscillator in form of a cavity in subsection 3.4.3, which inherits surprisingly different non-equilibrium properties due to its infinite-dimensional Hilbert space. Finally, we conclude with a planar rotor in 3.4.4, which, regarding the energy gaps, has the most complicated structure.

3.4.1. Flip-Flop interaction of a qubit and a qubit gas

We consider a system qubit with transition frequency ω_S and a gas of ancilla qubits with a possibly detuned frequency ω_A , as given by the internal Hamiltonians,

$$\hat{h}_{S/A} = \frac{\hbar\omega_{S/A}}{2} \hat{\sigma}_{S/A}^z = \frac{\hbar\omega_{S/A}}{2} (|1\rangle\langle 1| - |0\rangle\langle 0|). \quad (3.4.10)$$

For the system-ancilla coupling, we consider the flip-flop interaction given by,

$$\hat{v} = \hat{\sigma}_S^+ \otimes \hat{\sigma}_A^- + \hat{\sigma}_S^- \otimes \hat{\sigma}_A^+. \quad (3.4.11)$$

as in the collision model example of Subsection 3.1.1, which will allow for an instructive comparison. Additionally, we assume the ancilla to be in a Gibbs state at temperature T_A and the motion in a potentially different temperature T_M .

Notice that there are only three different transition energy values entering the Lindblad operator (3.4.5) of the master equation: $E \in \{0, \pm\hbar\omega_S\}$. The value zero implies no internal transition and hence requires the corresponding Lindblad operator to be diagonal. Given that the flip-flop interaction has zero diagonal elements, this means $\hat{J}_0^{n_A, k_A} = 0$. For the same reason we have $\hat{h}_{\text{eff}} = 0$.

The value $E = +\hbar\omega_S$, on the other hand, implies $n_S = 1$ and $k_S = 0$, which yields $\langle 1, n_A | \hat{v} | 0, k_A \rangle = \delta_{n_A, 0} \delta_{k_A, 1}$. The opposite value $E = -\hbar\omega_S$ yields, analogously, $n_S = 0$, $k_S = 1$ and $\langle 0, n_A | \hat{v} | 1, k_A \rangle = \delta_{n_A, 1} \delta_{k_A, 0}$. The internal energy changes only depend on the detuning $\omega_S - \omega_A$ and read,

$$\Delta_{10}^{01} = -\Delta_{01}^{10} = \hbar(\omega_S - \omega_A). \quad (3.4.12)$$

Altogether, the Lindblad operators become,

$$\begin{aligned} \hat{J}_{+\hbar\omega_S}^{n_A k_A} &= \delta_{1k_A} \delta_{0n_A} \sqrt{I \left(\frac{E_R}{k_B T_M}, \frac{\hbar(\omega_S - \omega_A)}{E_R} \right)} |1\rangle\langle 0|, \\ \hat{J}_{-\hbar\omega_S}^{n_A k_A} &= \delta_{0k_A} \delta_{1n_A} \sqrt{I \left(\frac{E_R}{k_B T_M}, -\frac{\hbar(\omega_S - \omega_A)}{E_R} \right)} |0\rangle\langle 1|, \end{aligned} \quad (3.4.13)$$

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and the dissipator can be expressed as,

$$\begin{aligned}
\Gamma \mathcal{C} &= \Gamma \sum_{n_A, k_A} p^{k_A} \sum_E \mathcal{D} \left[J_E^{n_A, k_A} \right] \\
&= \Gamma \left(p^1 I \left(\frac{E_R}{k_B T_M}, \frac{\hbar(\omega_S - \omega_A)}{E_R} \right) \mathcal{D} [\hat{\sigma}^+] + p^0 I \left(\frac{E_R}{k_B T_M}, -\frac{\hbar(\omega_S - \omega_A)}{E_R} \right) \mathcal{D} [\hat{\sigma}^-] \right) \\
&= \Gamma I \left(\frac{E_R}{k_B T_M}, \frac{\hbar(\omega_S - \omega_A)}{E_R} \right) \left(p^1 \mathcal{D} [\hat{\sigma}^+] + p^0 \exp \left(\frac{\hbar(\omega_S - \omega_A)}{k_B T_M} \right) \mathcal{D} [\hat{\sigma}^-] \right). \quad (3.4.14)
\end{aligned}$$

In the last line, we made use of the identity (3.4.9) for the integral function $I(\alpha, s)$, resulting in a common rate prefactor for both terms. Inserting $p^{k_A} = \langle k_A | \hat{\gamma}_A | k_A \rangle$ and pulling out a common factor of $\exp \left(\frac{\hbar(\omega_S - \omega_A)}{2k_B T_M} \right)$, we find an effectively thermal dissipator,

$$\Gamma \mathcal{C} = \tilde{\Gamma} \left(\exp \left(-\frac{\hbar\omega_S}{2k_B T_{\text{eff}}} \right) \mathcal{D} [\hat{\sigma}^+] + \exp \left(+\frac{\hbar\omega_S}{2k_B T_{\text{eff}}} \right) \mathcal{D} [\hat{\sigma}^-] \right) / Z_{\text{eff}}, \quad (3.4.15)$$

with the effective temperature ,

$$T_{\text{eff}} = \frac{T_A T_M}{\left[1 - \frac{\omega_A}{\omega_S} \right] T_A + \left[\frac{\omega_A}{\omega_S} \right] T_M} \quad (3.4.16)$$

and the rescaled rate,

$$\begin{aligned}
\tilde{\Gamma} &= \Gamma \frac{Z_{\text{eff}}}{Z_A} \exp \left(\frac{\hbar(\omega_S - \omega_A)}{2k_B T_M} \right) I \left(\frac{E_R}{k_B T_M}, \frac{\hbar(\omega_S - \omega_A)}{E_R} \right) \\
&= \Gamma \frac{Z_{\text{eff}}}{Z_A} \exp \left(\frac{\hbar|\omega_S - \omega_A|}{2k_B T_M} \right) I \left(\frac{E_R}{k_B T_M}, \frac{\hbar|\omega_S - \omega_A|}{E_R} \right). \quad (3.4.17)
\end{aligned}$$

The corresponding master equation thus lets the system qubit equilibrate to a Gibbs state at the effective temperature (3.4.16).

With these results at hand, we can now investigate how the effective temperature T_{eff} , and by that the equilibrium state, behaves. In the most natural case in which motion and internal degrees of freedom of the gas are at the same temperature $T_M = T_A = T$, the effective temperature is also $T_{\text{eff}} = T$. The system therefore thermalizes, exemplifying the general result from Subsection 3.3.1, which states that the system thermalizes. It is instructive to make a comparison to the framework of repeated interactions, where a detuned system did not thermalize, see Subsection 3.1.1. Note however, that for detunings that are large compared to the characteristic energy of the potential E_R , i.e. $\hbar|\omega_S - \omega_A| \gg E_R$, the effective rate $\tilde{\Gamma}$ becomes strongly suppressed, which can be seen from the upper bound,

$$\tilde{\Gamma} \leq \Gamma I \left(\frac{E_R}{k_B T}, -\frac{\hbar|\omega_S - \omega_A|}{E_R} \right), \quad (3.4.18)$$

which we derive in Appendix C.2.6 and the behavior of $I(\alpha, s)$ shown in Fig. 3.3. Hence,

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strongly detuned ancillas have virtually no effect on the system and show no thermalization for comparably short observation times.

While $T_A = T_M$ represents the standard setting of a low density heat bath, it is also possible to set up a nonequilibrium scenario of unequal temperatures. This could be realized by expanding a thermal gas through a nozzle, which reduces the kinetic energy. If the direct coupling between motional and internal degrees of the gas is negligible, the internal temperature remains unchanged, meaning $T_A > T_M$. In this scenario, a negative effective system temperature is achievable. Looking at (3.4.16), this is possible when $1 - \frac{\omega_A}{\omega_S} < 0$ and T_A is sufficiently hot compared to T_M . The exact condition is found to be,

$$1 - \frac{T_M}{T_A} > \frac{\omega_S}{\omega_A}, \quad (3.4.19)$$

which can only be fulfilled when the necessary conditions $\omega_A > \omega_S$ and $T_A > T_M$ are met.

The transitions from positive to negative effective temperatures occur due to a flip of the sign in the denominator of (3.4.16). Hence, continuously scanning through the parameters from the positive to the negative regime will inevitably yield a transition to an infinite positive temperature and an infinite negative temperature. This sudden jump in temperature becomes clear if one considers the fact that, for positive and negative infinite temperature, the equilibrium state is the maximally mixed state

$$\lim_{T_{\text{eff}} \rightarrow \pm\infty} \hat{\tau}_S = \mathbb{1}/2. \quad (3.4.20)$$

Therefore, the emergence of infinite temperature corresponds to the maximally mixed state, characterized by equal populations. Once the threshold towards negative temperature is crossed, the equilibrium state of the system starts to exhibit a higher population in the more-energetic excited state $|1\rangle$ than in the ground state $|0\rangle$. This *population inversion* is the strongest if T_{eff} is negative and close to zero. This optimal value of $T_{\text{eff}} = -0$ can be achieved asymptotically for $T_A \gg T_M$,

$$\lim_{T_A \rightarrow \infty} T_{\text{eff}} = \frac{T_M}{\left[1 - \frac{\omega_A}{\omega_S}\right]}. \quad (3.4.21)$$

Since T_M can, in principle, be chosen arbitrarily small, we can achieve negative effective temperatures arbitrarily close to zero. Thus, any population inversion

$$\hat{\tau}_S = q |1\rangle \langle 1| + (1 - q) |0\rangle \langle 0|, \quad \forall q \in [0, 1] \quad (3.4.22)$$

is within reach.

In such a population-inverted system, we can use a unitary transformation to extract useful energy, see Subsection 1.4.3. The maximal amount of energy that can be extracted is referred to as *ergotropy*. With respect to $\hat{h}_S$, the ergotropy is formally defined as shown

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in Subsection 1.4.3,

$$\mathcal{E}[\hat{\tau}_S] = \text{tr} \left\{ \hat{h}_S \hat{\tau}_S \right\} - \min_{\hat{U}} \text{tr} \left\{ \hat{h}_S \hat{U} \hat{\tau}_S \hat{U}^\dagger \right\}. \quad (3.4.23)$$

In a population-inverted state without coherences, the most energy is extracted by a unitary that flips the populations between the energy levels, which results in,

$$\mathcal{E}[\hat{\tau}_S] = \begin{cases} 0, & \text{for } q \leq 1/2 \\ \hbar\omega_S (2q - 1), & \text{else} \end{cases} = \begin{cases} 0, & \text{for } T_{\text{eff}} \geq 0 \\ -\hbar\omega_S \tanh\left(\frac{\hbar\omega_S}{2k_B T_{\text{eff}}}\right), & \text{else} \end{cases}, \quad (3.4.24)$$

where we substituted the expression for q in the second step.

The result extends in a straightforward manner to the case in which the system is a large spin, as we will see in Subsection 3.4.2.

3.4.2. Large spin in a qubit gas

We now consider a $2j + 1$ -dimensional large spin subjected to the qubit gas. The free Hamiltonian is given by,

$$\hat{h}_S = \omega_S \hat{S}_z = \hbar\omega_S \sum_{k_S=-N}^N k_S |k_S\rangle \langle k_S|. \quad (3.4.25)$$

Here, we take the interaction to be the standard linear spin-spin coupling as in the case of interacting magnetic dipoles, for example,

$$\hbar\hat{v} = \hat{\mathbf{S}} \cdot \hat{\boldsymbol{\sigma}} = \hat{S}_+ \hat{\sigma}_- + \hat{S}_- \hat{\sigma}_+ + \hat{S}_z \hat{\sigma}_z. \quad (3.4.26)$$

The raising and lowering operators of the spin are conveniently characterized by their effect on the spin-z basis,

$$\begin{aligned} \hat{S}_+ |k_S\rangle &= \hbar\sqrt{(j - k_S)(j + k_S + 1)} |k_S + 1\rangle \\ \hat{S}_- |k_S\rangle &= \hbar\sqrt{(j + k_S)(j - k_S + 1)} |k_S - 1\rangle. \end{aligned} \quad (3.4.27)$$

The first two terms in (3.4.26) effect an exchange of excitations as in the qubit case before (3.4.13), whereas the additional spin-z term corresponds to an interaction-induced energy level shift, which will be responsible for an effective Hamiltonian and a dephasing term in the master equation.

The spectrum of the spin is an equally spaced ladder, which means that the scattering Lindblad operators do not only induce a single transition between two specific energy levels separated by a multiple of $\hbar\omega_S$, but arbitrarily many across the whole ladder. Since the interaction operator (3.4.26) supports only single or zero quanta transitions between neighboring energy levels, we only have to consider the energy gaps $E \in \{0, \pm\hbar\omega_S\}$. Starting with the Lindblad operator $\hat{J}_E^{nk}$ associated to $E = +\hbar\omega_S$, we have the transitions

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$k + 1 \leftarrow k$, which require the ancilla qubit to de-excite, $0 \leftarrow 1$. This shows the total energy gap is of the form $\Delta_{k+1,k}^{0,1} = \hbar(\omega_S - \omega_A)$. The corresponding Lindblad operator similar to the qubit case reads,

$$\hat{J}_{\hbar(\omega_S - \omega_A)}^{01} = \sqrt{I\left(\frac{E_R}{k_B T_M}, \frac{\hbar(\omega_S - \omega_A)}{E_R}\right)} \begin{pmatrix} \hat{S}_+ \\ \hbar \end{pmatrix}. \quad (3.4.28)$$

The analogous procedure for $E = -\hbar\omega_S$ yields,

$$\hat{J}_{\hbar(\omega_A - \omega_S)}^{10} = \sqrt{I\left(\frac{E_R}{k_B T_M}, \frac{\hbar(\omega_A - \omega_S)}{E_R}\right)} \begin{pmatrix} \hat{S}_- \\ \hbar \end{pmatrix}. \quad (3.4.29)$$

The Lindblad operators corresponding to the energy transition $E = 0$ read,

$$\hat{J}_0^{11} = \sqrt{I\left(\frac{E_R}{k_B T}, 0\right)} \begin{pmatrix} \hat{S}_z \\ \hbar \end{pmatrix} \quad (3.4.30)$$

$$\hat{J}_0^{00} = -\sqrt{I\left(\frac{E_R}{k_B T}, 0\right)} \begin{pmatrix} \hat{S}_z \\ \hbar \end{pmatrix} = -\hat{J}_0^{11}, \quad (3.4.31)$$

where the minus sign in the second terms cancels anyway in the master equation.

As in the preceding example, we can use Observation 1 in Appendix C.2.5 to rewrite,

$$I\left(\frac{E_R}{k_B T}, -\frac{\hbar(\omega_S - \omega_A)}{E_R}\right) = e^{\beta\hbar(\omega_S - \omega_A)} I\left(\frac{E_R}{k_B T}, \frac{\hbar(\omega_S - \omega_A)}{E_R}\right), \quad (3.4.32)$$

and pull the common factor out of the dissipator in order to find,

$$\begin{aligned} \Gamma\mathcal{C} &= \sum_{n_A, k_A} p^{k_A} \sum_E \mathcal{D} \left[\hat{J}_E^{n_A k_A} \right] \\ &= \Gamma \left(p^1 \mathcal{D} \left[\hat{J}_0^{11} \right] + p^0 \mathcal{D} \left[\hat{J}_0^{00} \right] \right) + \Gamma \left(p^1 \mathcal{D} \left[\hat{J}_{\hbar(\omega_S - \omega_A)}^{01} \right] + p^0 \mathcal{D} \left[\hat{J}_{-\hbar(\omega_S - \omega_A)}^{10} \right] \right) \\ &= \Gamma_0 \mathcal{D} \left[\frac{\hat{S}_z}{\hbar} \right] + \frac{\tilde{\Gamma}}{Z_{\text{eff}}} \left(\exp \left(-\frac{\hbar\omega_S}{2k_B T_{\text{eff}}} \right) \mathcal{D} \left[\frac{\hat{S}_+}{\hbar} \right] + \exp \left(+\frac{\hbar\omega_S}{2k_B T_{\text{eff}}} \right) \mathcal{D} \left[\frac{\hat{S}_-}{\hbar} \right] \right), \end{aligned} \quad (3.4.33)$$

where we used that $\hat{J}_0^{00} = -\hat{J}_0^{11}$ and $p^0 + p^1 = 1$. The effective rate and temperature of this thermalizing dissipator are the same as in 3.1.1. The thermalizing terms are accompanied by an additional dephasing term at the effective rate,

$$\Gamma_0 = I\left(\frac{E_R}{k_B T_M}, 0\right) \Gamma. \quad (3.4.34)$$

3. Thermodynamics in a scattering environment

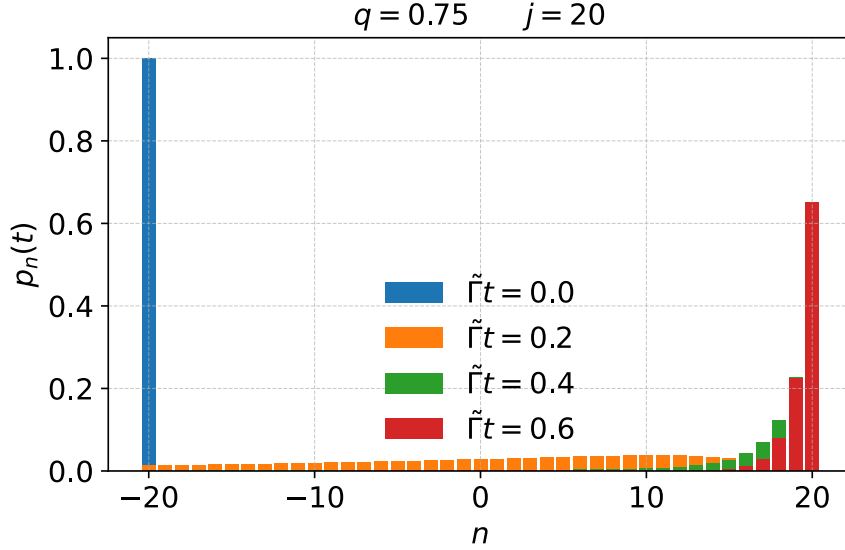


Figure 3.4.: Time evolution of the populations of a large spin with $j = 20$ subjected to a gas with negative effective temperature. Solving Equation (3.4.37) with $q = 0.75$. The system starts in the ground state $n = -j$. Then, the probabilities spread over bigger values of n , before accumulating at the right boundary.

In this example, we also have a non-zero effective Hamiltonian $\hat{h}_{\text{eff}}$ since

$$\langle n_S, k_A | \hbar \hat{v} | k_S, k_A \rangle = \langle n_S | \hat{S}_z | k_S \rangle \underbrace{\langle k_A | \hat{\sigma}_z | k_A \rangle}_{=\pm 1}. \quad (3.4.35)$$

Plugged into (3.4.8), we find,

$$\begin{aligned} \hat{h}_{\text{eff}} &= \sqrt{2\pi}^3 (nR^3) V_0 (p^1 - p^0) \frac{\hat{S}_z}{\hbar} \\ &= -\sqrt{2\pi}^3 (nR^3) V_0 \tanh\left(\frac{\hbar\omega_A}{2k_B T_A}\right) \frac{\hat{S}_z}{\hbar}. \end{aligned} \quad (3.4.36)$$

This could be viewed as an effective finite-temperature Lamb shift of the system resonance, which vanishes in the high-temperature limit.

Again, we consider the buildup of ergotropy in a more general out-of-equilibrium setting with different temperatures $T_A \neq T_M$ of motion and ancilla. For a diagonal initial state and in terms of the dimensionless time $\tau = \tilde{\Gamma}t$, we find the evolution of the populations to obey,

$$\begin{aligned} \frac{d}{d\tau} p_n(\tau) &= (j - n + 1)(j + n) [qp_{n-1}(\tau) - (1 - q)p_n(\tau)] \\ &\quad + (j + n + 1)(j - n) [(1 - q)p_{n+1}(\tau) - qp_n(\tau)], \end{aligned} \quad (3.4.37)$$

3. Thermodynamics in a scattering environment

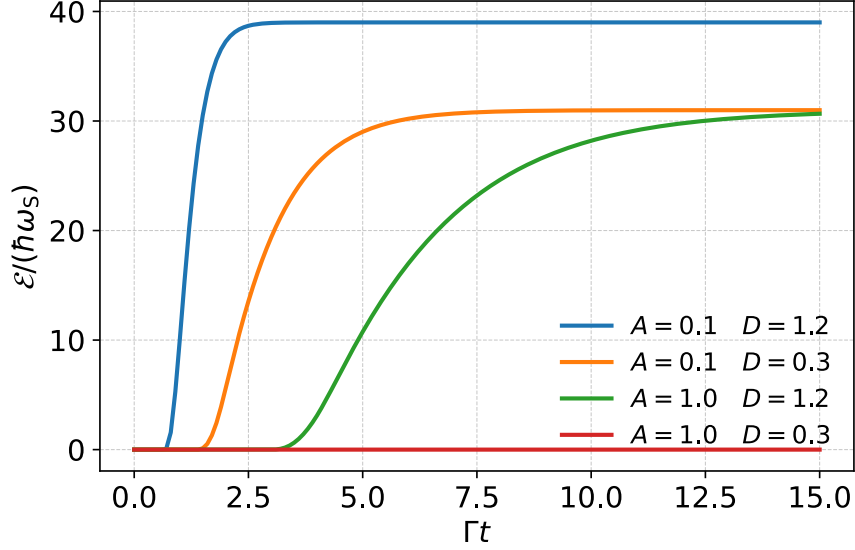


Figure 3.5.: Time evolution of the ergotropy of a spin with $j = 20$ subjected to a gas of two-level systems. We set $k_B T_M = E_R$ and vary the dimensionless quantities A and D . The difference $A - D$ characterizes the effective temperature, which becomes negative for $D > A$. The red line corresponds to a positive temperature and thus does not build up ergotropy and the orange and green line correspond to the same temperature, equilibrating them at the same ergotropy.

where q is given by (3.4.45). The results for a system, which is initially in the ground state $n = -j$ and for $q = 0.75$, can be seen in Fig. 3.4. Then the probabilities accumulate at high energies and form an inverted Gibbs state. Hence, ergotropy is built up. We investigate this further in Fig. 3.5, where the ergotropy of a $j = 20$ spin initially in the ground state is plotted versus the effective time Γt . We choose $k_B T_M = E_R$ and vary two dimensionless quantities that characterize the energy gap of the ancilla and the detuning,

$$A = \frac{\hbar T_A}{k_B T_A}, \quad D = \frac{\hbar(\omega_A - \omega_S)}{k_B T_M}. \quad (3.4.38)$$

The effective temperature is characterized by $A - D$. We see that, for $D > A$, ergotropy is built up after a short onset time. The value of ergotropy, at which the system saturates, is also determined by $A - D$.

This subsection presented the dynamical build up of ergotropy in finite-dimensional systems. In the following subsections, we will consider infinite-dimensional systems.

3.4.3. Cavity mode in a qubit gas

As the third example we choose a harmonic oscillator. Physically, this could be a mode of the electromagnetic field, interacting via dipole coupling of the $\mathbf{E}$ -field. The system

3. Thermodynamics in a scattering environment

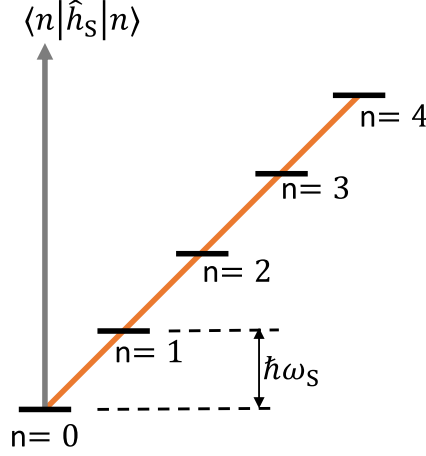


Figure 3.6.: First few energy levels of a harmonic oscillator. The gap between neighboring levels is always equal to $\hbar\omega_S$

is described by the Hamiltonian,

$$\hat{h}_S = \hbar\omega_S \hat{a}^\dagger \hat{a} = \hbar\omega_S \sum_{n=0}^{\infty} n |n\rangle \langle n|. \quad (3.4.39)$$

with the interaction,

$$\hat{E}d \propto (\hat{\sigma}^+ + \hat{\sigma}^-) (\hat{a} + \hat{a}^\dagger) \approx \hat{\sigma}^+ \hat{a} + \hat{\sigma}^- \hat{a}^\dagger = \hat{v}, \quad (3.4.40)$$

where we neglected the counter-rotating terms with the rotating wave approximation.

For infinite dimensional systems, caution is advised when using the scattering based master equation: A crucial point in the derivation was the assumption that the free evolution of the system is slow compared to the scattering interaction. A highly excited Fock state $|n\rangle$, for example, evolves with the frequency $n\omega_S$ and does not meet this condition anymore. For sufficiently low excitations, however, the condition is fulfilled and we can apply the master equation safely.

Since the spectrum of the harmonic oscillator is an equally spaced ladder, as in the case of the large spin, the derivation of the master equation follows analogously. The Lindblad operators (3.4.5) read,

$$\hat{J}_{\hbar\omega_S}^{0,1} = \sqrt{I \left(\frac{E_R}{k_B T}, \frac{\hbar(\omega_S - \omega_A)}{E_R} \right)} \hat{a}^\dagger \quad (3.4.41)$$

$$\hat{J}_{-\hbar\omega_S}^{1,0} = \sqrt{I \left(\frac{E_R}{k_B T}, -\frac{\hbar(\omega_S - \omega_A)}{E_R} \right)} \hat{a} \quad (3.4.42)$$

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and the full master equation becomes,

$$\Gamma \mathcal{C} = \tilde{\Gamma} \left(\exp \left(-\frac{\hbar\omega_S}{2k_B T_{\text{eff}}} \right) \mathcal{D} [\hat{a}^\dagger] + \exp \left(+\frac{\hbar\omega_S}{2k_B T_{\text{eff}}} \right) \mathcal{D} [\hat{a}] \right) / Z_{\text{eff}}. \quad (3.4.43)$$

Here, the effective temperature T_{eff} and the modified rate Γ are the same as for the qubit case and given by the formulas (3.4.16) and (3.4.17). Since the harmonic oscillator has an infinite number of energy levels, there is no fully population-inverted state and we thus cannot expect an equilibrium state for $T_{\text{eff}} < 0$. However, transient population inversion and ergotropy can in principle merge over time. To see whether this is the case, we consider the ergotropy dynamically: Since the off-diagonal elements evolve independently from the diagonal elements, and it is straightforward to see that they will decohere over time, we can make the Ansatz,

$$\hat{\rho}_S(t) = \sum_{n=0}^{\infty} p_n(t) |n\rangle \langle n|, \quad (3.4.44)$$

if the initial state was already diagonal, e.g. the ground state $|0\rangle \langle 0|$. With the abbreviation,

$$q = \exp \left(-\frac{\hbar\omega_S}{2k_B T_{\text{eff}}} \right) / Z_{\text{eff}} \quad (3.4.45)$$

and with respect to the dimensionless time $\tau = \tilde{\Gamma}t$, we find the evolution of the populations,

$$\frac{d}{d\tau} p_n(\tau) = nq p_{n-1}(\tau) + (n+1)(1-q) p_{n+1}(\tau) - [n+q] p_n(\tau) \quad (3.4.46)$$

For the extreme case $q = 1$ and the initial condition $p_n(0) = \delta_{n,0}$, this can be solved analytically by iteratively applying the method of variation of constants. The populations become,

$$p_n(\tau) = e^{-\tau} (1 - e^{-\tau})^n, \quad n \geq 0, \quad (3.4.47)$$

which is verified in Appendix C.2.7. The result implies that for any fixed time τ the probability of a high excitation n decays with a power law, $p_n(\tau) \propto (1 - e^{-\tau})^n$. Put differently, the lower the energy of a state, the higher its probability. Hence even in the ideal case of $q \rightarrow 1$, the master equation cannot generate population inversion at all times. Consequently,

$$\mathcal{E} [\hat{\rho}_S(\tau)] = 0, \quad \forall \tau. \quad (3.4.48)$$

For less extreme cases, we can invoke numerics to solve (3.4.46). For $q = 0.75$ and a system initially in the ground state, we find the dynamics shown in Fig. 3.7. We see that no population inversion occurs: As in the extreme $q = 1$ case, the probability for all levels $p_n(\tau)$ is smaller than $p_k(\tau)$ for all $k < n$ and all times τ . This again shows that energy cannot be extracted unitarily. Hence, the ergotropy in this case is zero, as well. This shows a difference between finite and infinite-systems: In Subsection 3.4.2, the

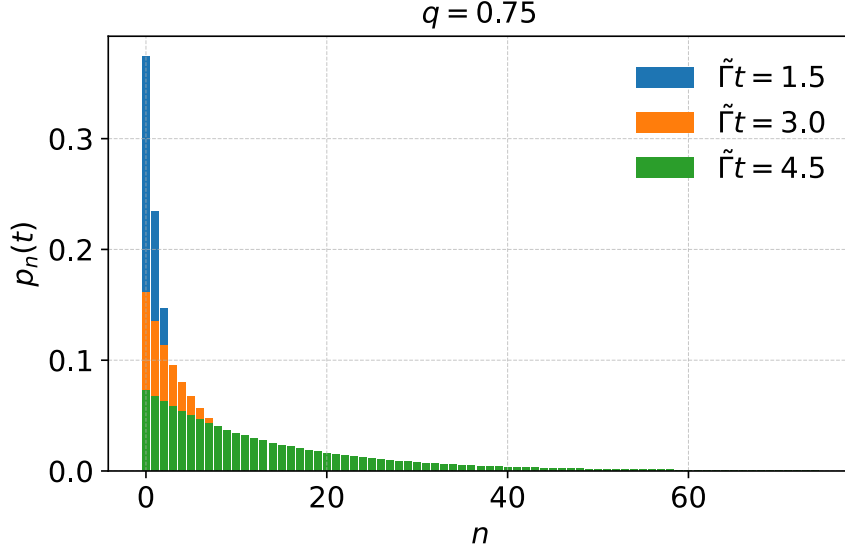


Figure 3.7.: Numerical results for the populations of a cavity mode subjected to a thermal gas with negative effective temperature, i.e. the solution to Equation (3.4.46) with $q = 0.75$. The system is initially in the ground state. Qualitatively, we see the same behavior as in the $q = 1$ case: The bigger n , the smaller the probability.

increasing populations eventually reach the highest energy eigenstate and accumulate there, forming an inverted Gibbs state. However, we can also build up ergotropy in the current situation when the initial state carries at least some ergotropy, say $p_n(0) = \delta_{n,1}$. Then the ground state remains unoccupied for all times τ if we choose $q = 1$. The populations of the ground state and an higher energetic state can then be exchanged in order to reduce the system energy. A discussion of the requirement of initial ergotropy can be found in [171].

In summary, we derived the master equation for a cavity mode and investigated the possibilities to build up ergotropy with a negative effective temperature. While the energy of the system got increased steadily due to the occupation of higher levels n , there is no ergotropy production at all if the initial state has no ergotropy. This is rooted in the fact that the system is infinite-dimensional as can be seen from comparison with the similar but finite-dimensional example of the large spin in Subsection 3.4.2.

3.4.4. Planar rotor in a qubit gas

As the final example, we will now discuss a quantum planar rotor subjected to a gas. The main difference to the preceding examples lies in the fact that the energies of the free Hamiltonian grow quadratically,

$$\hat{h}_S = \frac{\hat{L}_z^2}{2I}. \quad (3.4.49)$$

3. Thermodynamics in a scattering environment

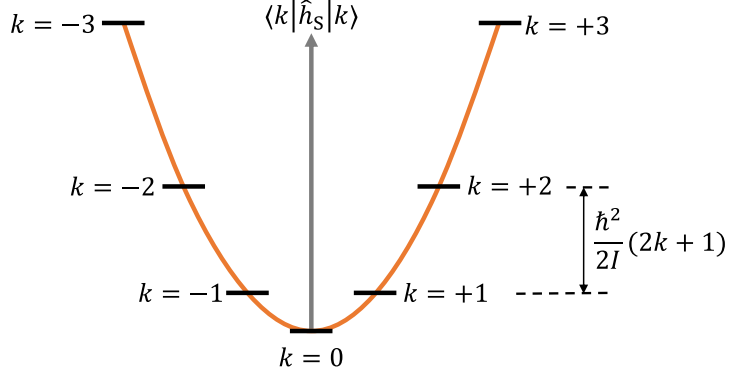


Figure 3.8.: First few energy levels of free planar rotor. The gap between neighboring levels varies with k . Due to the $\hat{L}_z^2$ term in the Hamiltonian, the spectrum is symmetric under $k \leftrightarrow -k$

As a consequence, there are now infinitely many transition energies, which results in individual Lindblad operators for each such resonance.

As in the cavity mode example in Subsection 3.4.3, the approximations in order to derive the master equation will break down if the rotor's kinetic energy is too high. Since it is self-evident in this example which operators and states belong to S and which belong to A, we drop the subscripts A/S in the following. In particular, the frequency of the ancilla qubits is just denoted by ω .

We assume a coupling via the orientations of rotor and spin, which could be understood as the interaction between a permanent magnet rotating in a fixed plane with magnetic dipoles associated to the ancilla spins,

$$\begin{aligned}
 \hat{v} &= \mathbf{e}(\hat{\alpha}) \cdot \hat{\boldsymbol{\sigma}} = \begin{pmatrix} \cos(\hat{\alpha}) \\ \sin(\hat{\alpha}) \\ 0 \end{pmatrix} \cdot \begin{pmatrix} \hat{\sigma}_x \\ \hat{\sigma}_y \\ \hat{\sigma}_z \end{pmatrix} \\
 &= \frac{1}{2} (\hat{L}_+ + \hat{L}_-) \otimes \hat{\sigma}_x + \frac{1}{2i} (\hat{L}_+ - \hat{L}_-) \otimes \hat{\sigma}_y \\
 &= \hat{L}_+ \otimes \frac{1}{2} (\hat{\sigma}_x - i\hat{\sigma}_y) + \hat{L}_- \otimes \frac{1}{2} (\hat{\sigma}_x + i\hat{\sigma}_y) \\
 &= \hat{L}_+ \otimes \hat{\sigma}^- + \hat{L}_- \otimes \hat{\sigma}^+,
 \end{aligned} \tag{3.4.50}$$

where we rewrote sine and cosine in terms of complex exponentials. Notice that, since $\hat{v}$ does not have any element of the form $\langle n, m | \hat{v} | k, m \rangle$, there is no effective Hamiltonian,

$$\hat{h}_{\text{eff}} = 0. \tag{3.4.51}$$

In order to derive the Lindblad operators of the form (3.4.5), we have to consider the degeneracies of the free Hamiltonian depicted in Fig. 3.8. Although the free planar rotor is doubly degenerate due to the $k \leftrightarrow -k$ symmetry of the energy spectrum, the structure of $\hat{v}$ reduces the number of possible transitions that a single Lindblad operator can induce. Namely, $\hat{v}$ only introduces transitions between neighboring energy levels.

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Hence, we only have to consider energy gaps of the form $E = \frac{\hbar^2}{2I} [(k+1)^2 - k^2] = \frac{\hbar^2}{2I}(2k+1)$, $k \in \mathbb{Z}$. For each such gap, there are two possible transitions with this gap, $k+1 \leftarrow k$ and $[-k]-1 \leftarrow [-k]$. From the interaction $\hat{v}$ we know that an increase in angular momentum of the rotor decreases the spin of the qubit. Thus, we get the total internal energy changes $\Delta_{k+1,k}^{0,1} = \frac{\hbar^2}{2I}(2k+1) - \hbar\omega$ and $\Delta_{-k-1,k}^{1,0} = \frac{\hbar^2}{2I}(2k+1) + \hbar\omega$. All things considered, we obtain two types of Lindblad operators,

$$\hat{J}_k^{0,1} = \sqrt{I \left(\frac{E_R}{k_B T}, \frac{\frac{\hbar^2}{2I}(2k+1) - \hbar\omega}{E_R} \right)} |k+1\rangle \langle k| \quad (3.4.52)$$

$$\hat{J}_k^{1,0} = \sqrt{I \left(\frac{E_R}{k_B T}, \frac{\frac{\hbar^2}{2I}(2k+1) + \hbar\omega}{E_R} \right)} |-k-1\rangle \langle -k|. \quad (3.4.53)$$

These can be rewritten in form of rescaled raising and lowering operators of angular momentum,

$$\hat{J}_k^+ = \sqrt{I \left(\frac{E_R}{k_B T}, \frac{\frac{\hbar^2}{2I}(1+2k) - \hbar\omega}{E_R} \right)} |k+1\rangle \langle k| \quad (3.4.54)$$

$$\hat{J}_k^- = \sqrt{I \left(\frac{E_R}{k_B T}, \frac{\frac{\hbar^2}{2I}(1-2k) + \hbar\omega}{E_R} \right)} |k-1\rangle \langle k|. \quad (3.4.55)$$

The full system dynamics becomes,

$$\frac{d}{dt} \hat{\rho}_S = -\frac{i}{\hbar} \left[\frac{\hat{L}_z^2}{2I}, \hat{\rho}_S \right] + \Gamma \sum_{k \in \mathbb{Z}} \left(p^1 \mathcal{D} [\hat{J}_k^+] + p^0 \mathcal{D} [\hat{J}_k^-] \right). \quad (3.4.56)$$

Here, we have a more complicated master equation with an infinite number of Lindblad operators. This once more highlights the influence of the energy gaps of the free Hamiltonian for the collisional dynamics. However, the increased complexity can be useful: Since the parameter integral $I(\alpha, s)$ decays fast for large detunings (recall Fig. 3.3), only a few Lindblad operators in (3.4.56) significantly contribute to the overall dynamics. With a proper choice of parameters these relevant Lindblad operators can be chosen.

3.5. Conclusion

In this chapter, we have investigated scattering based collision models for the use in quantum thermodynamics. First, we discussed repeated interaction models and highlighted the fact that, in order to properly thermalize the system, the environment has to be tailored specifically to the system. This flaw in the description of physical scattering processes with given internal resonances was traced back to the energy introduced to

3. Thermodynamics in a scattering environment

the system by implicitly assuming that an interaction Hamiltonian be switched on and off. Scattering-based models do not suffer from this problem, because the interaction is mediated by motional degrees of freedom.

We presented the derivation of a low-density limit master equation. While the master equation without structured gas particles has been known in the literature since the seminal work of Dümcke [55], we presented a self-contained elaborate derivation in modern language, which included internal degrees of freedom of the gas.

The laws of thermodynamics were derived for the scattering master equation in the same fashion as they were shown for repeated interactions in [15]. The zeroth law implies that a system, if it thermalizes, reaches a Gibbs state at the bath's temperature. This Gibbs state is a fixed point of the dynamics under the microreversibility condition, aligning with a result from Dümcke [55]. In terms of thermalization, this confirms the advantage of scattering-based collision models over repeated interaction models. A more general comparison of repeated interactions and scattering based models in the elastic limit is presented in [172]. A complementary result is presented in [173], where systems without micro-reversibility are studied. There, the systems still thermalize although the local detailed balance condition is not fulfilled.

Some care had to be taken for the first law: It is tempting to consider the motional and internal degrees of freedom of the gas as two individual thermal baths. As we pointed out, this is not possible in a three-body interaction between system, ancilla, and motion. The bath must be viewed as a single composite reservoir that can be in thermal equilibrium or not. Alicki's dynamical version of the second law is directly applicable in the thermal equilibrium case here.

We presented several examples that highlight the importance of the energy structure of the free Hamiltonian on the open system dynamics, including a qubit, a large spin, a cavity, and a planar rotor. As a non-thermal application, we study the dynamics of motional and internal gas degrees of freedom at different temperatures. In the finite-dimensional examples, this can create population inversion and the build-up of ergotropy.

We are confident that the presented findings and the modernized derivation of the low density master equation will help the thermodynamics community to rediscover scattering based collision models. The ergotropy build-up in the non-thermal gas shows an uncontrolled way to generate work, where no external driving is required. This could be a promising new route away from quantum thermal machines that require an external clock and energetically expensive control fields.

A wide range of inspirations can be drawn from the deeply studied one-dimensional models [53, 160]: First of all, a detailed comparison of the one- and three-dimensional model would be very beneficial. If we knew, for example, that the models coincide in a certain regime, we could resort to the less complex one-dimensional model whenever we are in this regime. A fluctuation theorem for energy in a three-dimensional scattering environment could be generalized from [163].

4. Metrology for scattering interactions in electron microscopes

This chapter is based on publication [C].

4.1. Introduction to free-electron quantum optics

By enabling the resolution of structures, too small to be seen with the bare human eye, the field of microscopy has always been a key groundwork in other fields like biology, chemistry, material science, and many more. While microscopes work successfully in many cases with a beam of light, the use of visible light ultimately limits the size of the resolvable objects: The Rayleigh criterion states that the angular resolution $\Delta\varphi$ is related to the wavelength of light λ and the diameter of the aperture $2R$ [174],

$$\Delta\varphi = 1.22 \frac{\lambda}{2R}. \quad (4.1.1)$$

Since the size of the aperture R is usually bound by practical, sometimes even financial reasons, it is more efficient to use sufficiently short wavelengths λ . Unfortunately for visible light, the wavelength is lower bounded by $\sim 380\text{nm}$. However, much smaller wavelengths can be achieved by using the material waves of quantum particles that have a wavelength that behaves anti-proportional to their momentum p ,

$$\lambda = \frac{h}{p}. \quad (4.1.2)$$

As the name suggests, *electron microscopes* do use the material waves of electrons that, for example, have a wavelength of $\lambda \sim 3.7\text{pm}$ for an acceleration energy of $E = 100\text{keV}$. Note that $100 - 200\text{keV}$ are achieved in standard transmission electron microscopes and specialized high voltage electron microscopes can even reach up to 3MeV [8].

Until recently, electron microscopes have been mainly used in applied sciences and quantum effects, aside from the wave-particle duality on which the microscopes are based, have not been exploited. This changes now with the rise of the new field of *free-electron quantum optics*, where the free electrons are considered a quantum resource [10]. Two works that acknowledge and exploit the quantum nature of the electron are [11, 12], where quantum metrology is incorporated in order to derive the quantum

Fisher information (QFI) for samples that can be described by a potential $V(\mathbf{x})$.

In this work, we contribute to the field by presenting a detailed metrological analysis of the measurement of magnetic moments. For this, we consider two different tasks. On the one hand, we consider the strength of the magnetic moment to be an unknown parameter. We apply estimation theory and calculate the Fisher information to obtain bounds on the measurement precision. On the other hand, we assume that a magnetic moment of a known strength has been placed in the path of the electron or not. The task is then to guess whether there was a target or not with the optimal success probability, which is characterized by the trace distance. For both tasks, we first consider a model with a static magnetic moment that is not influenced by the interaction with the electron. Second, we promote the magnetic moment to be a quantum spin that can change in the interaction.

The chapter is structured as follows: We begin with the mathematical model in Section 4.2, where we derive the Hamiltonian for the two cases, with and without backaction, derive the respective scattering operators in the slowly-varying envelope approximation, and discuss a basis for angular momentum measurements. The sections 4.3 and 4.4 deal with the task of parameter estimation and are structured in the same manner. The former considers the case without backaction, while the latter includes backaction. In both cases, the QFI is calculated (or at least bounded) and then the classical Fisher information (FI) in position, momentum, and angular momentum measurements is compared. Analogously, the task of state discrimination is treated in Section 4.5 without backaction and in Section 4.6 with backaction. Finally, we conclude in Section 4.7

4.2. The Model

An electron microscope is a complicated machine that accelerates electrons, focuses them on a sample, applies lens transformations in form of electromagnetic fields, and reads out the electron position from a screen [8]. The accurate description of any of these steps is very involved and in many cases not even explored quantum mechanically. In order to perform a metrological analysis for such a machine, we assume the simplified model sketched in Fig. 4.1 and discussed in this section.

The electron is described by a pure motional quantum state that interacts with a magnetic moment on the beam axis. For this interaction, we distinguish two scenarios: In the first scenario, the magnetic moment is static and the electron state remains pure. In the second case, the magnetic moment is sourced by a spin-1/2 and evolves together with the electron wave function. Since we are agnostic to the state of the spin, we have to consider the reduced state of the electron, which is mixed. After the interaction, we want to measure the electron in order to estimate the parameter ϑ that was encoded in the magnetic moment. Different measurements can be realized by applying a unitary transformation $\hat{U}$ in form of electron optics, before the electron hits the screen. Any measurement basis can be achieved by a proper choice of $\hat{U}$. However, the realizable unitaries in an experiment are often very limited.

This section is structured in the following way: We derive the general interaction

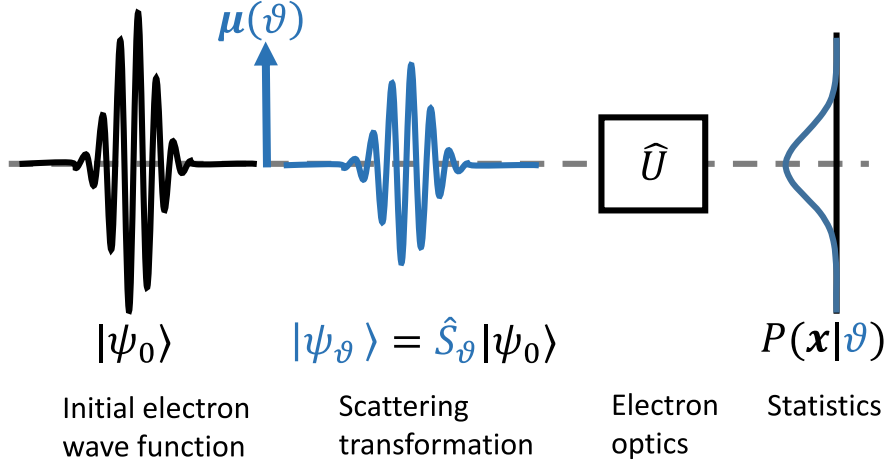


Figure 4.1.: Sketch of the general setting for sensing magnetic moments with an electron microscope: The electron wave packet, represented by the motional state $|\psi_0\rangle$, interacts with a magnetic moment $\boldsymbol{\mu}$ that is placed on the beam axis of the microscope. The parameter ϑ , which characterizes the magnetic moment $\boldsymbol{\mu}$, is encoded in the motional state of the electron via the scattering transformation $|\psi_\vartheta\rangle = \hat{S}_\vartheta |\psi_0\rangle$. After the application of some electron optics $\hat{U}$, e.g. lenses, the electron hits the screen at a position $\mathbf{x}$, distributed according to the likelihood function, $P(\mathbf{x}|\vartheta) = |\langle \mathbf{x} | \hat{U} \hat{S}_\vartheta |\psi_0\rangle|^2$. In case that the magnetic moment is also a dynamics quantum system, $|\psi_0\rangle$ has to be replaced by an initial state of both systems, $|\psi_M, \psi_S\rangle$, and the likelihood has to be calculated from the reduced state $\hat{\rho}_\vartheta = \hat{U} \text{tr}_S \{ \hat{S}_\vartheta |\psi_M, \psi_S\rangle \langle \psi_M, \psi_S| \hat{S}_\vartheta^\dagger \} \hat{U}^\dagger$. After the interaction, an estimate for the parameter ϑ is constructed from the , in general, parameter dependent measurement statistics. The choice of the unitary $\hat{U}$ effectively changes the basis, in which we perform the measurement. If all unitaries were realizable, we could perform any measurement. In practice, however, the set is very limit.

Hamiltonian from the minimal coupling principle for electromagnetic interactions in Subsection 4.2.1. In Subsection 4.2.2, we derive the scattering operator for the static magnetic moment by employing the so-called slowly-varying envelope approximation. The following Subsection 4.2.3 is focused on the derivation of the scattering operator with backaction using the same approximations. Finally, we discuss the angular momentum eigenbasis in Subsection 4.2.4.

4.2.1. General Hamiltonian

For our model of an electron microscope experiment, depicted in Fig. 4.1, we now derive the Hamiltonian describing the interaction between a probe electron and a sample, which is either given by a static magnetic moment or sourced by a quantum mechanical spin. The former case omits any backaction of the electron on the sample, as typically considered for sizable magnetic materials. In the latter case, the electron and the sample spin will become entangled, as we will discuss later. The interaction is mediated by the electromagnetic field, described by the scalar and vector potentials $(\Phi(\mathbf{x}), \mathbf{A}(\mathbf{x}))$. For the quantum sample, we will restrict to the case of a spin-1/2 system at fixed position, to which we associate the magnetic moment operator, $\boldsymbol{\mu} = \mu \hat{\boldsymbol{\sigma}}$. The strength of the magnetic moment would be given by Bohr magneton, $\mu_B = e\hbar/(2m_e)$, in the case of an unpaired atomic electron spin, or by the nuclear magneton in the case of a nuclear spin. We assume that the spin is aligned with respect to a magnetic field along the axis $\mathbf{r}$, which results in two bare energy levels separated by the Larmor frequency ω_0 . This scenario is well described by the *minimal coupling* Hamiltonian [64],

$$\hat{H} = \frac{1}{2m} (\hat{\mathbf{p}} - (-e)\mathbf{A}(\hat{\mathbf{x}}))^2 + (-e)\Phi(\hat{\mathbf{x}}) + \hbar\omega_0 (\mathbf{r} \cdot \hat{\boldsymbol{\sigma}}), \quad (4.2.1)$$

where e is the elementary charge and m is the mass of the probe electron with position and momentum operators, $\hat{\mathbf{x}}$, $\hat{\mathbf{p}}$. In Coulomb gauge, the electromagnetic potentials sourced by a magnetic moment $\boldsymbol{\mu}$ are,

$$\Phi(\mathbf{x}) = 0 \quad (4.2.2)$$

$$\mathbf{A}(\mathbf{x}) = \frac{\mu_0}{4\pi} \frac{\boldsymbol{\mu} \times \mathbf{x}}{|\mathbf{x}|^3}. \quad (4.2.3)$$

Here, the magnetic moment $\boldsymbol{\mu}$ is either a static vector or a quantum operator $\sim \hat{\boldsymbol{\sigma}}$, but we shall omit the operator hat in the general case.

With the expressions for the potentials at hand, we first expand the kinetic energy term in (4.2.1),

$$\frac{1}{2m} (\hat{\mathbf{p}} - (-e)\mathbf{A}(\hat{\mathbf{x}}))^2 = \frac{\hat{\mathbf{p}}^2}{2m} + \frac{e}{2m} \{\mathbf{A}(\hat{\mathbf{x}}), \hat{\mathbf{p}}\} + \frac{e^2}{2m} \mathbf{A}^2(\hat{\mathbf{x}}), \quad (4.2.4)$$

The anti-commutator between the vector potential $\mathbf{A}(\hat{\mathbf{x}})$ and the canonical momentum $\hat{\mathbf{p}}$ can be simplified using the identity $\hat{\mathbf{A}} \cdot \hat{\mathbf{p}} = \hat{\mathbf{p}} \cdot \hat{\mathbf{A}}$, as shown in Appendix D.1.1. Moreover,

applying the cyclic invariance of the triple product¹ while respecting the operator order,

$$\left\{ \hat{\mathbf{A}}(\hat{\mathbf{x}}), \hat{\mathbf{p}} \right\} = 2\hat{\mathbf{A}}(\hat{\mathbf{x}}) \cdot \hat{\mathbf{p}} = 2\frac{\mu_0}{4\pi} \boldsymbol{\mu} \cdot \left(\frac{\hat{\mathbf{x}}}{|\hat{\mathbf{x}}|^3} \times \hat{\mathbf{p}} \right). \quad (4.2.5)$$

We can now argue that the $\mathbf{A}^2$ term is negligible compared to the anti-commutator. To this end, we identify the angular momentum operator $\hat{\mathbf{L}} = \hat{\mathbf{x}} \times \hat{\mathbf{p}}$ and introduce the *Bohr magneton* μ_B and the *classical electron radius* r_e ,

$$\mu_B = \frac{e}{2m} \hbar \approx 9.3 \cdot 10^{-24} \frac{\text{J}}{\text{T}} \quad (4.2.6)$$

$$r_e = \frac{\mu_0}{4\pi} \frac{e^2}{m} \approx 3 \cdot 10^{-15} \text{ m}. \quad (4.2.7)$$

In these natural units, we find,

$$\frac{e}{2m} \left\{ \hat{\mathbf{A}}(\hat{\mathbf{x}}), \hat{\mathbf{p}} \right\} = \frac{1}{2m} \left(\frac{\hbar}{r_e} \right)^2 \left(\frac{\boldsymbol{\mu}}{\mu_B} \right) \left(\frac{r_e}{|\hat{\mathbf{x}}|} \right)^3 \left(\frac{\hat{\mathbf{L}}}{\hbar} \right). \quad (4.2.8)$$

The $\hat{\mathbf{A}}^2$ term, on the other hand, can be expressed as,

$$\frac{e^2}{2m} \mathbf{A}^2(\hat{\mathbf{x}}) = \frac{1}{2m} \left(\frac{\hbar}{r_e} \right)^2 \frac{1}{4} \left(\frac{r_e}{|\hat{\mathbf{x}}|} \right)^4 \left(\frac{\boldsymbol{\mu}}{\mu_B} \right)^2 \left(1 - \left(\frac{\boldsymbol{\mu} \cdot \hat{\mathbf{x}}}{|\boldsymbol{\mu}| |\hat{\mathbf{x}}|} \right)^2 \right). \quad (4.2.9)$$

Note that, while the relevant length scale of the interaction is the classical electron radius r_e , a realistic length scale of the electron wavefunction is at least five orders of magnitude larger than r_e . Thus, due to the additional factor of $r_e/|\hat{\mathbf{x}}|$, the $\mathbf{A}^2$ term is suppressed by a factor of 10^{-5} compared to the anti-commutator. For this reason, we drop the $\mathbf{A}^2$ term and arrive at the Hamiltonian,

$$\hat{H} = \frac{\hat{\mathbf{p}}^2}{2m} + \hbar\omega_0(\mathbf{r} \cdot \hat{\boldsymbol{\sigma}}) + \frac{\hbar r_e}{2m} \left(\frac{\boldsymbol{\mu}}{\mu_B} \right) \cdot \left(\frac{\hat{\mathbf{x}}}{|\hat{\mathbf{x}}|^3} \times \hat{\mathbf{p}} \right). \quad (4.2.10)$$

The magnetic moment, used in (4.2.3), is perfectly localized at the origin, where it creates numerous mathematical problems in the form of divergences. A solution lies in the more physical assumption of a spatially extended source of the magnetic moment, e.g., due to the finite wavefunction spread of the particle carrying the magnetic spin. Inspired by the ground state of a harmonic oscillator, we choose a Gaussian probability density $P_G(\mathbf{y})$ with zero mean and standard deviation Δ . It smears out the magnetic source and thus the vector potential,

$$\mathbf{A}_G(\mathbf{x}) = \int d^3\mathbf{y} P_G(\mathbf{y}) \mathbf{A}(\mathbf{x} - \mathbf{y}). \quad (4.2.11)$$

¹Recall $\mathbf{a} \cdot (\mathbf{b} \times \mathbf{c}) = \mathbf{b} \cdot (\mathbf{c} \times \mathbf{a}) = \mathbf{c} \cdot (\mathbf{a} \times \mathbf{b})$.

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The solution of the integral, derived in Appendix D.1.2, is the original vector potential multiplied by a radial function $\mathbf{A}_G(\mathbf{x}) = F(|\mathbf{x}|)\mathbf{A}(\mathbf{x})$. The function reads,

$$F(|\mathbf{x}|) = \operatorname{erf}\left(\frac{|\mathbf{x}|}{\sqrt{2}\Delta}\right) - \sqrt{\frac{2}{\pi}} \frac{|\mathbf{x}|}{\Delta} \exp\left(-\frac{1}{2}\left(\frac{|\mathbf{x}|}{\Delta}\right)^2\right). \quad (4.2.12)$$

Taylor expansion shows that F converges to zero sufficiently fast so as to alleviate the singularity of $\mathbf{A}$,

$$F(|\mathbf{x}|) = \frac{1}{3}\sqrt{\frac{2}{\pi}}\left|\frac{\mathbf{x}}{\Delta}\right|^3 + \mathcal{O}\left(\left|\frac{\mathbf{x}}{\Delta}\right|^5\right). \quad (4.2.13)$$

For large arguments $|\mathbf{x}| \gg \Delta$, on the other hand, the function approaches unity, $F(\mathbf{x}) \rightarrow 1$. Hence, the regularized vector potential $\mathbf{A}_G$ approximates the original $\mathbf{A}$ for large arguments, but does not diverge at the origin.

Going through the same arguments as for unregularized Hamiltonian ((4.2.10)), we find the regularized Hamiltonian,

$$\hat{H} = \frac{\hat{\mathbf{p}}^2}{2m} + \hbar\omega_0(\mathbf{r} \cdot \hat{\boldsymbol{\sigma}}) + \frac{\hbar r_e}{2m} \left(\frac{\boldsymbol{\mu}}{\mu_B}\right) \cdot \left(F(|\hat{\mathbf{x}}|) \frac{\hat{\mathbf{x}}}{|\hat{\mathbf{x}}|^3} \times \hat{\mathbf{p}}\right). \quad (4.2.14)$$

In the following chapter, we will use two versions of (4.2.14): One backaction-less version, in which the magnetic moment is static. This means that there is no dynamical spin degree of freedom, $\omega_0 = 0$, and the magnetic moment points in a fixed direction $\boldsymbol{\mu} = \mu\mathbf{n}$,

$$\hat{H}_N = \frac{\hat{\mathbf{p}}^2}{2m} + \frac{\hbar r_e}{2m} \left(\frac{\mu}{\mu_B}\right) \mathbf{n} \cdot \left(F(|\hat{\mathbf{x}}|) \frac{\hat{\mathbf{x}}}{|\hat{\mathbf{x}}|^3} \times \hat{\mathbf{p}}\right). \quad (4.2.15)$$

In the case with quantum backaction, the magnetic moment is sourced by a quantum spin, $\boldsymbol{\mu} = \mu\hat{\boldsymbol{\sigma}}$, and the Hamiltonian reads,

$$\hat{H}_{BA} = \frac{\hat{\mathbf{p}}^2}{2m} + \hbar\omega_0(\mathbf{r} \cdot \hat{\boldsymbol{\sigma}}) + \frac{\hbar r_e}{2m} \left(\frac{\mu}{\mu_B}\right) \hat{\boldsymbol{\sigma}} \cdot \left(F(|\hat{\mathbf{x}}|) \frac{\hat{\mathbf{x}}}{|\hat{\mathbf{x}}|^3} \times \hat{\mathbf{p}}\right). \quad (4.2.16)$$

4.2.2. Scattering operator without backaction

Based on the Hamiltonian $\hat{H}_N$ for an electron interacting with a static magnetic moment, we derive the Scattering operator that connects the electron wave function before and after an interaction. The forward momentum along the beam axis $\mathbf{p}_0$ is much larger than the deflected momenta $\mathbf{p}_\perp$ in an electron microscope. For this reason, we employ the so-called *slowly varying envelope approximation* (SVEA) [175] that is well suited for such a scenario.

In order to apply the SVEA, it is convenient to express the state $|\psi(t)\rangle$ by a slowly varying *envelope* state $|u(t)\rangle$ boosted with the forward momentum $\mathbf{p}_0$ and modified with

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a time dependent phase $E_0 t$,

$$|\psi(t)\rangle = \exp\left(i\frac{\mathbf{p}_0 \cdot \hat{\mathbf{x}} - E_0 t}{\hbar}\right) |u(t)\rangle. \quad (4.2.17)$$

The Schrödinger equation, $i\hbar |\dot{\psi}(t)\rangle = \hat{H}_N |\psi(t)\rangle$, can be transformed for the envelope state as,

$$\begin{aligned} i\hbar \frac{d}{dt} |u(t)\rangle &= \left[\exp\left(-i\frac{\mathbf{p}_0 \cdot \hat{\mathbf{x}}}{\hbar}\right) \hat{H}_N \exp\left(i\frac{\mathbf{p}_0 \cdot \hat{\mathbf{x}}}{\hbar}\right) - E_0 \right] |u(t)\rangle \\ &= \left[\frac{(\hat{\mathbf{p}} + \mathbf{p}_0)^2}{2m} + \frac{\hbar r_e}{2m} \left(\frac{\mu}{\mu_B}\right) \mathbf{n} \cdot \left(F(|\mathbf{x}|) \frac{\mathbf{x}}{|\mathbf{x}|^3} \times (\hat{\mathbf{p}} + \mathbf{p}_0) \right) - E_0 \right] |u(t)\rangle. \end{aligned} \quad (4.2.18)$$

Note that the Schrödinger equation for $|u(t)\rangle$ is still exact. Now, we assume that the supported transverse momenta are much smaller than the forward momentum,

$$\hat{\mathbf{p}}^2 \ll \frac{\mathbf{p}_0 \cdot \hat{\mathbf{p}}}{m}, \quad \& \quad |\hat{\mathbf{p}}| \ll |\mathbf{p}_0|. \quad (4.2.19)$$

These inequalities have to be understood in the sense that $\langle \mathbf{p} | u(t) \rangle$ only deviates significantly from zero, where these conditions are met. Neglecting the sub-dominant terms and setting $E_0 = \mathbf{p}_0^2/(2m)$ yields,

$$i\hbar \frac{d}{dt} |u(t)\rangle \approx \left[\frac{\mathbf{p}_0}{m} \cdot \hat{\mathbf{p}} + \frac{\hbar r_e}{2m} \left(\frac{\mu}{\mu_B}\right) \mathbf{n} \cdot \left(F(|\mathbf{x}|) \frac{\mathbf{x}}{|\mathbf{x}|^3} \times \mathbf{p}_0 \right) \right] |u(t)\rangle. \quad (4.2.20)$$

We further assume that the beam axis shows along the z -axis, $\mathbf{p}_0 = p_0 \mathbf{e}_z$, and define $v_0 = p_0/m$ arriving at the effective SVEA Schrödinger equation for the envelope state,

$$i\hbar \frac{d}{dt} |u(t)\rangle = \left[v_0 \hat{p}_z + v_0 \frac{\hbar r_e}{2} \left(\frac{\mu}{\mu_B}\right) F(|\mathbf{x}|) \frac{n_x \hat{y} - n_y \hat{x}}{|\mathbf{x}|^3} \right] |u(t)\rangle. \quad (4.2.21)$$

In order to employ scattering theory, we identify $v_0 \hat{p}_z$ as the free Hamiltonian and the remainder as the interaction $\hat{V}$. Hence, the interaction picture interaction operator is,

$$\hat{V}^{(I)}(t) = e^{i(v_0 t) \hat{p}_z / \hbar} \hat{V} e^{-i(v_0 t) \hat{p}_z / \hbar} = \frac{\hbar r_e}{2} \left(\frac{\mu}{\mu_B}\right) v_0 (n_x \hat{y} - n_y \hat{x}) \frac{F(|\hat{\mathbf{x}} + v_0 t \mathbf{e}_z|)}{|\hat{\mathbf{x}} + v_0 t \mathbf{e}_z|^3}. \quad (4.2.22)$$

The scattering operator can now be calculated by means of (1.2.26), where we can omit the time ordering since the interaction operator commutes with itself for different times,

$$\hat{S} = \exp\left(-i\frac{r_e}{2} \left(\frac{\mu}{\mu_B}\right) (n_x \hat{y} - n_y \hat{x}) \int_{-\infty}^{+\infty} v_0 dt \frac{F(|\hat{\mathbf{x}} + v_0 t \mathbf{e}_z|)}{|\hat{\mathbf{x}} + v_0 t \mathbf{e}_z|^3}\right). \quad (4.2.23)$$

Notice that the integral in the scattering operator no longer depends on the $\hat{z}$ operator, because the integration is over all possible values of z . We split the position operator

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into a part along the beam axis and a part lying in the orthogonal plane, $\hat{\mathbf{x}} = \hat{\mathbf{x}}_{\perp} + \hat{z}\mathbf{e}_z$, and use the result of the integral, calculated in Appendix D.1.3, which results in,

$$\hat{S} = \exp \left(-ir_e \left(\frac{\mu}{\mu_B} \right) \left(1 - e^{-\frac{1}{2} \left(\frac{\hat{\mathbf{x}}_{\perp}}{\Delta} \right)^2} \right) \frac{(n_x \hat{y} - n_y \hat{x})}{|\mathbf{x}_{\perp}|^2} \right). \quad (4.2.24)$$

This is a multiplicative phase in position space that only varies in the plane perpendicular to the beam axis. Using polar coordinates in this plane, $(\hat{x}, \hat{y}) = \hat{r}(\cos(\hat{\varphi}), \sin(\hat{\varphi}))$ and $(n_x, n_y) = |\mathbf{n}_{\perp}|(\cos(\varphi_n), \sin(\varphi_n))$, we can write,

$$\frac{(n_x \hat{y} - n_y \hat{x})}{|\mathbf{x}_{\perp}|} = |\mathbf{n}_{\perp}|(\cos(\varphi_n) \sin(\hat{\varphi}) - \sin(\varphi_n) \cos(\hat{\varphi})) = |\mathbf{n}_{\perp}| \sin(\hat{\varphi} - \varphi_n). \quad (4.2.25)$$

Without loss of generality, we can set $\varphi_n = 0$, which corresponds to a rotation of our coordinate system with respect to the orientation of the magnetic moment. Assuming that the size of the wave functions is of the same order of magnitude as the regulator length scale Δ or larger, it is convenient to relate all lengths scales to Δ . We arrive at the scattering operator,

$$\hat{S}_{\vartheta} = \exp[-i\vartheta |\mathbf{n}_{\perp}| g(\hat{r}) \sin(\hat{\varphi})]. \quad (4.2.26)$$

Here, we introduce a radial function,

$$g(\hat{r}) = \left(1 - e^{-\frac{1}{2} \left(\frac{\hat{r}}{\Delta} \right)^2} \right) \frac{\Delta}{\hat{r}} \quad (4.2.27)$$

and the dimensionless parameter $\vartheta = \frac{r_e \mu}{\Delta \mu_B}$ that encodes the strength of the magnetic moment. Reasonable assumptions of the order of magnitude of the relevant quantities are $\mu/\mu_B \sim 1$ and $r_e/\Delta \sim 10^{-5}$. Thus, $\vartheta \sim 10^{-5}$ can be expected.

The scattering operator in (4.2.26) is derived for the envelope state $|u(t)\rangle$. However, it can be seen that this operator commutes with the unitary transformation (4.2.17) that mediates between the boosted and unboosted state,

$$\left[\hat{S}_{\vartheta}, \exp \left(i \frac{\mathbf{p}_0 \cdot \hat{\mathbf{x}} - E_0 t}{\hbar} \right) \right] = 0. \quad (4.2.28)$$

Conclusively, we can apply the scattering operator directly to the original state $|\psi(t)\rangle$.

In this subsection, we have seen that, under the SVEA approximation, the scattering operator acts solely on the two-dimensional function space defined in the x-y plane. We use this to reduce the three-dimensional problem to a two-dimensional problem, where the incident plane is mapped onto the measurement screen, as depicted in Fig. 4.2. This resembles a typical scenario for the diffraction of electromagnetic waves: Huygens' principle states that any point of a wavefront can be considered as a point source generating the next wavefront, which is the basis for the Fraunhofer diffraction [176] of matter waves. In general, this is not applicable to quantum mechanics since the three-dimensional wavefunction has to be normalized. In this case however, the norm of the two-dimensional slices is already preserved by the scattering operator, justifying our

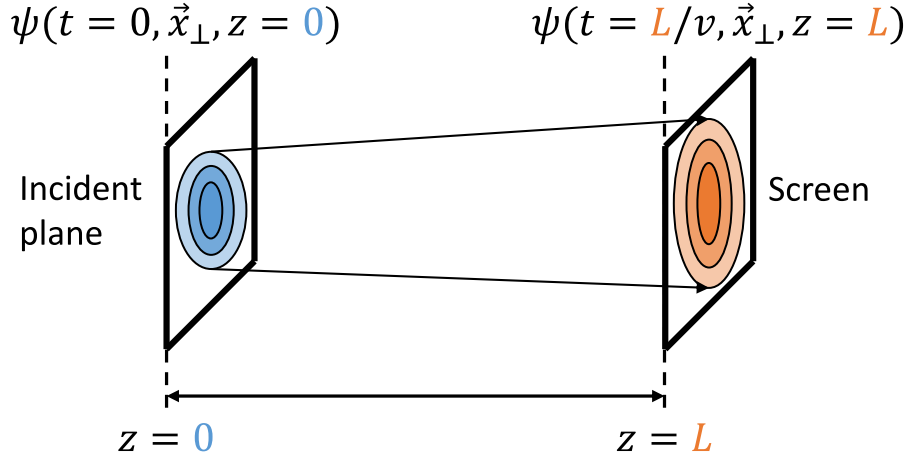


Figure 4.2.: Effective evolution of the electron wave function in an TEM: Instead of the full three-dimensional description, we describe the evolution and diffraction of the electron beam as a mapping from a incident plane at $z=0$ onto the plane of the screen at $z=L$. This procedure is possible because of the SVEA approximation that yields a unitary operator on the function space in the x - y plane.

approach.

A deep connection between the scattering operator and the angular momentum of the planar wavefunction becomes evident after applying the *Jacobi-Anger expansion* [177] in expression (4.2.26),

$$\hat{S}_\vartheta = \sum_{k \in \mathbb{Z}} (-1)^k J_k(\vartheta |\mathbf{n}_\perp| g(\hat{r})) e^{ik\hat{\varphi}}, \quad (4.2.29)$$

where the J_k denote Bessel functions. Hence in the case of a static magnetic sample, the scattering operator maps a state with zero angular momentum onto a superposition of all angular momenta weighted by radius-dependent Bessel functions.

4.2.3. Scattering operator with backaction

In the preceding subsection 4.2.2, we derived a scattering operator for a static magnetic moment constant in time. For a small sample, however, this ignores the fact that the magnetic moment itself is affected by the presence of the electron. This, on the other hand, alters the evolution of the electron. In this subsection, we take the dynamic nature of the magnetic moment into account by modeling it as a quantum spin. We derive and discuss the scattering operator for this case.

Recall the backaction Hamiltonian including the spin dynamics from Subsection 4.2.1,

$$\hat{H}_{\text{BA}} = \frac{\hat{\mathbf{p}}^2}{2m} + \hbar\omega_0(\mathbf{r} \cdot \hat{\sigma}_z) + \frac{\hbar r_e}{2m} \left(\frac{\mu}{\mu_B} \right) \hat{\boldsymbol{\sigma}} \cdot \left(F(|\hat{\mathbf{x}}|) \frac{\hat{\mathbf{x}}}{|\hat{\mathbf{x}}|^3} \times \hat{\mathbf{p}} \right). \quad (4.2.30)$$

Analogous to the case without backaction we can describe our state $|\psi(t)\rangle$, now including

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the electron motion and the state of the sample spin, as a boosted envelope state $|u(t)\rangle$,

$$|\psi(t)\rangle = \exp\left(i\frac{\mathbf{p}_0 \cdot \hat{\mathbf{x}} - E_0 t}{\hbar}\right) |u(t)\rangle. \quad (4.2.31)$$

Under the same approximations, we find the effective Hamiltonian for the evolution of the envelope state,

$$\hat{H}_{\text{BA}}^{\text{eff}} = \underbrace{v_0 \hat{p}_z + \frac{\hbar \omega_0}{2} (\mathbf{r} \cdot \hat{\boldsymbol{\sigma}})}_{=\hat{H}_0} + \underbrace{\frac{\hbar r_e}{2} \left(\frac{\mu}{\mu_B}\right) v_0 \left(F(|\hat{\mathbf{x}}|) \frac{\hat{\boldsymbol{\sigma}} \cdot (\hat{\mathbf{x}} \times \mathbf{e}_z)}{|\hat{\mathbf{x}}|^3}\right)}_{=\hat{V}}. \quad (4.2.32)$$

In the interaction picture, the Hamiltonian reduces to the transformed interaction term,

$$\begin{aligned} \hat{V}^{(I)}(t) &= e^{i\hat{H}_0 t/\hbar} \hat{V} e^{-i\hat{H}_0 t/\hbar} \\ &= \frac{\hbar r_e}{2} \left(\frac{\mu}{\mu_B}\right) v_0 F(|\hat{\mathbf{x}} + v_0 t \mathbf{e}_z|) \frac{(R(\mathbf{r}, \omega_0 t) \hat{\boldsymbol{\sigma}}) \cdot (\hat{\mathbf{x}} \times \mathbf{e}_z)}{|\hat{\mathbf{x}} + v_0 t \mathbf{e}_z|^3} \\ &= \frac{\hbar r_e}{2} \left(\frac{\mu}{\mu_B}\right) v_0 \hat{\boldsymbol{\sigma}} \cdot \frac{F(|\hat{\mathbf{x}} + v_0 t \mathbf{e}_z|) R(\mathbf{r}, -\omega_0 t)}{|\hat{\mathbf{x}} + v_0 t \mathbf{e}_z|^3} \begin{pmatrix} \hat{y} \\ -\hat{x} \\ 0 \end{pmatrix}, \end{aligned} \quad (4.2.33)$$

where $R(\mathbf{r}, \omega_0 t)$ is a rotation matrix of angle $\omega_0 t$ around the $\mathbf{r}$ -axis. In the third line, we apply the transpose of the rotation $R(\mathbf{r}, -\omega_0 t)$ to the other side of the euclidean scalar product, $\hat{\mathbf{x}} \times \mathbf{e}_z$.

With (4.2.33) at hand, we can express the scattering operator as a time ordered exponential (1.2.26),

$$\hat{S} = \mathcal{T} \exp \left(-i \frac{r_e}{2} \left(\frac{\mu}{\mu_B}\right) \hat{\boldsymbol{\sigma}} \cdot \int_{-\infty}^{+\infty} v_0 dt \frac{F(|\hat{\mathbf{x}} + v_0 t \mathbf{e}_z|) R(\mathbf{r}, -\omega_0 t)}{|\hat{\mathbf{x}} + v_0 t \mathbf{e}_z|^3} \begin{pmatrix} \hat{y} \\ -\hat{x} \\ 0 \end{pmatrix} \right). \quad (4.2.34)$$

The time integral can be rewritten as,

$$\begin{aligned} &\int_{-\infty}^{+\infty} v_0 dt \frac{F(|\hat{\mathbf{x}} + v_0 t \mathbf{e}_z|) R(\mathbf{r}, -\omega_0 t)}{|\hat{\mathbf{x}} + v_0 t \mathbf{e}_z|^3} \\ &= R\left(\mathbf{r}, \frac{\omega_0}{v_0} \hat{z}\right) \int_{-\infty}^{+\infty} dz \frac{F(|\hat{\mathbf{x}}_{\perp} + z \mathbf{e}_z|) R\left(\mathbf{r}, \frac{\omega_0}{v_0} (-z)\right)}{|\hat{\mathbf{x}}_{\perp} + z \mathbf{e}_z|^3}, \end{aligned} \quad (4.2.35)$$

see Appendix D.1.4 for details. Here, the rotation matrix R appears twice: On the one hand, the leading term that depends on the $\hat{z}$ operator, $R(\mathbf{r}, \omega_0 \hat{z}/v_0)$, accounts for the fact that the magnetic moment precesses in time with respect to the free Hamiltonian. Thus, every z component of the wave packet sees a differently oriented magnetic moment. If the wavepacket is much shorter than the characteristic length scale v_0/ω_0 , this rotation

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can be neglected, $R(\mathbf{r}, \omega_0 \hat{z}/v_0) \approx \mathbb{1}$. On the other hand, the rotation matrix appears inside the integral. However, given that z is large, the integrand decays on a scale of σ , where σ is the spread of the wavefunction in the plane perpendicular to the beam axis. If we assume $\sigma \ll v_0/\omega_0$, the integrand vanishes before the rotation matrix can deviate significantly from identity. We therefore set $R(\mathbf{r}, -\omega_0 \hat{z}/v_0) \approx \mathbb{1}$. In both cases, neglecting the rotation matrix requires the length scale v_0/ω_0 to be large. Since ω_0 is proportional to a magnetic bias field, which can theoretically be chosen arbitrarily small, we assume both conditions to be met.

Having shown that the rotation matrices can be neglected, we recognize the remaining integral as an already known expression from the case without backaction, calculated in D.1.3. We see that the time ordering can be omitted and find the scattering operator,

$$\begin{aligned} \hat{S}_\vartheta &= \exp \left(-i \left(\frac{r_e \mu}{\Delta \mu_B} \right) \frac{\Delta}{|\hat{\mathbf{x}}_\perp|} \left(1 - e^{-\frac{1}{2} \left(\frac{\hat{\mathbf{x}}_\perp}{\Delta} \right)^2} \right) \frac{\hat{\sigma}_x \hat{y} - \hat{\sigma}_y \hat{x}}{|\hat{\mathbf{x}}_\perp|} \right) \\ &= \exp \left(-i \vartheta g(\hat{r}) [\hat{\sigma}_x \sin(\hat{\varphi}) - \hat{\sigma}_y \cos(\hat{\varphi})] \right). \end{aligned} \quad (4.2.36)$$

Here we once again make use of the previous notation from (4.2.26),

$$\vartheta = \frac{r_e \mu}{\Delta \mu_B} \quad \& \quad g(r) = \frac{\Delta}{r} \left(1 - e^{-\frac{1}{2} \left(\frac{r}{\Delta} \right)^2} \right). \quad (4.2.37)$$

Expanding the trigonometric functions into complex exponentials, yields an alternative form of the angular part of the scattering operator,

$$\hat{\sigma}_x \sin(\hat{\varphi}) - \hat{\sigma}_y \cos(\hat{\varphi}) = i (\hat{\sigma}_+ e^{-i\hat{\varphi}} - \hat{\sigma}_- e^{i\hat{\varphi}}). \quad (4.2.38)$$

In this form, the scattering operator reads,

$$\hat{S}_\vartheta = \exp \left(\vartheta g(\hat{r}) [\hat{\sigma}_+ e^{-i\hat{\varphi}} - \hat{\sigma}_- e^{i\hat{\varphi}}] \right). \quad (4.2.39)$$

This operator exponential simplifies further due to the fact that the angular part squares to the negative identity operator,

$$[e^{-i\hat{\varphi}} \hat{\sigma}_+ - e^{i\hat{\varphi}} \hat{\sigma}_-]^2 = -\{\hat{\sigma}_+, \hat{\sigma}_-\} = -\mathbb{1}. \quad (4.2.40)$$

A power series expansion then leaves us with,

$$\hat{S}_\vartheta = \cos(\vartheta g(\hat{r})) + \sin(\vartheta g(\hat{r})) [e^{-i\hat{\varphi}} \hat{\sigma}_+ - e^{i\hat{\varphi}} \hat{\sigma}_-], \quad (4.2.41)$$

as we discuss in Appendix D.1.5. This final form embodies angular momentum conservation as the interaction of the electron with the spin can at most transfer $\pm \hbar$ of orbital angular momentum to the electron wavefunction. Compare this to the previous case without backaction (4.2.29), which supports the transfer of any multiple of $\hbar$ in orbital angular momentum.

4.2.4. Angular momentum measurements

The scattering operators in both cases, with and without backaction, can be phrased in terms of the operators $e^{\pm i\hat{\varphi}} = (\hat{x} \pm i\hat{y})/|\hat{\mathbf{x}}_{\perp}|$. We will now exploit this structure by constructing a basis that resembles the angular momentum basis for the planar rotor, see Chapter 2. On these states, the operators $e^{\pm i\hat{\varphi}}$ act like raising and lowering operators.

Recall that the radius operator $\hat{r} = \sqrt{\hat{x}^2 + \hat{y}^2}$ commutes with the angular momentum operator $\hat{L}_z$. Thus, there is a basis of mutual eigenstates of $\hat{r}$ and $\hat{L}_z$, $|r, k\rangle$, in which $r \in [0, \infty)$ is a continuous label and $k \in \mathbb{Z}$ is discrete. As we discuss in Appendix D.2.2, the basis is orthonormal and complete,

$$\langle r', k' | r, k \rangle = \frac{1}{r} \delta^{(1)}(r' - r) \delta_{k'k} \quad (4.2.42)$$

$$\int_0^\infty dr \, r \sum_{k \in \mathbb{Z}} |r, k\rangle \langle r, k| = \mathbb{1}, \quad (4.2.43)$$

and translates to position space by,

$$\langle \mathbf{x}_{\perp} | r, k \rangle = \frac{1}{r} \delta^{(1)}(|\mathbf{x}_{\perp}| - r) \frac{1}{\sqrt{2\pi}} e^{ik\varphi_x}. \quad (4.2.44)$$

From the position space representation, we directly see how $e^{\pm i\hat{\varphi}}$ acts on the angular basis,

$$e^{\pm i\hat{\varphi}} |r, k\rangle = |r, k \pm 1\rangle. \quad (4.2.45)$$

Therefore, we refer to $e^{\pm i\hat{\varphi}}$ as *angular momentum kicks*.

We can also expand operators in the new basis, where we are especially interested in radial multiplications and angular momentum kicks,

$$f(\hat{r}) e^{il\hat{\varphi}} = \int_0^\infty dr \, r \sum_{k \in \mathbb{Z}} f(r) |r, k + l\rangle \langle r, k|. \quad (4.2.46)$$

Finally, we calculate the transition amplitudes between the new angular basis and momentum eigen states. To this end, we once more use the Jacobi-Anger expansion of plane waves [177],

$$\langle \mathbf{x}_{\perp} | \mathbf{p}_{\perp} \rangle = \frac{1}{2\pi\hbar} \sum_{n \in \mathbb{Z}} i^n J_n \left(\frac{|\mathbf{x}_{\perp}| |\mathbf{p}_{\perp}|}{\hbar} \right) e^{in(\varphi_x - \varphi_p)}. \quad (4.2.47)$$

Inserting a resolution of identity and performing the integral in polar coordinates, yields

the transition amplitude,

$$\begin{aligned}
 \langle r, k | \mathbf{p}_\perp \rangle &= \int d^2 \mathbf{x}_\perp \langle r, k | \mathbf{x}_\perp \rangle \langle \mathbf{x}_\perp | \mathbf{p}_\perp \rangle \\
 &= \frac{1}{\sqrt{2\pi^3} \hbar} \int d^2 \mathbf{x}_\perp \frac{1}{r} \delta^{(1)}(r - |\mathbf{x}_\perp|) e^{-ik\varphi_x} \sum_{n \in \mathbb{Z}} i^n J_n \left(\frac{|\mathbf{x}_\perp| |\mathbf{p}_\perp|}{\hbar} \right) e^{in(\varphi_x - \varphi_p)} \\
 &= \frac{1}{\sqrt{2\pi} \hbar} \sum_{n \in \mathbb{Z}} i^n e^{-in\varphi_p} J_n \left(\frac{r |\mathbf{p}_\perp|}{\hbar} \right) \int_0^{2\pi} \frac{d\varphi_x}{2\pi} e^{i(n-k)\varphi_x} \\
 &= \frac{i^k}{\hbar} J_k \left(\frac{r |\mathbf{p}_\perp|}{\hbar} \right) \frac{e^{-ik\varphi_p}}{\sqrt{2\pi}}.
 \end{aligned} \tag{4.2.48}$$

The presented basis is not only useful to describe the effect of the scattering operators, with and without backaction, but also to define measurements of angular momentum for optimal sensing.

4.3. Sensing without backaction

Building on the scattering operator that we derived in Subsection 4.2.2, we now investigate the task of quantum sensing for the static magnetic moment. The advantage in this case is that the motional state of the electron remains pure, which makes analytical calculations mostly tractable. In Subsection 4.3.1, we begin with the calculation of the QFI that, as an upper bound for the CFI, serves as a point of reference. We proceed with the CFI for momentum measurements in Subsection 4.3.2 and consider angular momentum measurements in Subsection 4.3.3.

4.3.1. Derivation of the quantum Fisher information

The QFI is an important quantity that characterizes the amount of information about and unknown parameter that is contained in a quantum state, as discussed in Subsection 1.5.2. Here, we will calculate and discuss the QFI of the motional state of an electron after scattering off a magnetic moment,

$$|\psi_\vartheta\rangle = \hat{S}_\vartheta |\psi_0\rangle. \tag{4.3.1}$$

Since this scattering transformation introduces a ϑ -proportional phase, we can apply the results for the phase estimation case, provided in Subsection 1.5.2. Therefore, the QFI $\mathcal{J}_{\text{NB}}$ becomes parameter independent and reads,

$$\begin{aligned}
 \mathcal{J}_{\text{NB}} &= 4\mathbb{V}_{|\psi_\vartheta\rangle} [-|\mathbf{n}_\perp| g(\hat{r}) \sin(\hat{\varphi})] \\
 &= 4|\mathbf{n}_\perp|^2 \left(\langle \psi_0 | g(\hat{r})^2 \sin^2(\hat{\varphi}) | \psi_0 \rangle - (\langle \psi_0 | g(\hat{r}) \sin(\hat{\varphi}) | \psi_0 \rangle)^2 \right).
 \end{aligned} \tag{4.3.2}$$

This expression simplifies significantly when we consider radial wavefunctions, i.e., wave-

functions with no dependence on the azimuthal angle, as incoming states. The second term then vanishes due to the averaging of the sine,

$$\langle \psi_0 | g(\hat{r}) \sin(\hat{\varphi}) | \psi_0 \rangle = \int_0^\infty dr \, r g(\hat{r}) |\psi_0(r)|^2 \underbrace{\int_0^{2\pi} d\varphi \, \sin(\varphi)}_{=0} = 0. \quad (4.3.3)$$

The first term reduces to

$$\begin{aligned} \langle \psi_0 | g(\hat{r})^2 \sin^2(\hat{\varphi}) | \psi_0 \rangle &= \int_0^\infty dr \, r g(\hat{r})^2 |\psi_0(r)|^2 \underbrace{\int_0^{2\pi} d\varphi \, \sin^2(\varphi)}_{=\pi} \\ &= \frac{1}{2} \int_0^\infty dr \, r g(\hat{r})^2 |\psi_0(r)|^2 \int_0^{2\pi} d\varphi \\ &= \frac{1}{2} \langle \psi_0 | g(\hat{r})^2 | \psi_0 \rangle. \end{aligned} \quad (4.3.4)$$

Combining the two results yields a compact formula for the QFI of radial wavefunctions,

$$\mathcal{J}_{\text{NB}} = 2|\mathbf{n}_\perp|^2 \langle \psi_0 | g(\hat{r})^2 | \psi_0 \rangle = |\mathbf{n}_\perp|^2 \mathcal{J}_{\text{NB}}^{\text{max}}. \quad (4.3.5)$$

Equation (4.3.5) plays a central role in this chapter for two reasons: On the one hand, the QFI is the optimal sensitivity for parameter ϑ , which we will use to find the optimal measurement basis. On the other hand, the expression $\mathcal{J}_{\text{NB}}^{\text{max}} = 2 \langle \psi_0 | g(\hat{r})^2 | \psi_0 \rangle$ connects to the classical Fisher information evaluated in other bases, as we will show in subsections 4.3.2 and 4.3.3. Since the overall formula (4.3.5) is independent of the specific choice of the initial state $|\psi_0\rangle$ and the function $g(\hat{r})$, this connection holds for general initial state and $g(\hat{r})$ as well.

In order to better understand equation (4.3.5), we now choose a centered radial Gaussian with standard deviation σ as the initial state,

$$\langle \mathbf{x}_\perp | \psi_0 \rangle = \psi_0(|\mathbf{x}_\perp|) = \frac{1}{(2\pi\sigma^2)^{1/4}} \exp\left(-\frac{1}{4} \left(\frac{\mathbf{x}_\perp}{\sigma}\right)^2\right). \quad (4.3.6)$$

Using polar coordinates and executing the trivial angular integral yields,

$$\begin{aligned} \mathcal{J}_{\text{NB}} &= 2|\mathbf{n}_\perp|^2 \left(\frac{\Delta}{\sigma}\right)^2 \int_0^\infty \frac{dr}{r} e^{-(r/\sigma)^2/2} \left(1 - e^{-(r/\Delta)^2/2}\right)^2 \\ &= \frac{2|\mathbf{n}_\perp|^2}{\chi^2} \int_0^\infty \frac{d\tau}{\tau} e^{-(\tau/\chi)^2/2} \left(1 - e^{-\tau^2/2}\right)^2 \\ &= \frac{2|\mathbf{n}_\perp|^2}{\chi^2} \ln\left(\frac{1 + \chi^2}{\sqrt{1 + 2\chi^2}}\right), \end{aligned} \quad (4.3.7)$$

where we used the solution of the integral, derived in Appendix D.2.1, in the third line.

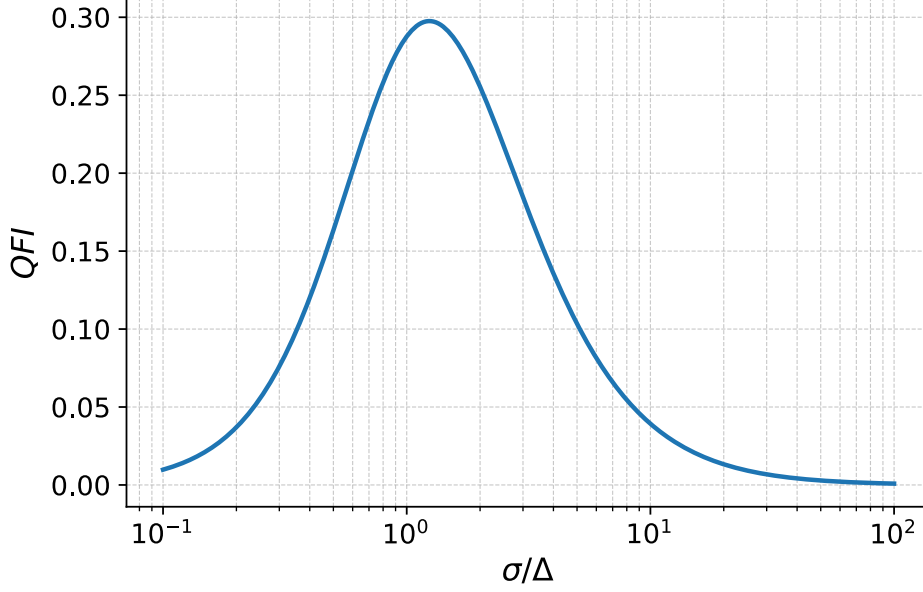


Figure 4.3.: Quantum Fisher information $\mathcal{J}_{\text{NB}}^{\text{max}}$ with respect to the dimensionless parameter ϑ for a radial Gaussian with standard deviation σ interacting with a classical magnetic moment. We find the maximal value of $\mathcal{J}_{\text{max}} \approx 0.297$ at $\sigma_{\text{max}} \approx 1.2\Delta$.

The QFI according to (4.3.7) is plotted semi-logarithmically in Fig. 4.3 versus the dimensionless standard deviation σ/Δ for a magnetic dipole that is oriented in the x - y plane, $|\mathbf{n}_{\perp}| = 1$. We see that the largest values are found for $\sigma \sim \Delta$, where the maximum of $\mathcal{J}_{\text{NB}}^{\text{max}} \approx 0.297$ is found at $\sigma_{\text{max}} \approx 1.2\Delta$. This observation is in accordance with intuition: For a large wavefunction, i.e. $\sigma \gg \Delta$, only a small fraction of the wavefunction significantly interacts with the field of the magnetic moment, which reduces the sensitivity. For small wavefunctions ($\sigma \ll \Delta$), on the other hand, the wavefunction only interacts with a fraction of the field and important features might not be imprinted on it, which also reduces sensitivity. Hence, the optimal value is where the size of the wavefunction and the sample are of the same order of magnitude.

Practically, technical limitations in focusing the electron beam force us to work in the regime of $\sigma \gtrsim 10^2\Delta$ at the right boundary of Fig. 4.3, where the QFI vanishes asymptotically like,

$$\mathcal{J}_{\text{NB}} \approx \frac{|\mathbf{n}_{\perp}|^2}{\chi^2} (2 \log(\chi) - \log(2)). \quad (4.3.8)$$

The results of this section yield the optimal measurement precision for estimating the parameter ϑ , (4.3.7). In the following subsections, 4.3.2 and 4.3.3, we will compare the classical Fisher information (CFI) in specific quantum measurements to this ideal value.

4.3.2. Optimality of momentum measurements

With the analytic formula of the QFI (4.3.7), we know the optimal value of the CFI that we can achieve by any measurement. Here, we discuss the measurements of position and momentum and show that the momentum measurement is already optimal.

Since the scattering operator imprints a position dependent phase on the state, the likelihood function obtained from a position measurement is independent from the parameter,

$$P(\mathbf{x}_\perp|\vartheta) = |\langle \mathbf{x}_\perp | \hat{S}_\vartheta | \psi_0 \rangle|^2 = |\langle \mathbf{x}_\perp | \psi_0 \rangle|^2 = P(\mathbf{x}_\perp|0), \quad \forall \vartheta. \quad (4.3.9)$$

Thus, position measurements do not yield any information about the parameter. The opposite is the case for momentum measurements, where the ultimate bound given by the QFI is reached. We show this in two different ways with slightly different conditions.

First, we consider a radial state $|\psi_0\rangle$ and a small parameter $\vartheta \ll 1$. Recall that we estimated a realistic value of $\vartheta \sim 10^{-5}$ in the derivation of the scattering operator in Subsection 4.2.2. Therefore, the following perturbative approach is well justified: we expand the post-scattering state as,

$$|\psi_\vartheta\rangle = |\psi_0\rangle + \vartheta |\partial\psi_0\rangle + \mathcal{O}(\vartheta^2), \quad (4.3.10)$$

with $|\partial\psi_0\rangle = -ig(\hat{r})\sin(\hat{\varphi})|\psi_0\rangle$. Next, we show that $\langle \mathbf{p}_\perp | \psi_0 \rangle \in \mathbb{R}$ and $\langle \mathbf{p}_\perp | \partial\psi_0 \rangle \in \mathbb{R}$. This is true since the radial function $\langle \mathbf{x}_\perp | \psi_0 \rangle$ is symmetric in x, y , which makes the Fourier transform real. For $|\partial\psi_0\rangle$, recall that $\sin(\varphi) = y/r$, which makes the function odd in y -direction. Thus, the Fourier transform is purely imaginary, which is countered by the extra factor of $-i$. The real-valued expressions are sufficient to invoke the optimality criterion, presented in Subsection 1.5.3. However, note that the optimality in this case is only guaranteed asymptotically for $\vartheta \rightarrow 0$.

The second approach is non-perturbative but has stronger requirements on the state: As we will see, the wavefunction has to be radial and real-valued, $\langle \mathbf{x}_\perp | \psi_0 \rangle = \psi_0(|\mathbf{x}_\perp|) \in \mathbb{R}$. In order to exactly evaluate $\langle \mathbf{p}_\perp | \psi_\vartheta \rangle = \langle \mathbf{p}_\perp | \hat{S}_\vartheta | \psi_0 \rangle$, we rewrite the plane-wave amplitudes $\langle \mathbf{x}_\perp | \mathbf{p}_\perp \rangle$ in polar coordinates by means of the Jacobi-Anger expansion [177],

$$\begin{aligned} \langle \mathbf{x}_\perp(r, \varphi_x) | \mathbf{p}_\perp(p, \varphi_p) \rangle &= \frac{1}{\sqrt{2\pi\hbar}} \exp\left(i \frac{\mathbf{x}_\perp \cdot \mathbf{p}_\perp}{\hbar}\right) = \frac{1}{\sqrt{2\pi\hbar}} \exp\left(i \frac{pr}{\hbar} \cos(\varphi_x - \varphi_p)\right) \\ &= \frac{1}{\sqrt{2\pi\hbar}} \sum_{k \in \mathbb{Z}} i^k J_k\left(\frac{pr}{\hbar}\right) e^{ik(\varphi_x - \varphi_p)}. \end{aligned} \quad (4.3.11)$$

Together with the similar expansion of the scattering operator (4.2.29), we can start the

evaluation by inserting a resolution of identity in position space,

$$\begin{aligned}
 \langle \mathbf{p}_\perp(p, \varphi_p) | \hat{S}_\vartheta | \psi_0 \rangle &= \int d^2 \mathbf{x}_\perp \langle \mathbf{p}_\perp | \mathbf{x}_\perp \rangle \langle \mathbf{x}_\perp | \hat{S} | \psi_0 \rangle \\
 &= \int_0^\infty \frac{dr}{2\pi\hbar} r 2\pi \left[\sum_{n \in \mathbb{Z}} (ie^{i\varphi_p})^n J_n\left(\frac{pr}{\hbar}\right) J_n(\vartheta |\mathbf{n}_\perp| g(r)) \right] \psi_0(r) \\
 &= \int_0^\infty \frac{dr}{\hbar} r J_0 \left(\sqrt{\left[\frac{pr}{\hbar}\right]^2 + [\vartheta |\mathbf{n}_\perp| g(r)]^2 - 2 \left[\frac{pr}{\hbar}\right] [\vartheta |\mathbf{n}_\perp| g(r)] \cos\left(\varphi_p + \frac{\pi}{2}\right)} \right) \psi_0(r) \\
 &= \int_0^\infty \frac{dr}{\hbar} r J_0 \left(\sqrt{\left[\frac{pr}{\hbar}\right]^2 + [\vartheta |\mathbf{n}_\perp| g(r)]^2 + 2 \left[\frac{pr}{\hbar}\right] [\vartheta |\mathbf{n}_\perp| g(r)] \sin(\varphi_p)} \right) \psi_0(r) \in \mathbb{R}.
 \end{aligned} \tag{4.3.12}$$

Executing the φ_x integration, which is immediately possible since the state is radial, collapsed the double sum in a single sum. In the third line, we expressed the sum over Bessel functions by Neuman's addition theorem for Bessel functions [177],

$$\sum_{n=-\infty}^{\infty} e^{in\alpha} J_n(x) J_n(y) = J_0 \left(\sqrt{x^2 + y^2 - 2xy \cos(\alpha)} \right). \tag{4.3.13}$$

After rephrasing the cosine as a sine, we note that the whole expression is real-valued since the wavefunction is real by assumption. From the last line of (4.3.12) it can be seen that the derivative with respect to ϑ , $\langle \mathbf{p}_\perp | \partial \psi_\vartheta \rangle$ is real-valued, too. This meets the condition of the optimality criterion from Subsection 1.5.3. Hence for an initially radial and real wavefunction, a momentum measurement yields the optimal precision to estimate ϑ .

In summary, we have shown that either a radial state to lowest order in ϑ , or a radial and real-valued state for general ϑ , is sufficient for the CFI to saturate the QFI $\mathcal{J}_{\text{NB}}$ in a momentum measurement. Notably, in both cases, neither $g(r)$ nor $\psi_0(r)$ needs to be specified further. We emphasize that the presented conditions on the initial state are sufficient but not necessary. Therefore, a more general proof might eliminate additional restrictions on the state. The fact that momentum measurements are a standard procedure in electron microscopy makes this result highly relevant for the practical estimation of magnetic moments.

4.3.3. Optimality of angular momentum measurements

In Subsection 4.3.2, we showed that position measurements do not yield any information, while momentum measurements reach the optimal precision predicted by the QFI. Although the optimality of momentum measurements effectively settles the case for static magnetic moments from a practical standpoint, the form of the scattering operator (4.2.29) has not been explicitly exploited. This scattering operator maps a radial initial state, i.e. a state without angular momentum, onto a superposition of all possible angular momentum states, where the weights depend on the parameter. Here, we will

use this to infer the parameter ϑ by measuring whether the post scattering state contains any angular momentum or not.

In the following, we present a dichotomic measurement of angular momentum and show that, for small ϑ and radial initial states, its optimality. To this end, we use the angular basis, introduced in Subsection 4.2.4. The measurement, $(\hat{\Pi}, \mathbb{1} - \hat{\Pi})$, is fully characterized by $\hat{\Pi}$, which detects any state with zero angular momentum,

$$\hat{\Pi} = \int_0^\infty dr \, r |r, 0\rangle \langle r, 0|. \quad (4.3.14)$$

The radial initial state contains no angular momentum and reads,

$$|\psi\rangle = \int_0^\infty dr \, r \langle r, 0|\psi\rangle |r, 0\rangle. \quad (4.3.15)$$

After the scattering, however, it is a superposition of all possible angular momenta,

$$|\psi_\vartheta\rangle = \hat{S}_\vartheta |\psi\rangle = \sum_{n \in \mathbb{Z}} (-1)^n \int_0^\infty dr \, r J_n(\vartheta |\mathbf{n}_\perp| g(r)) \langle r, 0|\psi\rangle |r, n\rangle. \quad (4.3.16)$$

The probability to measure zero angular momentum after the radial state interacted with the sample reads,

$$\begin{aligned} P(k=0|\vartheta) &= \langle \psi_\vartheta | \hat{\Pi} | \psi_\vartheta \rangle = \int_0^\infty dr \, r |\langle r, 0 | \psi_\vartheta \rangle|^2 \\ &= \int_0^\infty dr \, r \langle \psi_0 | r, 0 \rangle J_0(\vartheta |\mathbf{n}_\perp| g(r)) \langle r, 0 | \psi_0 \rangle \\ &= \langle \psi_0 | \left(\int_0^\infty dr \, r \sum_{k \in \mathbb{Z}} J_0(\vartheta |\mathbf{n}_\perp| g(r))^2 |r, k\rangle \langle r, k| \right) | \psi_0 \rangle \\ &= \langle \psi_0 | J_0(\vartheta |\mathbf{n}_\perp| g(\hat{r}))^2 | \psi_0 \rangle, \end{aligned} \quad (4.3.17)$$

where we rephrased the integral to make the appearance of the $g(\hat{r})^2$ operator explicit. For small ϑ , the Bessel function can be expanded, $J_0(\vartheta |\mathbf{n}_\perp| g(r))^2 = 1 - \vartheta^2 |\mathbf{n}_\perp|^2 g(r)^2 / 2 + \mathcal{O}(\vartheta^4)$, which simplifies the probability,

$$\begin{aligned} P(k=0|\vartheta) &= 1 - \vartheta^2 \frac{|\mathbf{n}_\perp|^2}{2} \langle \psi_0 | g(\hat{r})^2 | \psi_0 \rangle + \mathcal{O}(\vartheta^4) \\ &= 1 - \vartheta^2 \frac{1}{4} \mathcal{J}_{\text{NB}} + \mathcal{O}(\vartheta^4). \end{aligned} \quad (4.3.18)$$

Hence, the opposite probability in this dichotomic setting is $P(k \neq 0|\vartheta) = \vartheta^2 \mathcal{J}_{\text{NB}}^{\text{max}} / 4 +$

$\mathcal{O}(\vartheta^4)$. The corresponding CFI is derived from these probabilities,

$$\mathcal{I} = \frac{(\partial P(k=0|\vartheta))^2}{P(k=0|\vartheta)} + \frac{(\partial P(k \neq 0|\vartheta))^2}{P(k \neq 0|\vartheta)} = \mathcal{J}_{\text{NB}} + \mathcal{O}(\vartheta^2). \quad (4.3.19)$$

Hence, the CFI for the dichotomic measurement of angular momentum reaches the QFI and thus yields optimal precision.

Summarized, the interaction introduces a parameter-dependent occupation of non-zero angular momentum. Thus, we can optimally infer the parameter from the occupation of the state with zero angular momentum. In contrast to position and momentum measurements, this measurement is not directly available in typical electron microscope setups. However, it shows a similarity to works on superresolution techniques: In [7], for example, the distance d of two incoherent light sources is inferred. There, the signal fully occupies the lowest Hermite Gauss mode for $d = 0$ and higher modes get occupied for $d > 0$. Then, a measurement of the lowest mode is sufficient to reach the QFI.

4.4. Sensing with backaction

After discovering the optimal measurements in the case without backaction, we now consider a magnetic moment that is sourced by a quantum spin. Thus, the interaction influences the sample dynamically. The motion of the electron and the spin entangle and, since we can not read out the spin state, the reduced state of the electron becomes mixed. While this significantly increases the complexity of the problem, we are still able to derive analytical results.

In Subsection 4.4.1, we establish an upper bound for the QFI. Second, we formally derive the channel that describes the evolution of the motional state of the electron in Subsection 4.4.2. By means of the bound for the QFI and the CFI for an angular momentum measurement, we are able to derive the QFI in this case in Subsection 4.4.3. Finally, we derive the CFI for momentum measurements.

4.4.1. Pure state QFI

In general, computing the QFI for mixed states is a hard problem and rarely admits analytic solutions, especially for infinite-dimensional systems. In order to arrive at an upper bound and a rough idea of the order of magnitude of the QFI, we assume that we have access to the combined state of the sample spin and the probe electron and compute its pure state QFI.

We start from an initial product state of the electron, labeled by M, and the spin state of the sample, labeled by S, because the systems are spatially separated before the interaction. According to (1.5.16), we can express the pure-state QFI by the variance of the generator $\hat{G} = g(\hat{r}) (\sin(\hat{\varphi})\hat{\sigma}_x - \cos(\hat{\varphi})\hat{\sigma}_y)$,

$$\tilde{\mathcal{J}}_{\text{BA}} = 4\mathbb{V}_{|\psi_{\text{M}}, \psi_{\text{S}}\rangle} [g(\hat{r}) (\sin(\hat{\varphi})\hat{\sigma}_x - \cos(\hat{\varphi})\hat{\sigma}_y)]. \quad (4.4.1)$$

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The expectation value of $\hat{G}$ vanishes due to the radial symmetry of the initial wave packet,

$$\begin{aligned} & \langle \psi_M, \psi_S | g(\hat{r}) [\sin(\hat{\varphi})\hat{\sigma}_x - \cos(\hat{\varphi})\hat{\sigma}_y] | \psi_M, \psi_S \rangle \\ &= \underbrace{\langle \psi_M | g(\hat{r}) \sin(\hat{\varphi}) | \psi_M \rangle}_{=0} \langle \psi_S | \hat{\sigma}_x | \psi_S \rangle - \underbrace{\langle \psi_M | g(\hat{r}) \cos(\hat{\varphi}) | \psi_M \rangle}_{=0} \langle \psi_S | \hat{\sigma}_y | \psi_S \rangle \\ &= 0. \end{aligned} \quad (4.4.2)$$

Thus, the QFI is four times the second moment of $\hat{G}$. This expression simplifies by the fact that the angular part of $\hat{G}$ squares to identity, $(\sin(\hat{\varphi})\hat{\sigma}_x - \cos(\hat{\varphi})\hat{\sigma}_y)^2 = \mathbb{1}$. Therefore, the QFI reduces to

$$\begin{aligned} \tilde{\mathcal{J}}_{\text{BA}} &= 4 \langle \psi_M, \psi_S | g^2(\hat{r}) [\sin(\hat{\varphi})\hat{\sigma}_x - \cos(\hat{\varphi})\hat{\sigma}_y]^2 | \psi_M, \psi_S \rangle \\ &= 4 \langle \psi_M, \psi_S | g^2(\hat{r}) \mathbb{1} | \psi_M, \psi_S \rangle = 4 \langle \psi_M | g^2(\hat{r}) | \psi_M \rangle \langle \psi_S | \psi_S \rangle \\ &= 4 \langle \psi_M | g^2(\hat{r}) | \psi_M \rangle. \end{aligned} \quad (4.4.3)$$

This expression is not only independent from the state of the sample $|\psi_S\rangle$, but also relates to the QFI (4.3.5) without backaction for the case that the magnetic moment is oriented in the plane perpendicular to the beam axis ($|\mathbf{n}_\perp| = 1$), (4.3.5), $\mathcal{J}_{\text{NB}}^{\text{max}} = \langle \psi_M | g(\hat{r})^2 | \psi_M \rangle$, yields,

$$\tilde{\mathcal{J}}_{\text{BA}} = 2\mathcal{J}_{\text{NB}}. \quad (4.4.4)$$

We conclude that there is no more than twice the amount of information contained in the combined sample-probe state as in the case without backaction. The intuition behind this result is the following: Due to the interaction, basically the same phase is imprinted on the motion and the spin state. In principle, we could now probe both systems individually and obtain twice the sensitivity.

In an electron microscope experiment, we do not have access to the sample state, of course. The accessible QFI is that of the reduced post-scattering state,

$$\hat{\rho}_\vartheta = \Lambda_\vartheta[\hat{\rho}_M] = \text{tr}_S \left\{ \hat{S}_\vartheta \hat{\rho}_M \otimes \hat{\rho}_S \hat{S}_\vartheta^\dagger \right\}. \quad (4.4.5)$$

Clearly, this must be less than the QFI of the combined state as tracing over a subsystem is a quantum channel that cannot increase the QFI [178],

$$\mathcal{J} \left[\text{tr}_S \left\{ \hat{S}_\vartheta \hat{\rho}_M \otimes \hat{\rho}_S \hat{S}_\vartheta^\dagger \right\} \right] \leq \mathcal{J} \left[\hat{S}_\vartheta \hat{\rho}_M \otimes \hat{\rho}_S \hat{S}_\vartheta^\dagger \right]. \quad (4.4.6)$$

We can further decompose the mixed states into convex combinations of pure states,

$$\hat{\rho}_M = \sum_k q_k \hat{\rho}_M^{(k)}, \quad \hat{\rho}_S = \sum_n p_n \hat{\rho}_S^{(n)}. \quad (4.4.7)$$

Together with the convexity of the QFI [179], this yields,

$$\begin{aligned} \mathcal{J} \left[\hat{S}_\vartheta \hat{\rho}_M \otimes \hat{\rho}_S \hat{S}_\vartheta^\dagger \right] &\leq \sum_{k,n} q_k p_n \mathcal{J} \left[\hat{S}_\vartheta \hat{\rho}_M^{(k)} \otimes \hat{\rho}_S^{(n)} \hat{S}_\vartheta^\dagger \right] = 4 \sum_{k,n} q_k p_n \text{tr} \left\{ g(\hat{r})^2 \hat{\rho}_M^{(k)} \right\} \\ &= 4 \text{tr} \left\{ g(\hat{r})^2 \hat{\rho}_M \right\}. \end{aligned} \quad (4.4.8)$$

Considering pure motional states, we find the useful bound,

$$\mathcal{J}_{\text{BA}} = \mathcal{J} \left[\Lambda_\vartheta \left[|\psi_M\rangle \langle \psi_M| \right] \right] \leq 4 \langle \psi_M | g(\hat{r})^2 | \psi_M \rangle = 2 \mathcal{J}_{\text{NB}}^{\text{max}}, \quad (4.4.9)$$

that is valid for any $g(r)$, any radial state $|\psi_M\rangle$, and arbitrary spin state $\hat{\rho}_S$.

4.4.2. Scattering channel

In this subsection, we derive the explicit form of the channel that describes the evolution of the motional state of the electron. It is obtained by tracing out the spin degree of freedom after the joint scattering transformation,

$$\Lambda_{\vartheta, \mathbf{c}} [\hat{\rho}_M] = \text{tr}_S \left\{ \hat{S}_\vartheta (\hat{\rho}_M \otimes \hat{\rho}_S) \hat{S}_\vartheta^\dagger \right\}. \quad (4.4.10)$$

The channel $\Lambda_{\vartheta, \mathbf{c}}$ depends on the parameter ϑ and the Bloch vector $\mathbf{c}$, characterizing a general state of the sample,

$$\hat{\rho}_S = \frac{1}{2} (\mathbb{1} + \mathbf{c} \cdot \hat{\boldsymbol{\sigma}}), \quad |\mathbf{c}| \leq 1. \quad (4.4.11)$$

For the derivation, we introduce a simpler notation in terms of three operators,

$$\hat{K}_0 = \cos(\vartheta g(\hat{r})), \quad \hat{K}_+ = \sin(\vartheta g(\hat{r})) e^{+i\hat{\varphi}}, \quad \hat{K}_- = \sin(\vartheta g(\hat{r})) e^{-i\hat{\varphi}}. \quad (4.4.12)$$

With these, the backaction scattering operator (4.2.41) becomes,

$$\begin{aligned} \hat{S}_\vartheta &= \cos(\vartheta g(\hat{r})) + \sin(\vartheta g(\hat{r})) \left[e^{-i\hat{\varphi}} \hat{\sigma}_+ - e^{i\hat{\varphi}} \hat{\sigma}_- \right] \\ &= \hat{K}_0 + \left(\hat{K}_- \hat{\sigma}_+ - \hat{K}_+ \hat{\sigma}_- \right). \end{aligned} \quad (4.4.13)$$

The resulting channel reads,

$$\begin{aligned} \Lambda_{\mathbf{c}, \vartheta} [\hat{\rho}_M] &= \hat{K}_0 \hat{\rho}_M \hat{K}_0^\dagger + \frac{1}{2} \left(\hat{K}_+ \hat{\rho}_M \hat{K}_+^\dagger + \hat{K}_- \hat{\rho}_M \hat{K}_-^\dagger \right) + \frac{c_z}{2} \left(\hat{K}_+ \hat{\rho}_M \hat{K}_+^\dagger - \hat{K}_- \hat{\rho}_M \hat{K}_-^\dagger \right) \\ &\quad + \frac{c_x}{2} \left(\hat{K}_0 \hat{\rho}_M \left[\hat{K}_- - \hat{K}_+ \right]^\dagger + \left[\hat{K}_- - \hat{K}_+ \right] \hat{\rho}_M \hat{K}_0^\dagger \right) \\ &\quad + \frac{ic_y}{2} \left(\left[\hat{K}_+ + \hat{K}_- \right] \hat{\rho}_M \hat{K}_0^\dagger - \hat{K}_0 \hat{\rho}_M \left[\hat{K}_+ + \hat{K}_- \right]^\dagger \right), \end{aligned} \quad (4.4.14)$$

where the lengthy but straightforward calculation is presented in Appendix D.3.1. This simplifies further when we define the complex number $c_x + ic_y = |\mathbf{c}_\perp| e^{i\varphi_c}$,

$$\begin{aligned} \Lambda_{\mathbf{c},\vartheta} [\hat{\rho}_M] &= \hat{K}_0 \hat{\rho}_M \hat{K}_0^\dagger + \frac{1}{2} \left(\hat{K}_+ \hat{\rho}_M \hat{K}_+^\dagger + \hat{K}_- \hat{\rho}_M \hat{K}_-^\dagger \right) + \frac{c_z}{2} \left(\hat{K}_+ \hat{\rho}_M \hat{K}_+^\dagger - \hat{K}_- \hat{\rho}_M \hat{K}_-^\dagger \right) \\ &+ i|\mathbf{c}_\perp| \left(\hat{K}_0 \hat{\rho}_M \sin(\vartheta g(\hat{r})) \sin(\hat{\varphi} - \varphi_c) - \sin(\vartheta g(\hat{r})) \sin(\hat{\varphi} - \varphi_c) \hat{\rho}_M \hat{K}_0 \right). \end{aligned} \quad (4.4.15)$$

Note that we can set $\varphi_c = 0$ without loss of generality by choosing the polar coordinate system accordingly.

With the scattering channel at hand, we can derive the likelihood function for measurements in position and momentum. As in the case without backaction, the position measurement does not carry any information of the parameter ϑ ,

$$P(\mathbf{x}_\perp | \vartheta) = \langle \mathbf{x}_\perp | \Lambda_{\mathbf{c},\vartheta} [\hat{\rho}_M] | \mathbf{x}_\perp \rangle = \langle \mathbf{x}_\perp | \hat{\rho}_M | \mathbf{x}_\perp \rangle = P(\mathbf{x}_\perp | 0), \quad \forall \vartheta, \mathbf{c}. \quad (4.4.16)$$

For the momentum measurement, we additionally assume the incoming motional state of the electron to be pure, $\hat{\rho}_M = |\psi_0\rangle \langle \psi_0|$. The likelihood then reads,

$$\begin{aligned} P(\mathbf{p}_\perp | \vartheta) &= |\langle \mathbf{p}_\perp | \hat{K}_0 | \psi_0 \rangle|^2 + \frac{1+c_z}{2} |\langle \mathbf{p}_\perp | \hat{K}_+ | \psi_0 \rangle|^2 + \frac{1-c_z}{2} |\langle \mathbf{p}_\perp | \hat{K}_- | \psi_0 \rangle|^2 \\ &+ 2|\mathbf{c}_\perp| \text{Im} \left\{ \langle \mathbf{p}_\perp | \sin(\vartheta g(\hat{r})) \sin(\hat{\varphi}) | \psi_0 \rangle \langle \psi_0 | \hat{K}_0 | \mathbf{p}_\perp \rangle \right\}. \end{aligned} \quad (4.4.17)$$

We will use this result in the following subsections to evaluate the CFI for angular momentum and momentum measurements.

4.4.3. Optimality of angular momentum measurements

Analogous to the case without backaction, discussed in Subsection 4.3.3, we show that, for small parameter values ϑ and radial input states $|\psi_0\rangle$, a dichotomic measurement of angular momentum is sufficient to reach a CFI of $\mathcal{I} = 2\mathcal{J}_{\text{NB}}^{\text{max}}$, which was proven to be an upper bound for the QFI in Subsection 4.4.1. We consider the same measurement $(\hat{\Pi}, \mathbb{1} - \hat{\Pi})$ as before, with

$$\hat{\Pi} = \int_0^\infty dr \, r |r, 0\rangle \langle r, 0|. \quad (4.4.18)$$

Upon examination of the scattering channel (4.4.15), only the first term leaves the angular momentum of the input state unchanged. Thus, the probability to remain in a

state of zero angular momentum reads,

$$\begin{aligned}
 P(k=0|\vartheta) &= \int_0^\infty dr \, r \langle r, 0 | \hat{K}_0 | \psi_0 \rangle \langle \psi_0 | \hat{K}_0^\dagger | r, 0 \rangle \\
 &= \int_0^\infty dr \, r \langle \psi_0 | r, 0 \rangle \cos(\vartheta g(r))^2 \langle r, 0 | \psi_0 \rangle \\
 &= \langle \psi_0 | \left(\int_0^\infty dr \, r \sum_{k \in \mathbb{Z}} \cos(\vartheta g(r))^2 |r, k\rangle \langle r, k| \right) | \psi_0 \rangle \\
 &= \langle \psi_0 | \cos(\vartheta g(\hat{r}))^2 | \psi_0 \rangle.
 \end{aligned} \tag{4.4.19}$$

For small ϑ , the cosine can be expanded, which simplifies the probability to

$$P(k=0|\vartheta) = 1 - \vartheta^2 \langle \psi | g(\hat{r})^2 | \psi \rangle + \mathcal{O}(\vartheta^4) = 1 - \frac{\vartheta^2}{2} \mathcal{J}_{\text{NB}}^{\text{max}} + \mathcal{O}(\vartheta^4). \tag{4.4.20}$$

This yields the CFI for a dichotomic measurement,

$$\begin{aligned}
 \mathcal{I} &= (\vartheta^2 \mathcal{J}_{\text{NB}}^{\text{max}2} + \mathcal{O}(\vartheta^4)) \left(\frac{1}{\frac{\vartheta^2}{2} \mathcal{J}_{\text{NB}}^{\text{max}} + \mathcal{O}(\vartheta^4)} + \frac{1}{1 - \frac{\vartheta^2}{2} \mathcal{J}_{\text{NB}}^{\text{max}} + \mathcal{O}(\vartheta^4)} \right) \\
 &= 2\mathcal{J}_{\text{NB}}^{\text{max}} + \mathcal{O}(\vartheta^2).
 \end{aligned} \tag{4.4.21}$$

The bound, $\mathcal{I} \leq \mathcal{J}_{\text{BA}}$, is true by definition of the QFI. In Subsection 4.4.1, on the other hand, we found the upper bound, $\mathcal{J}_{\text{BA}} \leq 2\mathcal{J}_{\text{NB}}^{\text{max}}$. Together with (4.4.21) these inequalities imply the QFI in the backaction case to be,

$$\mathcal{J}_{\text{BA}} = 2\mathcal{J}_{\text{NB}}^{\text{max}} + \mathcal{O}(\vartheta^2). \tag{4.4.22}$$

We see that, for small parameters, the QFI in the backaction case is twice as high as the QFI in the case without backaction. This behavior can be motivated from the angular momentum preserving nature of the interaction: The evolution of the spin is already determined from the evolution of the electron. Hence, we can expect to obtain the information, in form of CFI, from measuring the motional state of the electron and the state of the spin.

4.4.4. CFI in diffraction mode

In Subsection 4.3.2, we learned that measurements in diffraction mode are optimal in the case without backaction if the initial state $|\psi_0\rangle$ is radial and real-valued (in position space). Under the same conditions and for small parameters, we show that the momentum measurement CFI in the case with backaction still reaches the QFI from the case without backaction. However, the actual QFI in this scenario is twice as high and the momentum measurement is thus not optimal.

For the calculation, we can use the likelihood function (4.4.17). We shall now derive explicit expressions for the matrix elements that appear there. First, we calculate the

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terms imparting angular momentum kicks by $\pm\hbar$, using the transition amplitude between momentum eigenstates and the angular eigenbasis (4.2.48),

$$\begin{aligned}
\langle \mathbf{p}(p, \varphi_p) | \hat{K}_{\pm} | \psi_0 \rangle &= \int_0^\infty dr \, r \sum_{k \in \mathbb{Z}} \langle \mathbf{p}(p, \varphi_p) | \hat{K}_{\pm} | r, k \rangle \langle r, k | \psi_0 \rangle \\
&= \int_0^\infty dr \, r \langle \mathbf{p}(p, \varphi_p) | \hat{K}_{\pm} | r, 0 \rangle \langle r, 0 | \psi_0 \rangle \\
&= \int_0^\infty dr \, r \sin(\vartheta g(r)) \langle \mathbf{p}(p, \varphi_p) | r, \pm 1 \rangle \langle r, 0 | \psi_0 \rangle \\
&= -\frac{i}{\hbar} \frac{e^{\pm i\varphi_p}}{\sqrt{2\pi}} \int_0^\infty \frac{dr \, r}{\hbar} \sin(\vartheta g(r)) J_1\left(\frac{pr}{\hbar}\right) \langle r, 0 | \psi_0 \rangle.
\end{aligned} \tag{4.4.23}$$

An analogous calculation yields the matrix element of the term imparting not kick,

$$\begin{aligned}
\langle \mathbf{p}(p, \varphi_p) | \hat{K}_0 | \psi_0 \rangle &= \int_0^\infty dr \, r \cos(\vartheta g(r)) \langle \mathbf{p}_\perp | r, 0 \rangle \langle r, 0 | \psi_0 \rangle \\
&= \frac{1}{\sqrt{2\pi}} \int_0^\infty \frac{dr \, r}{\hbar} \cos(\vartheta g(r)) J_0\left(\frac{pr}{\hbar}\right) \langle r, 0 | \psi_0 \rangle.
\end{aligned} \tag{4.4.24}$$

The last term we need for the momentum likelihood function is a linear combination of the two kick terms,

$$\begin{aligned}
\langle \mathbf{p}(p, \varphi_p) | \sin(\hat{\varphi}) \sin(\vartheta g(\hat{r})) | \psi_0 \rangle &= \frac{1}{2i} \langle \mathbf{p}(p, \varphi_p) | (\hat{K}_+ - \hat{K}_-) | \psi_0 \rangle \\
&= -i \sin(\varphi_p) \frac{1}{\sqrt{2\pi}} \int_0^\infty \frac{dr \, r}{\hbar} \sin(\vartheta g(r)) J_1\left(\frac{pr}{\hbar}\right) \langle r, 0 | \psi_0 \rangle.
\end{aligned} \tag{4.4.25}$$

Noting that $\left| \langle \mathbf{p}(p, \varphi_p) | \hat{K}_+ | \psi_0 \rangle \right|^2 = \left| \langle \mathbf{p}(p, \varphi_p) | \hat{K}_- | \psi_0 \rangle \right|^2$, we can now express the likelihood function (4.4.17) as,

$$P(p, \varphi_p) = |a(p|\vartheta)|^2 + |b(p|\vartheta)|^2 - 2|\mathbf{c}_\perp| \sin(\varphi_p) a(p|\vartheta) b(p|\vartheta). \tag{4.4.26}$$

The functions a and b , which depend only on the radial momentum, are given by,

$$a(p|\vartheta) = \int_0^\infty \frac{dr \, r}{\sqrt{2\pi\hbar}} \cos(\vartheta g(r)) J_0\left(\frac{pr}{\hbar}\right) \langle r, 0 | \psi_0 \rangle, \tag{4.4.27}$$

$$b(p|\vartheta) = \int_0^\infty \frac{dr \, r}{\sqrt{2\pi\hbar}} \sin(\vartheta g(r)) J_1\left(\frac{pr}{\hbar}\right) \langle r, 0 | \psi_0 \rangle. \tag{4.4.28}$$

We differentiate the likelihood function with respect to the parameter (omitting the arguments),

$$\partial P = 2(a\partial a + b\partial b) - 2|\mathbf{c}_\perp| \sin(\varphi_p) (a\partial b + b\partial a). \tag{4.4.29}$$

The derivatives on the right hand side can be calculated by differentiation under the

integral,

$$\partial a(p|\vartheta) = - \int_0^\infty \frac{dr}{\sqrt{2\pi\hbar}} r g(r) \sin(\vartheta g(r)) J_0\left(\frac{pr}{\hbar}\right) \langle r, 0|\psi_0\rangle \quad (4.4.30)$$

$$\partial b(p|\vartheta) = + \int_0^\infty \frac{dr}{\sqrt{2\pi\hbar}} r g(r) \cos(\vartheta g(r)) J_1\left(\frac{pr}{\hbar}\right) \langle r, 0|\psi_0\rangle. \quad (4.4.31)$$

These expressions simplify significantly for $\vartheta = 0$, so that

$$\begin{aligned} P(p, \varphi_p|0) &= |a|^2, \\ \partial P(p, \varphi_p|0) &= -2|\mathbf{c}_\perp| \sin(\varphi_p) a \partial b. \end{aligned} \quad (4.4.32)$$

Therefore, the CFI can be written in this case as,

$$\mathcal{I}(0) = \int d^2\mathbf{p}_\perp \frac{4|\mathbf{c}_\perp|^2 \sin(\varphi_p)^2 |a|^2 |\partial b|^2}{|a|^2} = 4\pi |\mathbf{c}_\perp|^2 \int_0^\infty dp p |\partial b(p|0)|^2. \quad (4.4.33)$$

The remaining integral can be simplified by making use of an integral representation of the delta function in terms of Bessel functions,

$$\begin{aligned} & \int_0^\infty dp p |\partial b(p|0)|^2 \\ &= \int_0^\infty dp p \int_0^\infty \int_0^\infty \frac{dr_1 dr_2 r_1 r_2}{2\pi\hbar^2} g(r_1) g(r_2) J_1\left(\frac{pr_1}{\hbar}\right) J_1\left(\frac{pr_2}{\hbar}\right) \langle \psi_0|r_1, 0\rangle \langle r_2, 0|\psi_0\rangle \\ &= \int_0^\infty \int_0^\infty \frac{dr_1 dr_2 r_1 r_2}{2\pi\hbar^2} g(r_1) g(r_2) \left(\int_0^\infty dp p J_1\left(\frac{pr_1}{\hbar}\right) J_1\left(\frac{pr_2}{\hbar}\right) \right) \langle \psi_0|r_1, 0\rangle \langle r_2, 0|\psi_0\rangle \\ &= \int_0^\infty \int_0^\infty \frac{dr_1 dr_2 r_1 r_2}{2\pi\hbar^2} g(r_1) g(r_2) \left(\frac{\hbar^2}{r_1} \delta^{(1)}(r_1 - r_2) \right) \langle \psi_0|r_1, 0\rangle \langle r_2, 0|\psi_0\rangle \\ &= \int_0^\infty \frac{dr}{2\pi} r \langle \psi_0|r, 0\rangle g(r)^2 \langle r, 0|\psi_0\rangle = \frac{1}{2\pi} \langle g(\hat{r})^2 \rangle_0, \end{aligned} \quad (4.4.34)$$

Altogether, the CFI for momentum measurements becomes,

$$\mathcal{I}(0) = |\mathbf{c}_\perp|^2 \mathcal{J}_{\text{NB}}^{\text{max}}. \quad (4.4.35)$$

Notice the similarity to the case without backaction, where the QFI (Subsection 4.3.1) and the CFI for momentum measurements (Subsection 4.3.2) read $\mathcal{J}_{\text{NB}} = |\mathbf{n}_\perp|^2 \mathcal{J}_{\text{NB}}^{\text{max}}$. Accordingly, the coherence of the spin state, $|\mathbf{c}_\perp|$, now plays the role of the component of the classical magnetic moment lying in the plane perpendicular to the beam axis, $\mathbf{n}_\perp$. Put differently, the precision of momentum measurements with backaction depends strongly on the state of the sample spin and ranges from zero to the optimal value without backaction, $\mathcal{J}_{\text{NB}}^{\text{max}}$. Experimentally, a bias field perpendicular to the beam axis aligns the spin accordingly and enables high-precision sensing. However, even for the maximal value of $|\mathbf{c}_\perp| = 1$, this does not reach the QFI for the case with backaction, $\mathcal{J}_{\text{BA}} = 2\mathcal{J}_{\text{NB}}^{\text{max}}$. Hence for a properly aligned spin, the precision of momentum measurements is still high.

However, they are far from optimal.

4.5. State discrimination without backaction

After the discussion of quantum sensing, we now turn to another metrological task, called *state discrimination*. Here, the strength of the magnetic moment is known, but we do not know whether a sample was placed in the machine or not. We try to maximize the success probability to infer from measurement data whether the sample was there or not. The sample is assumed to be static.

This section is similarly structured as the sensing section without backaction: First, we calculate the optimal quantum bound in form of the quantum trace distance in Subsection 4.5.1. Then, we calculate the classical trace distance for the probability distributions obtained from quantum measurements. We evaluate the trace distance in a continuum of *defocused* bases between position and momentum in Subsection 4.5.2. Finally, we consider the angular momentum measurement in Subsection 4.5.3.

4.5.1. Quantum trace distance

The quantum trace distance between the unscattered electron state $|\psi_0\rangle$ and the scattered state, $|\psi_\vartheta\rangle = \hat{S}_\vartheta |\psi_0\rangle$, is best calculated with the Jacobi-Anger form of the scattering operator (4.2.29), which we repeat here for convenience,

$$\hat{S}_\vartheta = \sum_{n \in \mathbb{Z}} (-1)^n J_n(\vartheta |\mathbf{n}_\perp| g(\hat{r})) e^{in\hat{\varphi}}. \quad (4.5.1)$$

Assuming a radial initial state $|\psi_0\rangle$, the scalar product reduces to the expectation value,

$$\langle \psi_0 | \psi_\vartheta \rangle = \langle \psi_0 | J_0(\vartheta |\mathbf{n}_\perp| g(\hat{r})) | \psi_0 \rangle = \langle J_0(\vartheta |\mathbf{n}_\perp| g(\hat{r})) \rangle_0. \quad (4.5.2)$$

With this, the quantum trace distance can be given as (1.5.37),

$$D_Q(|\psi_0\rangle, |\psi_\vartheta\rangle) = \sqrt{1 - \langle J_0(\vartheta |\mathbf{n}_\perp| g(\hat{r})) \rangle_0^2}. \quad (4.5.3)$$

For small parameter values ϑ , we expand the Bessel function and find a connection to the QFI 4.3.5,

$$\begin{aligned} D_Q(|\psi_0\rangle, |\psi_\vartheta\rangle) &= \sqrt{1 - \langle \psi_0 | \left(1 - \vartheta^2 \frac{|\mathbf{n}_\perp|^2}{2} g(\hat{r})^2 + \mathcal{O}(\vartheta^4) \right) | \psi_0 \rangle} \\ &= \frac{\vartheta}{\sqrt{2}} \sqrt{|\mathbf{n}_\perp| \langle g(\hat{r})^2 \rangle_0} \sqrt{1 + \mathcal{O}(\vartheta^2)} \\ &= \frac{\vartheta}{2} \sqrt{\mathcal{J}_{\text{NB}}} + \mathcal{O}(\vartheta^3). \end{aligned} \quad (4.5.4)$$

The optimal measurement, saturating the bound, is known to be the projector onto

the positive eigenspace $|+\rangle\langle+|$ of the rank-two operator $|\psi_\vartheta\rangle\langle\psi_\vartheta| - |\psi_0\rangle\langle\psi_0|$ [70]. We find this projector by establishing an orthonormal basis on the span of $|\psi_0\rangle$ and $|\psi_\vartheta\rangle$,

$$\begin{aligned} |0\rangle &= |\psi_0\rangle \\ |\vartheta\rangle &= \frac{|\psi_\vartheta\rangle - \langle\psi_0|\psi_\vartheta\rangle |\psi_0\rangle}{\sqrt{1 - |\langle\psi_0|\psi_\vartheta\rangle|^2}}. \end{aligned} \quad (4.5.5)$$

Note that for radial states, the scalar product is real-valued,

$$\langle\psi_0|\psi_\vartheta\rangle = \langle\psi_0|\hat{S}_\vartheta|\psi_0\rangle = \langle J_n(\vartheta|\mathbf{n}_\perp|g(\hat{r}))\rangle_0 \in \mathbb{R}. \quad (4.5.6)$$

Thus, we can parametrize the basis by the trace distance $D_Q(|\psi_0\rangle, |\psi_\vartheta\rangle)$, for which we will now omit the arguments, and get,

$$|\vartheta\rangle = \frac{1}{D_Q} \left(|\psi_\vartheta\rangle - \sqrt{1 - D_Q^2} |\psi_0\rangle \right). \quad (4.5.7)$$

In the new basis, the difference of the states reads,

$$|\psi_\vartheta\rangle\langle\psi_\vartheta| - |\psi_0\rangle\langle\psi_0| = D_Q \begin{pmatrix} D_Q & \sqrt{1 - D_Q^2} \\ \sqrt{1 - D_Q^2} & -D_Q \end{pmatrix}. \quad (4.5.8)$$

This matrix can be diagonalized straightforwardly, leading to the sought positive eigenvector,

$$\begin{aligned} |+\rangle &= \sqrt{\frac{1 + D_Q}{2}} |\vartheta\rangle + \sqrt{\frac{1 - D_Q}{2}} |0\rangle \\ &= \frac{1}{D_Q} \left(\sqrt{\frac{1 + D_Q}{2}} |\psi_\vartheta\rangle - \sqrt{\frac{1 - D_Q}{2}} |\psi_0\rangle \right), \end{aligned} \quad (4.5.9)$$

to the eigenvalue D_Q .

Hence, the optimal measurement to distinguish $|\psi_\vartheta\rangle$ from $|\psi_0\rangle$ is given by $(|+\rangle\langle+|, \mathbb{1} - |+\rangle\langle+|)$. However, the practical implementation of such a measurement in a laboratory setting remains unclear. There, the exact states $|\psi_0\rangle$ and $|\psi_\vartheta\rangle$ might not even be known. In order to find a practical measurement scheme, we will consider more standardized measurement settings in the following sections.

4.5.2. Discrimination in defocus planes

In the preceding subsection, we already presented how to solve the state discrimination problem optimally with a dichotomic measurement. Since the implementation of such a measurement is non-trivial or even impossible, we now investigate how well this task can be performed by experimentally realizable measurements.

We start with a momentum measurement: Applying (1.5.32) to the momentum space

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probability distribution, $P(\mathbf{p}_\perp|\vartheta) = |\langle \mathbf{p}_\perp | \psi_\vartheta \rangle|^2$, we have to solve a non-trivial integral for classical trace distance,

$$D_C(P(\mathbf{p}_\perp|\vartheta), P(\mathbf{p}_\perp|0)) = \frac{1}{2} \int d^2\mathbf{p}_\perp |P(\mathbf{p}_\perp|\vartheta) - P(\mathbf{p}_\perp|0)|, \quad (4.5.10)$$

which we evaluate numerically. Since the classical trace distance for momentum measurements is significantly worse than the quantum trace distance, we extend the range of measurements.

In order to restrict ourselves to experimentally realizable measurements, we construct the measurement by first applying a unitary corresponding to realizable electron optics and then measuring in the position basis, compare Fig. 4.1. To this end, we consider an electron lens, which imprints a position-dependent phase on the electron wavefunction,

$$\hat{U}_{\text{lense}} = \exp\left(-i\frac{k_0}{2f}\hat{\mathbf{x}}_\perp^2\right), \quad (4.5.11)$$

where f is the focal length and $k_0 = p_0/\hbar$ is the wavenumber corresponding to the forward momentum p_0 , see Subsection 4.2.2. The second unitary is given by the free propagation of the electron,

$$\hat{U}_{\text{free}} = \exp\left(-i\frac{\mathbf{p}_\perp^2}{2m\hbar}t\right) = \exp\left(-i\frac{z}{2k_0}\frac{\mathbf{p}_\perp^2}{\hbar^2}\right), \quad (4.5.12)$$

in which we substituted the time span t by the distance the electron traveled in this time span, z . Combining the two unitaries,

$$\hat{U}_{\text{optics}} = \hat{U}_{\text{free}}\hat{U}_{\text{lense}}\hat{U}_{\text{free}} \quad (4.5.13)$$

enables us to scan through a continuum of bases by, e.g, fixing f and varying z .

Before we go into the general case, we shortly discuss the two extreme cases: A trivial example is the case, where the electron does not propagate at all, $z = 0$. Then, we effectively measure the original wavefunction in position space. Note that the lens picture breaks down for $z = 0$ since it requires the sample, the lens, and the screen to be at the same position. For completeness, however, we include this case in the following analysis, too. The other extreme case is obtained from the so-called $2f$ -setup, where the free propagation length z before and after the lens is chosen to be the focal length, $z = f$. Applying this transformation before measuring in the position basis, effectively measures the original wavefunction in the momentum basis as can be seen from inserting two resolutions of identity,

$$\langle \mathbf{x}_\perp | \hat{U}_{2f} | \psi \rangle = \frac{\hbar k_0}{f} \left\langle \mathbf{p}_\perp = \frac{\hbar k_0}{f} \mathbf{x}_\perp \middle| \psi \right\rangle, \quad (4.5.14)$$

where we ignore an unimportant phase and the notation on the right hand side denotes

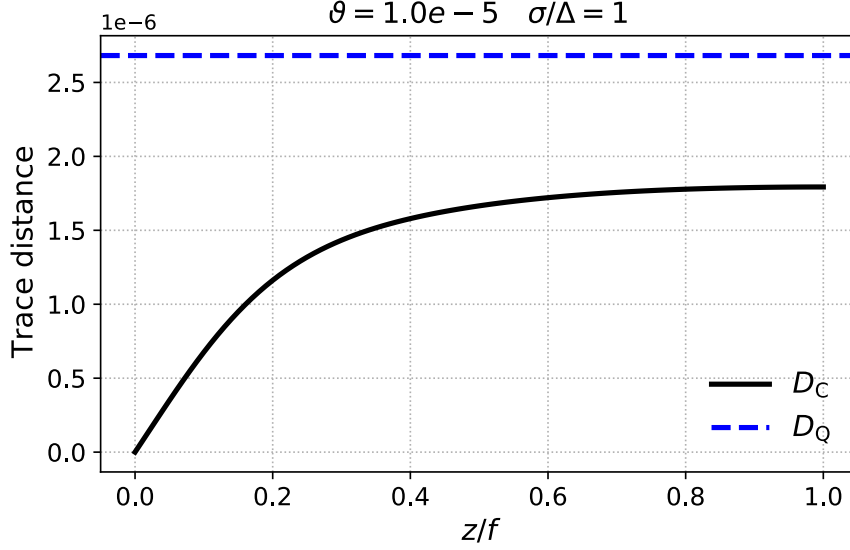


Figure 4.4.: Plot of the classical trace distance (black) versus the free propagation distance in units of the focal length, z/f , in the setting described by (4.5.13). Each value characterizes a defocused measurement basis between position and momentum space. For comparison, the basis-independent quantum trace distance is depicted in blue. Here the lens is characterized by $k_0\Delta^2/f = 0.5$ and the parameter admits the realistic value of $\vartheta = 10^{-5}$. Here the state is a Gaussian wavepacket that is of the same size as the regulator $\sigma = \Delta$. The classical trace distance is zero for $z = 0$, i.e. in position space, and increases monotonously reaching the highest value for momentum measurements $z = f$. The optimal bound of the quantum trace distance is reached for no z

the momentum space wavefunction evaluated at a momentum of $\hbar k_0/f \mathbf{x}_\perp^2$.

For fixed focal length f , we will now consider the continuum of measurements for $z \in [0, f]$. We consider all lengths in units of the regulator Δ and find the dimensionless quantity $\frac{k_0\Delta^2}{f}$ that characterizes the lens transformation. For numerical stability, we choose this parameter to be $\frac{k_0\Delta^2}{f} = \frac{1}{2}$. The classical trace distance is plotted versus the continuum of measurement bases, labeled by z/f in Fig. 4.4 and Fig. 4.5. In Fig. 4.4, we assume the standard deviation of the initial wavefunction to be of the size of the regulator. We see that, as expected, the classical trace distance is zero in position space, i.e. $z = 0$. For increasing z , the classical trace distance increases monotonously and reaches the largest value for momentum measurements, $z = f$. The basis-independent quantum trace distance is in any case larger than the classical trace distance. The behavior changes if we increase the size of the wavefunction to four times the size of the regulator, as shown in Fig. 4.5. In this case, the classical trace distance increases

²Note that the $2f$ transformation can be rewritten as the propagator of a harmonic oscillator. An evolution that corresponds to an angle of $\pi/2$ thus rotates the position wave function on the momentum wavefunction.

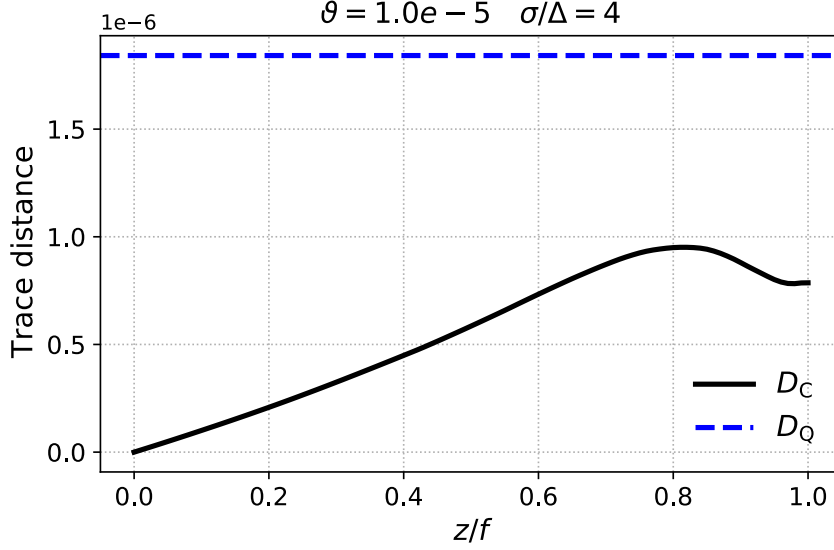


Figure 4.5.: Plot of the classical trace distance (black) versus the free propagation distance in units of the focal length, z/f . Compare Fig. 4.4. Here the state is a Gaussian wavepacket that is four times larger than the regulator $\sigma = 4\Delta$. The classical trace distance is zero in position space and increases monotonously until it reaches its maximum. In contrast to Fig. 4.4, this maximum is already reached at $z \approx 0.8f$. The optimal bound of the quantum trace distance is reached for no z .

monotonously until the maximal value is reached at the measurement basis labeled by $z \approx 0.8f$. This measurement also exceeds the momentum measurement. In the limit of small parameters $\vartheta \ll 1$ and large wavefunction $\sigma \gg \Delta$ we can motivate this behavior: The transformation of the state is well described by the first order contribution,

$$\hat{S}_\vartheta |\psi_0\rangle = |\psi_0\rangle + \vartheta |\partial\psi_0\rangle + \mathcal{O}(\vartheta^2). \quad (4.5.15)$$

Upon measuring in an arbitrary basis ($|\xi\rangle$), the difference of likelihoods is governed by the overlap of $\langle\xi|\psi_0\rangle$ and $\langle\xi|\partial\psi_0\rangle$,

$$\left| \langle\xi|\hat{S}_\vartheta|\psi_0\rangle \right|^2 - |\langle\xi|\psi_0\rangle|^2 = 2\vartheta \text{Re} \{ \langle\psi_0|\xi\rangle \langle\xi|\partial\psi_0\rangle \} + \mathcal{O}(\vartheta^2), \quad (4.5.16)$$

which can only be maximal when both terms, $\langle\xi|\psi_0\rangle$ and $\langle\xi|\partial\psi_0\rangle$, are roughly of the same size. In position space, we know that $\langle\xi|\psi_0\rangle$ has a standard deviation of σ and the derivative, $\langle\xi|\partial\psi_0\rangle$, is modulated by the interaction with length scale Δ . Hence, we could already conclude from $\sigma \gg \Delta$ that the trace distance is small in position space. In momentum space, on the other hand, the standard deviations behave like $\sigma_p \sim 1/\sigma$ and $\Delta_p \sim 1/\Delta$. We have the opposite situation as in position space and the overlap is small again. Suppose the parameter z is slowly varied from 0 to f . During this process, the wavepacket continuously shrinks, while the interaction length scale increases. At an

intermediate point, both acquire the same size, resulting in maximal overlap.

Summarizing, we presented a continuum of bases based on a lens transformation. We showed that the quantum trace distance is never reached. For large wavepackets $\sigma \gg \Delta$, a measurement in a defocused plane is advantageous over the momentum measurement.

4.5.3. Discrimination by angular momentum measurements

In this subsection, we show how well we can tell apart the scattered and unscattered state from the statistics obtained from the measurement $\hat{\Pi}$, $\mathbb{1} - \hat{\Pi}$ that discriminates states with zero angular momentum from states with non-zero angular momentum. In principle, this measurement has the same disadvantage as the optimal measurement discussed in Subsection 4.5.1, since an experimental implementation is not known. However, the optimal measurement has the additional drawback that the measurement depends on the post-scattering state, which is here not the case. Additionally, the angular momentum measurement has already proven optimal in the task of sensing.

We assume a radial initial state $|\psi_0\rangle$ and re-use the probability from Subsection 4.3.3,

$$P(k=0|\vartheta) = \langle J_0(\vartheta|\mathbf{n}_\perp|g(\hat{r}))^2 \rangle_0. \quad (4.5.17)$$

Together with the opposite probability we obtain the full outcome statistics,

$$P_\vartheta = (P(k=0|\vartheta), 1 - P(k=0|\vartheta)) \quad (4.5.18)$$

that need to be distinguished from $P_0 = (1, 0)$.

A straightforward calculation yields the classical trace distance,

$$\begin{aligned} D_C(P_0, P_\vartheta) &= \frac{1}{2} |1 - P(k=0|\vartheta)| + \frac{1}{2} |0 - P(k \neq 0|\vartheta)| \\ &= 1 - \langle J_0(\vartheta|\mathbf{n}_\perp|g(\hat{r}))^2 \rangle_0. \end{aligned} \quad (4.5.19)$$

We see that the classical and the quantum trace distance are related by the variance of a multiplication operator,

$$\begin{aligned} D_Q(|\psi_0\rangle, |\psi_\vartheta\rangle)^2 - D_C(P_0, P_\vartheta) &= \langle J_0(\vartheta|\mathbf{n}_\perp|g(\hat{r}))^2 \rangle_0 - \langle J_0(\vartheta g(\hat{r})) \rangle_0^2 \\ &= \mathbb{V}[J_0(\vartheta|\mathbf{n}_\perp|g(\hat{r}))]_0. \end{aligned} \quad (4.5.20)$$

We can upper bound the square by noting that the trace distance is between zero and one,

$$D_Q(|\psi_0\rangle, |\psi_\vartheta\rangle) \geq D_C(P_0, P_\vartheta) + \mathbb{V}[J_0(\vartheta|\mathbf{n}_\perp|g(\hat{r}))]_0. \quad (4.5.21)$$

This shows that the optimal value can not be achieved with angular momentum measurements, unless the initial state is chosen to be an eigenstate of $J_0(\vartheta|\mathbf{n}_\perp|g(\hat{r}))$. Although this bound is not necessarily strict, it reveals a way to potentially optimize the initial state by minimizing the variance of $J_0(\vartheta|\mathbf{n}_\perp|g(\hat{r}))$.

A relation between the QFI and the trace distance becomes evident if we assume small

parameters $\vartheta \ll 1$,

$$\begin{aligned} D_C(P_0, P_\vartheta) &= 1 - \langle \psi_0 | \left(1 - \frac{\vartheta^2}{2} g(\hat{r})^2 + \mathcal{O}(\vartheta^4) \right) | \psi_0 \rangle \\ &= \frac{\vartheta^2}{2} |\mathbf{n}_\perp|^2 \langle g(\hat{r})^2 \rangle_0 + \mathcal{O}(\vartheta^4) = \frac{\vartheta^2}{4} |\mathbf{n}_\perp|^2 \mathcal{J}_{\text{NB}}^{\text{max}}. \end{aligned} \quad (4.5.22)$$

Concluding, we have calculate the classical trace distance for the dichotomic angular momentum measurement. We found a bound that shows the inferiority of this classical trace distance compared with the quantum trace distance. Finally, we expanded the expression and once more found the quantum Fisher information for the case without backaction.

4.6. State discrimination with backaction

In this section, we focus on the state discrimination task for the case with backaction. As before, the backaction makes the reduced electron state of motion mixed, which increases the complexity compared to the case without backaction.

First, we derive an upper bound for the quantum trace distance in Subsection 4.6.1. Second, we consider the angular momentum measurement in Subsection 4.6.2.

4.6.1. An upper bound for the trace distance

The (quantum) trace distance between the unscattered $\hat{\rho}_0$ and a scattered state $\hat{\rho}_{\text{out}} = \Lambda_{\vartheta, \mathbf{c}}[\hat{\rho}_0]$ is analytically hard to compute. Here we derive a bound for the trace distance, based on the natural representation of the channel,

$$\Lambda_{\vartheta, \mathbf{c}}[\hat{\rho}_M] = \text{tr}_S \left\{ \hat{S}_\vartheta (\hat{\rho}_M \otimes \hat{\rho}_S) \hat{S}_\vartheta^\dagger \right\}. \quad (4.6.1)$$

We will show that, if the initial electron wavefunction $|\psi_0\rangle$ is radial, the trace distance between unscattered and scattered state is bounded from above,

$$D_Q(|\psi_0\rangle \langle \psi_0|, \Lambda_{\vartheta, \mathbf{c}}[|\psi_0\rangle \langle \psi_0|]) \leq \frac{1 - |\mathbf{c}|^2}{2} + \frac{1 + |\mathbf{c}|^2}{2} \sqrt{1 - \langle \cos(\vartheta g(\hat{r})) \rangle_0^2}. \quad (4.6.2)$$

In order to prove this, we start with the derivation in the pure case, where we distinguish $|\psi_M, \psi_S\rangle$ and $\hat{S}_\vartheta |\psi_M, \psi_S\rangle$. The trace distance in this case only depends on the scalar product of the two states, which reads,

$$\langle \psi_M, \psi_S | \hat{S}_\vartheta | \psi_M, \psi_S \rangle = \langle \hat{K}_0 \rangle_M + \langle \hat{K}_- \rangle_M \langle \hat{\sigma}_+ \rangle_S - \langle \hat{K}_+ \rangle_M \langle \hat{\sigma}_- \rangle_S. \quad (4.6.3)$$

For radial states $|\psi_M\rangle$, the last two terms vanish due to $\langle \hat{K}_\pm \rangle_M = 0$ and the trace distance

becomes

$$D_Q \left(|\psi_M, \psi_S\rangle, \hat{S}_\vartheta |\psi_M, \psi_S\rangle \right) = \sqrt{1 - \langle \hat{K}_0 \rangle_M^2} = \sqrt{1 - \langle \cos(\vartheta g(\hat{r})) \rangle_M^2}. \quad (4.6.4)$$

Next, we consider the sample to be in a mixed state $\hat{\rho}_S$, which can be decomposed into a mixture of orthogonal pure states,

$$\hat{\rho}_S = \sum_{j=0}^1 p_j |\psi_S^{(j)}\rangle \langle \psi_S^{(j)}|. \quad (4.6.5)$$

Two orthogonal states are perfectly distinguishable, resulting in unit trace distance. Using this fact and the convexity of the trace distance, we obtain an upper bound for the mixed sample state,

$$\begin{aligned} & D_Q \left(|\psi_M\rangle \langle \psi_M| \otimes \hat{\rho}_S, \hat{S}_\vartheta |\psi_M\rangle \langle \psi_M| \otimes \hat{\rho}_S \hat{S}_\vartheta^\dagger \right) \\ & \leq \sum_{j,k=0}^1 p_j p_k D_Q \left(|\psi_M\rangle \langle \psi_M| \otimes |\psi_S^{(j)}\rangle \langle \psi_S^{(j)}|, \hat{S}_\vartheta |\psi_M\rangle \langle \psi_M| \otimes |\psi_S^{(k)}\rangle \langle \psi_S^{(k)}| \hat{S}_\vartheta^\dagger \right) \\ & = 2p_0 p_1 + (p_0^2 + p_1^2) \sqrt{1 - \langle \cos(\vartheta g(\hat{r})) \rangle_M^2} \\ & = 2 \det(\hat{\rho}_S) + \text{tr} \{ \hat{\rho}_S^2 \} \sqrt{1 - \langle \cos(\vartheta g(\hat{r})) \rangle_M^2}, \end{aligned} \quad (4.6.6)$$

where the $j = k$ terms yield the result for pure states (4.6.4). Finally, we identify the two probability expressions as the determinant and the purity of the initial spin state. In terms of the Bloch vector $\mathbf{c}$, we have [70],

$$\det(\hat{\rho}_S) = \frac{1 - |\mathbf{c}|^2}{4}, \quad \text{tr} \{ \hat{\rho}_S^2 \} = \frac{1 + |\mathbf{c}|^2}{2}. \quad (4.6.7)$$

Finally, the fact that taking the partial trace can only decrease the quantum trace distance yields the desired bound (4.6.2).

4.6.2. Trace distance for angular momentum measurements

In this subsection, we calculate the classical trace distance between the probability distributions that we obtain from an angular momentum measurement of the unscattered state $|\psi_0\rangle \langle \psi_0|$ and scattered $\Lambda_{\vartheta, \mathbf{c}}[|\psi_0\rangle \langle \psi_0|]$ state. We use the dichotomic measurement $(\hat{\Pi}, \mathbf{1} - \hat{\Pi})$, introduced in Subsection 4.3.3 and the probability to measure no angular momentum, known from Subsection 4.4.3,

$$P(k = 0 | \vartheta) = \langle \cos(\vartheta g(\hat{r}))^2 \rangle_0. \quad (4.6.8)$$

Consistently, the opposite probability can be rephrased to be,

$$\begin{aligned} P(k \neq 0|\vartheta) &= 1 - P(k = 0|\vartheta) = \langle \psi_0 | \mathbb{1} | \psi_0 \rangle - \langle \psi_0 | \cos(\vartheta g(\hat{r}))^2 | \psi_0 \rangle \\ &= \langle \sin(\vartheta g(\hat{r}))^2 \rangle_0. \end{aligned} \quad (4.6.9)$$

We use the tuple notation for the probability distribution

$$P_\vartheta = (P(k = 0|\vartheta), P(k \neq 0|\vartheta)), \quad (4.6.10)$$

in which the distribution of the unscattered state reads $P_0 = (1, 0)$. The trace distance between the two distributions is calculated to be,

$$\begin{aligned} D_C(P_0, P_\vartheta) &= \frac{1}{2} |1 - P(k = 0|\vartheta)| + \frac{1}{2} |0 - P(k \neq 0|\vartheta)| \\ &= \langle \psi_0 | \sin(\vartheta g(\hat{r}))^2 | \psi_0 \rangle. \end{aligned} \quad (4.6.11)$$

We see that, at least for pure spin states ($|\mathbf{c}| = 1$), this relates to the bound from the last subsection (4.6.2) in the same fashion as the angular momentum measurement trace distance to the quantum trace distance in the non backaction case (4.5.21),

$$\sqrt{1 - \langle \cos(\vartheta g(\hat{r})) \rangle_0^2} \geq D_C(P_0, P_\vartheta) + \mathbb{V}[\cos(\vartheta g(\hat{r}))]_0. \quad (4.6.12)$$

For small parameters ϑ , we can expand $\sin(\vartheta g(\hat{r}))^2 = \vartheta^2 g(\hat{r})^2 + \mathcal{O}(\vartheta^4)$ and find once more the expression of the maximal QFI,

$$\begin{aligned} D_C(P_0, P_\vartheta) &= \vartheta^2 \langle \psi_0 | g(\hat{r})^2 | \psi_0 \rangle + \mathcal{O}(\vartheta^4) \\ &= \frac{\vartheta^2}{2} \mathcal{J}_{\text{NB}}^{\text{max}} + \mathcal{O}(\vartheta^4). \end{aligned} \quad (4.6.13)$$

4.7. Discussion and outlook

In this project, we investigated the interaction of the electron beam in an electron microscope with a sample in form of a magnetic moment. We differentiated between a static and a dynamic sample sourced by a quantum spin. The interaction with the former preserves the purity of the electron's motional state, whereas the latter introduces backaction on the sample and results in a mixed state for the reduced electron state. For both cases, we derived the scattering operator under the slowly-varying envelope approximation.

Based on the scattering description, we explored the limits of two metrological tasks. The first task is the estimation of the strength of the magnetic moment in the framework of quantum sensing. Our analysis is based on the Fisher information, which characterizes the measurement precision. While the classical Fisher information (CFI) depends on the measurement basis, the quantum Fisher information (QFI) is basis-independent and serves as the optimal upper bound for the CFI. We derived the QFI and compared it

with the CFI in several bases. The results are summarized in Table 4.1.

All these results can be expressed in terms of the expectation value $\mathcal{J}_{\text{NB}}^{\text{max}} = 2 \langle \psi_0 | g(\hat{r})^2 | \psi_0 \rangle$ with respect to the motional initial state, which we assume to be radial. In the case without backaction, the QFI is proportional to the portion of the magnetic moment that points in a direction perpendicular to the beam axis, $|\mathbf{n}_\perp|$. The measurements in momentum and, for small parameter, in angular momentum reach the bound given by the QFI and are thus optimal. The fact that momentum measurements are a standard procedure in electron microscopy makes the corresponding result relevant for experiments. The position measurement, on the other hand, is completely insensitive to the magnetic moment.

The results change when we consider the case with backaction. In this case and for small ϑ the QFI is double as high as before. This value is only reached for angular momentum measurements. For a properly aligned sample spin, however, we can still reach half of the QFI with a momentum measurement. Since an experimental realization of the angular momentum measurement is unclear, the momentum measurement remains a feasible options. Notice that the momentum space CFI is basically the same for both cases, no backaction and backaction. The only difference is that the static direction of the magnetic moment $\mathbf{n}_\perp$ has to be replaced by the corresponding Bloch vector $\mathbf{c}$, which characterizes the initial spin state of the sample.

	No backaction	Backaction
QFI	$ \mathbf{n}_\perp ^2 \mathcal{J}_{\text{NB}}^{\text{max}}$	$2\mathcal{J}_{\text{NB}}^{\text{max}} + \mathcal{O}(\vartheta^2)$
CFI(position)	0	0
CFI(momentum)	$ \mathbf{n}_\perp ^2 \mathcal{J}_{\text{NB}}^{\text{max}}$	$ \mathbf{c}_\perp ^2 \mathcal{J}_{\text{NB}}^{\text{max}} + \mathcal{O}(\vartheta^2)$
CFI(ang. mom.)	$ \mathbf{n}_\perp ^2 \mathcal{J}_{\text{NB}}^{\text{max}} + \mathcal{O}(\vartheta^2)$	$2\mathcal{J}_{\text{NB}}^{\text{max}} + \mathcal{O}(\vartheta^2)$

Table 4.1.: Comparison between QFI and CFI for measurements in image mode (position space), diffraction mode (momentum space), and of angular momenta, for the cases with and without backaction. The initial wavefunction $|\psi_0\rangle$ is assumed to be radial. All results are given in terms of the maximal QFI in the case without backaction, $\mathcal{J}_{\text{NB}}^{\text{max}} = 2 \langle \psi_0 | g(\hat{r})^2 | \psi_0 \rangle$. The vector $\mathbf{n}$ denotes the direction of the classical magnetic moment, where $0 \leq |\mathbf{n}_\perp| \leq 1$ describes the portion in the plane orthogonal to the beam axis, $|\mathbf{n}_\perp| = \sqrt{n_x^2 + n_y^2}$. Similarly, $\mathbf{c}$ denotes the initial Bloch vector of the quantum mechanical source of the magnetic moment and $\mathbf{c}_\perp$ its projection in the $x - y$ -plane. When no scaling $\mathcal{O}(\vartheta^n)$ is indicated, the result holds for all parameters ϑ . For no backaction, the momentum measurement as well as the angular momentum measurement yield the optimal precision, while position measurements are completely insensitive to magnetic moments. In the backaction case, on the other hand, the angular momentum measurement is still optimal, while the momentum measurement can at best reach half of the optimum. Note the subtle difference that the QFI in absence of backaction depends crucially on $|\mathbf{n}_\perp|$, while the QFI in the case with backaction is independent of the coherence $|\mathbf{c}_\perp|$.

4. Metrology for scattering interactions in electron microscopes

The second metrological task, we did consider, was state discrimination. In this case, a magnetic moment of known strength is either positioned in the beam path or not. The task is to distinguish the two situations from measurement data. Here, the relevant quantity, which characterizes the success probability, is the basis-dependent classical trace distance. In analogy to the sensing task, the basis-independent quantum trace distance serves as the optimal upper bound of the classical trace distance. The optimal measurement is mathematically known and we present it for the case without backaction in Subsection 4.5.1. However, this measurement depends strongly on the state and a experimental realization is unclear.

The values of quantum and classical trace distance in various bases are presented in Table 4.2: Position measurements are still insensitive to the magnetic moment. The momentum measurement does not have an analytical form but, in the backactionless case, numerical results in Subsection 4.5.2 confirm that the classical trace distance is smaller than the quantum trace distance. The results also show that measurements in a defocused plane between position and momentum can improve the classical trace distance compared to momentum measurements. Measurements in the angular momentum basis were shown to be suboptimal, unless the $\mathbb{V}[J_0(\vartheta|\mathbf{n}_\perp|g(\hat{r}))]$ vanishes.

The scenario with backaction is not fully explored yet. We found an upper bound for the quantum trace distance. The angular momentum measurement does not reach this optimal value.

	No backaction	Backaction
D_Q	$\sqrt{1 - \langle J_0(\vartheta \mathbf{n}_\perp g(\hat{r})) \rangle_0^2}$	$\leq \frac{1- \mathbf{c} ^2}{2} + \frac{1+ \mathbf{c} ^2}{2} \sqrt{1 - \langle \cos(\vartheta g(\hat{r})) \rangle_0^2}$
$D_C(\text{position})$	0	0
$D_C(\text{momentum})$	-	-
$D_C(\text{ang. mom.})$	$1 - \langle J_0(\vartheta \mathbf{n}_\perp g(\hat{r}))^2 \rangle_0$	$\langle \sin(\vartheta g(\hat{r}))^2 \rangle_0$

Table 4.2.: Comparison of the trace distance with its classical analogous for measurement statistics in image mode (position space), diffraction mode (momentum space), and of angular momenta, for the cases with and without backaction. The used symbols are explained in Table 4.1. In the case without backaction

All things considered, the task of sensing magnetic moments is completed. The momentum measurement was shown to be optimal in the case without backaction and sufficient in the case with backaction. However, in future works the realizability of angular momentum measurements should be investigated. This could double the precision in the case without backaction.

The discrimination task is also well understood in the case without backaction. A momentum measurement is still efficient and can even be improved by going out of focus. For the case with backaction, a numerical analysis of momentum measurement and defocus planes has to be done.

Further research should consider the choice of the optimal initial state and could also include the spin degree of freedom of the electron. The connection between the two tasks

4. Metrology for scattering interactions in electron microscopes

could also be further explored. In the limit of small parameters, the Fisher information and the trace distance are clearly related in our case, see Table 4.3.

	No backaction	Backaction
D_Q	$\frac{\vartheta}{2} \mathbf{n}_\perp \sqrt{\mathcal{J}_{\text{NB}}^{\text{max}}} + \mathcal{O}(\vartheta^3)$	$\leq \frac{1- \mathbf{c} ^2}{2} + \frac{1+ \mathbf{c} ^2}{2} \frac{\vartheta}{\sqrt{2}} \sqrt{\mathcal{J}_{\text{NB}}^{\text{max}}} + \mathcal{O}(\vartheta^3)$
$D_C(\text{position})$	0	0
$D_C(\text{momentum})$	-	-
$D_C(\text{ang. mom.})$	$\frac{\vartheta^2}{4} \mathbf{n}_\perp ^2 \mathcal{J}_{\text{NB}}^{\text{max}} + \mathcal{O}(\vartheta^4)$	$\frac{\vartheta^2}{2} \mathcal{J}_{\text{NB}}^{\text{max}} + \mathcal{O}(\vartheta^4)$

Table 4.3.: Same scenario as in Table 4.2 but expanded for small parameters $\vartheta \ll 1$. In this regime, the distances can be expressed in terms of maximal non backaction QFI, $\mathcal{J}_{\text{NB}}^{\text{max}}$, too.

5. Conclusion

In this thesis, we presented two projects in the field of quantum thermodynamics and one project in the field of quantum metrology. In contrast to the majority of research in these areas, we put the focus on quantum systems that represent continuous-variable (and non-harmonic) mechanical degrees of freedom.

In the first work on *quantum thermodynamics*, we presented two realizations of the Otto cycle with a planar rotor as the working medium. For each realization, we compared between the cycle performance with a quantum and a classical rotor in order to identify genuine quantum effects. There, the main focus was on the operation regimes, i.e. whether the machine operates as an engine, a refrigerator, or a (useless) heater. In the first realization, the rotor resembles an electric dipole in an electric field of periodically modulated strength, which is mathematically equivalent to a mathematical pendulum. We showed that the parameter regime for which the machine admits a useful (engine or refrigerator) operation mode is consistently smaller in the quantum case, displaying true quantum disadvantage. In the second realization, the rotor represents a magnetic dipole moment associated to a ring current, which interacts with an external magnetic field of periodically modulated strength. It acquires potential energy proportional to the angular momentum and the field strength. We proved that, classically, this machine can neither operate as an engine nor as a refrigerator. This changes when we consider a quantum planar rotor close to its motional ground state: With the cold reservoir being in the deep quantum regime, a window of work production opens and the machine operates as an engine. Although the work output in this regime is dominated by fluctuations, it demonstrates a genuine quantum advantage, because engine operation is only made possible due to the quantization of angular momentum.

These findings have several implications for quantum thermodynamics: First, the example of an engine that is only possible due to quantum mechanics deepens the understanding of quantum advantage and how to reach it. We further show that unusual working media with a non-trivial operator algebra, such as the planar rotor, hold the potential to reveal many interesting and unexplored quantum features. Mechanical degrees of freedom are particularly useful in this regard, because they have a natural classical counterpart. The periodic nature of the rotor make it also an ideal starting point for the construction of autonomous heat engines [180], which do not require external timing control of the cycle strokes. Besides using the rotor as a working medium, future work could also investigate the transmission of motion in quantum gears, building on [181]. Due to the current progress in quantum rotor experiments [50], it might well be possible to realize one of the presented thermal machines in the near future.

The second project in *quantum thermodynamics* deals with the fundamental question of collisional open system dynamics in gas environments and proper thermalization

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therein. We presented a detailed master equation derivation from first principles for a gas with internal degrees of freedom, and we verified its consistency with the laws of thermodynamics. Unlike the often used repeated interaction models, our master equation correctly predicts proper thermalization to a Gibbs state with the gas temperature regardless of the mismatch between the internal resonances of the system and the gas particles. Intriguingly, we found that a non-equilibrium setting in which the internal and motional degrees of freedom of the gas are thermal with respect to different temperatures can act as an uncontrolled work source: For properly chosen parameters, a build-up of ergotropy can then be observed in the system.

Since a perfect vacuum is practically unattainable, the influence of residual gases must be taken into account in most experimental setups. This makes collisional open system dynamics a relevant environmental scenario. Although low-density master equations were discussed before, our derivation incorporates internal degrees of freedom for the gas particles, admitting potentially useful non-equilibrium scenarios of uncontrolled work production. In addition, our representation aligns with the modern language of thermodynamics, lowering the entry barrier for people from the field. Given that system and gas obey time-reversal symmetry, the system always thermalizes to a Gibbs state at the gas temperature. This is an advantage compared to repeated interaction models, where the environment must be specifically engineered to achieve proper thermalization. In contrast to one-dimensional scattering models [53], our three-dimensional version achieves thermodynamic consistency from the Maxwell-Boltzmann distribution of gas momenta at thermal equilibrium instead of an artificial effusion distribution [54].

While the decoherence rates of low-density master equations have already been checked experimentally in the diffusive limit [182], an experimental proof of proper thermalization remains open. In the future, we will consider an alternative derivation that drops the assumption of slow system dynamics, provided that the considered observables are close to diagonal in the energy basis.

In our *quantum metrology* project, we discussed the possibilities of sensing the magnetic moments of individual quantum spins in state-of-the-art electron microscopes. We derived the scattering operator for electrons interacting with two types of samples: one with a static magnetic moment that does not change due to the presence of the electron and one in which the magnetic moment is sourced by a quantum spin. In the second case, quantum backaction of the electron on the sample spin qualitatively changes the analysis. We derived the outcome statistics for the two cases by applying the respective scattering operator and detecting electrons in either the position, momentum, or angular momentum basis. From the statistics, we calculated the Fisher information for the cases with and without backaction and every measurement in order to find the basis with the best precision to sense the strength of the magnetic moment. We also derived the quantum Fisher information, which represents the optimal bound on the classical Fisher information. By comparison, we found that momentum and angular momentum measurements are optimal in the case without backaction. In the case with backaction, on the other hand, only the angular momentum measurement is optimal.

Besides sensing, we also explored quantum state discrimination between the scattered and unscattered state of an electron. There, the task is to decide whether a magnetic

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moment of a known strength was placed on the beam axis or not. We derived the optimal success probability to decide correctly, characterized by the (quantum) trace distance. Then we compared the classical trace distance in the before mentioned bases. In contrast to the Fisher information, the optimum is never reached in those bases.

Our assessment of scattering metrology with electrons extends previous work [11] to quantum-limited sensing of magnetic moments. By that, it contributes to the emerging field of free-electron quantum optics [10]. Since this work is a joint project with the experimental group of Philipp Haslinger, which is performing state-of-the-art experiments in the field [46–48], we are confident that the results will directly influence the design of upcoming experiments.

In follow-up works, we plan to investigate how to implement orbital angular momentum measurements on an electron beam, which we have shown to be optimal even in the case with backaction. Another open question is whether the results can be improved by considering initial electron beam wave packets that are not radially symmetric or that contain entanglement between subsequent electrons. Finally, we will also include the internal degrees of freedom of the electron to estimate their effect and to exploit them for more precise sensing.

All projects considered, we presented new results in the fields of quantum thermodynamics and quantum metrology. The three projects have in common that the systems under consideration are mechanical degrees of freedom, which led to results that would not have been possible with discrete finite-dimensional systems.

A. Appendix for Basics

A.1. Appendix for linear motion and scattering theory

A.1.1. Alternative definition of the scattering operator

Here, we want to show the equality of two different definitions of the scattering operator,

$$\hat{S} = \lim_{T \rightarrow \infty} \hat{U}_0^\dagger(T) \hat{U}(2T) \hat{U}_0(T)^\dagger = \lim_{T \rightarrow \infty} \mathcal{T} \exp \left(-\frac{i}{\hbar} \int_{-T}^T dt \hat{V}^{(I)}(t) \right), \quad (\text{A.1.1})$$

where it is sufficient to show this for any finite T . Note that the right hand side is the formal solution of the interaction picture time evolution mapping some state $|\psi^I(+T)\rangle \leftarrow |\psi^I(-T)\rangle$. This can also be achieved by the product,

$$|\psi^I(+T)\rangle \approx e^{-i\hat{V}^{(I)}(t_N)\Delta t/\hbar} e^{-i\hat{V}^{(I)}(t_{N-1})\Delta t/\hbar} \dots e^{-i\hat{V}^{(I)}(t_0)\Delta t/\hbar} |\psi^I(-T)\rangle, \quad (\text{A.1.2})$$

where $\Delta t = 2T/N$ and The formula becomes exact in the limit of $N \rightarrow \infty$. Recalling the definition of the interaction picture operator,

$$\hat{V}^{(I)}(t) = \hat{U}_0^\dagger(t) \hat{V} \hat{U}_0(t), \quad (\text{A.1.3})$$

we find,

$$e^{-i\hat{V}^{(I)}(t)\Delta t/\hbar} = \hat{U}_0^\dagger(t) e^{-i\hat{V}\Delta t/\hbar} \hat{U}_0(t). \quad (\text{A.1.4})$$

Altogether, we can write,

$$\begin{aligned} & \mathcal{T} \exp \left(-\frac{i}{\hbar} \int_{-T}^T dt \hat{V}^{(I)}(t) \right) \\ & \approx \hat{U}_0^\dagger(T) e^{-i\hat{V}\Delta t/\hbar} \underbrace{\left[\hat{U}_0(T) \hat{U}_0^\dagger(T - \Delta t) \right]}_{=\hat{U}_0(\Delta t)} e^{-i\hat{V}\Delta t/\hbar} \underbrace{\left[\hat{U}_0(T - \Delta t) \hat{U}_0^\dagger(T - 2\Delta t) \right]}_{=\hat{U}_0(\Delta t)} \dots \hat{U}_0(-T) \\ & = \hat{U}_0^\dagger(T) \left(e^{-i\hat{V}\Delta t/\hbar} \hat{U}_0(\Delta t) \right)^N \hat{U}_0^\dagger(T) \\ & = \hat{U}_0^\dagger(T) \left(e^{-i\hat{V}(2T/N)/\hbar} e^{-i\hat{H}_0(2T/N)/\hbar} \right)^N \hat{U}_0^\dagger(T), \end{aligned} \quad (\text{A.1.5})$$

where we approximated the evolution by the operator product (A.1.2), expressed the interaction picture operator by (A.1.3), combined the neighboring free evolutions, and summarized the repeating expressions with the power N . Note that, in the limit $N \rightarrow \infty$,

A. Appendix for Basics

the approximation in the first step becomes exact and the Trotter formula can be applied in the last expression to find,

$$\mathcal{T} \exp \left(-\frac{i}{\hbar} \int_{-T}^T dt \hat{V}^{(I)}(t) \right) = \hat{U}_0^\dagger(T) e^{-i(\hat{H}_0 + \hat{V})2T/\hbar} \hat{U}_0^\dagger(T) = \hat{U}_0^\dagger(T) \hat{U}(2T) \hat{U}_0^\dagger(T). \quad (\text{A.1.6})$$

Since this is true for all T , the limit $T \rightarrow \infty$ can be applied on both sides in order to find the desired result.

B. Appendix for Rotor engines

B.1. Variance of work

The operator of work and its variance for our two engine models from Chapter 2 are calculated in the following subsections.

B.1.1. General Formula

In Subsection 2.2.3 we encountered the operator of work in the form,

$$\hat{W}_{A \rightarrow B} = -\hat{W}_{C \rightarrow D} = (\lambda_c - \lambda_h)\hat{V}, \quad (\text{B.1.1})$$

where $\hat{V}$ is the controlled part of the Hamiltonian. The operator of work is evaluated with respect to the initial state of the corresponding trajectory, which are A and C in our case. For example, the work statistics along the unitary work stroke ($A \rightarrow B$) are given by the moments $\text{tr}\{\gamma_h \hat{W}_{A \rightarrow B}^n\}$. The total per-cycle work is the sum of both contributions, ($A \rightarrow B$) and ($C \rightarrow D$). Using that the end points of the two work strokes are independent, we can conveniently define the total full work operator on the tensor product of the Hilbert space with itself,

$$\begin{aligned} \hat{W} &= \hat{W}_{A \rightarrow B} \otimes \mathbb{1} + \mathbb{1} \otimes \hat{W}_{C \rightarrow D} \\ &= (\lambda_c - \lambda_h) \left(\hat{V} \otimes \mathbb{1} - \mathbb{1} \otimes \hat{V} \right), \end{aligned} \quad (\text{B.1.2})$$

which allows the calculation of arbitrary high moments with respect to the state $\hat{\gamma}_h \otimes \hat{\gamma}_c$.

In order to calculate the variance, we need the second moment of work, which reads,

$$\langle W^2 \rangle = (\lambda_c - \lambda_h)^2 \left(\langle V^2 \rangle_h - 2\langle V \rangle_h \langle V \rangle_c + \langle V^2 \rangle_c \right), \quad (\text{B.1.3})$$

while the squared expectation value becomes,

$$\langle W \rangle^2 = (\lambda_c - \lambda_h)^2 \left(\langle V \rangle_h^2 - 2\langle V \rangle_h \langle V \rangle_c + \langle V \rangle_c^2 \right). \quad (\text{B.1.4})$$

Upon subtraction, the mixed term vanishes and we find the work variance,

$$\mathbb{V}[W] = (\lambda_c - \lambda_h)^2 (\mathbb{V}_h[V] + \mathbb{V}_c[V]). \quad (\text{B.1.5})$$

This expression holds both for the quantum and the classical version.

B.1.2. Work variance of the electric dipole machine

For the electric dipole engine, the controlled part of the Hamiltonian reads,

$$\hat{V} = E \sin^2(\hat{\alpha}/2). \quad (\text{B.1.6})$$

Classically, its first and second moments with respect to both Gibbs states, hot and cold, can be calculated by taking the respective derivative of the classical partition function (2.3.2),

$$\left\langle \left[E \sin^2 \left(\frac{\alpha}{2} \right) \right]^n \right\rangle = \frac{(-k_B T)^n}{Z(\lambda, T)} \frac{\partial^n}{\partial \lambda^n} Z(\lambda, T). \quad (\text{B.1.7})$$

The derivatives can be obtained from elementary properties of the Bessel functions,

$$\left\langle E \sin^2 \left(\frac{\alpha}{2} \right) \right\rangle = \frac{E}{2} \left(1 - \frac{I_1(x)}{I_0(x)} \right) \quad (\text{B.1.8})$$

$$\left\langle \left[E \sin^2 \left(\frac{\alpha}{2} \right) \right]^2 \right\rangle = \frac{E^2}{2} \left[1 - \left(1 + \frac{1}{2x} \right) \frac{I_1(x)}{I_0(x)} \right], \quad (\text{B.1.9})$$

with the abbreviation $x = E\lambda/(2k_B T)$. From this the variance is found,

$$\mathbb{V} \left[E \sin^2 \left(\frac{\alpha}{2} \right) \right] = \frac{E^2}{4} \left[1 - \left(\frac{1}{x} + \frac{I_1(x)}{I_0(x)} \right) \frac{I_1(x)}{I_0(x)} \right]. \quad (\text{B.1.10})$$

For the quantum version, we use numerical methods.

B.1.3. Work variance of the magnetic dipole machine

Since the magnetic dipole engine only operates in the quantum regime, it suffices to calculate the variance for this scenario, where the controlled part of the Hamiltonian is proportional to the angular momentum operator,

$$\hat{V} = E \frac{\hat{L}_z}{\hbar}. \quad (\text{B.1.11})$$

Rewriting the expectation value of energy Eq. (2.4.2) and using the classical high temperature limit $\langle L_z \rangle_h \approx \hbar \lambda_h$ from (2.4.6), we find the second moment of angular momentum,

$$\begin{aligned} \langle L_z^2 \rangle_h &= 2\hbar^2 \left(\frac{\langle H_h \rangle_h}{E} + \lambda \frac{\langle L_z \rangle_h}{\hbar} \right) \\ &\approx \frac{2\hbar^2}{E} \langle H_h \rangle_h + 2(\hbar \lambda_h)^2. \end{aligned} \quad (\text{B.1.12})$$

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With this we can express the variance of angular momentum by the expectation value of $\hat{H}_h$, which we already derived in Eq. (2.4.7),

$$\mathbb{V}_h[L_z] = \frac{\hbar^2 k_B T_h}{E} = I k_B T_h. \quad (\text{B.1.13})$$

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C.1. Collision models

C.1.1. Calculation for Flip-Flop

Since all $\hat{S}_j$ commute with $\hat{F}_z$, we get,

$$\hat{U}(t) = \left(\cos(\bar{\omega}t) - i \sin(\bar{\omega}t) \hat{F}_z \right) \left(\cos(\Delta t) - i \sin(\Delta t) \mathbf{n} \cdot \hat{\mathbf{S}} \right). \quad (\text{C.1.1})$$

This can be explicitly written as,

$$\begin{aligned} \hat{U}(t) = & e^{-i\bar{\omega}t} |11\rangle \langle 11| + e^{i\bar{\omega}t} |00\rangle \langle 00| \\ & + \cos(\Delta t) (|01\rangle \langle 01| + |10\rangle \langle 10|) \\ & - i \sin(\Delta t) \left(\frac{g_0}{\Delta} (|01\rangle \langle 10| + |10\rangle \langle 01|) + \frac{\delta}{\Delta} (|10\rangle \langle 10| - |01\rangle \langle 01|) \right). \end{aligned} \quad (\text{C.1.2})$$

Assume that the second qubit is initially in a thermal state of the form $\hat{\rho}(p) = (1 - p) |1\rangle \langle 1| + p |0\rangle \langle 0|$. With this the Krauss operators for the first qubit become $\hat{K}_{ij}(t) = \langle i|_2 \hat{U}(t) |j\rangle_2$,

$$\begin{aligned} \hat{K}(t)_{11} &= e^{-i\bar{\omega}t} |1\rangle \langle 1| + A(t) |0\rangle \langle 0| \\ \hat{K}(t)_{00} &= e^{i\bar{\omega}t} |0\rangle \langle 0| + A^*(t) |1\rangle \langle 1| \\ \hat{K}(t)_{10} &= -i \frac{g_0}{\Delta} \sin(\Delta t) |0\rangle \langle 1| \\ \hat{K}(t)_{01} &= -i \frac{g_0}{\Delta} \sin(\Delta t) |1\rangle \langle 0|, \end{aligned} \quad (\text{C.1.3})$$

where $A(t) = \cos(\Delta t) + i \frac{\delta}{\Delta} \sin(\Delta t)$. We parametrize the first qubit by the bloch vector $\vec{r} = (r_x, r_y, r_z)$,

$$\hat{\rho}_S = \frac{1}{2} (\mathbb{1} + \vec{r} \cdot \hat{\boldsymbol{\sigma}}) = \begin{pmatrix} (1 + r_z)/2 & (r_x - ir_y)/2 \\ (r_x + ir_y)/2 & (1 - r_z)/2 \end{pmatrix}. \quad (\text{C.1.4})$$

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The channel can be written as,

$$\begin{aligned}
\Lambda_t [\hat{\rho}_S] &= \text{tr} \left\{ \hat{U}(t) \hat{\rho}_S \otimes ((1-p) |1\rangle \langle 1| + p |0\rangle \langle 0|) \hat{U}^\dagger(t) \right\} \\
&= \left[p \frac{1-r_z}{2} + (1-p) |A(t)|^2 \frac{1-r_z}{2} + p \left(\frac{g_0}{\Delta} \right)^2 \frac{1+r_z}{2} \sin^2(\Delta t) \right] |0\rangle \langle 0| \\
&+ \left[(1-p) \frac{1+r_z}{2} + p |A(t)|^2 \frac{1+r_z}{2} + (1-p) \left(\frac{g_0}{\Delta} \right)^2 \frac{1-r_z}{2} \sin^2(\Delta t) \right] |1\rangle \langle 1| \\
&+ e^{-i\bar{\omega}t} A^*(t) \frac{r_x - ir_y}{2} |1\rangle \langle 0| + e^{i\bar{\omega}t} A(t) \frac{r_x + ir_y}{2} |0\rangle \langle 1|. \tag{C.1.5}
\end{aligned}$$

For later use, it is convenient to be aware of the following ways to rewrite $|A(t)|^2$

$$\begin{aligned}
|A(t)|^2 &= \cos^2(\Delta t) + \left(\frac{\delta}{\Delta} \right)^2 \sin^2(\Delta t) \\
&= \left(\frac{g_0}{\Delta} \right)^2 \cos^2(\Delta t) + \left(\frac{\delta}{\Delta} \right)^2 \\
&= 1 - \left(\frac{g_0}{\Delta} \right)^2 \sin^2(\Delta t). \tag{C.1.6}
\end{aligned}$$

From the second and third line we can also deduce the important inequalities,

$$\frac{|\delta|}{\Delta} \leq |A(t)| \leq 1 \tag{C.1.7}$$

We can now use $p(1-r_z)/2 = p - p(1+r_z)/2$ in order to find the diagonal entries of the transformed density matrix,

$$\begin{aligned}
\langle 0 | \Lambda_t [\hat{\rho}_S] | 0 \rangle &= p + (1-p) |A(t)|^2 \frac{1-r_z}{2} + p \frac{1+r_z}{2} \left(\left(\frac{g_0}{\Delta} \right)^2 \sin^2(\Delta t) - 1 \right) \\
&= p + (1-p) |A(t)|^2 \frac{1-r_z}{2} - p \frac{1+r_z}{2} |A(t)|^2 \\
&= p + \frac{|A(t)|^2}{2} ([1-2p] - r_z). \tag{C.1.8}
\end{aligned}$$

Due to normalization this implies,

$$\langle 1 | \Lambda_t [\hat{\rho}_S] | 1 \rangle = 1 - \langle 0 | \Lambda_t [\hat{\rho}_S] | 0 \rangle = (1-p) - \frac{|A(t)|^2}{2} ([1-2p] - r_z) \tag{C.1.9}$$

and the transformation rule of the z -component of the Bloch vector,

$$r_z \xrightarrow{\Lambda_t} (1-2p) - |A(t)|^2 ([1-2p] - r_z). \tag{C.1.10}$$

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For the non-diagonal terms we use the polar decomposition of $A(t) = |A(t)|e^{i\varphi(t)}$,

$$A(t)e^{i\bar{\omega}t} = |A(t)|e^{i(\bar{\omega}t+\varphi(t))} = |A(t)|e^{i\vartheta(t)} \quad (\text{C.1.11})$$

and then use,

$$\begin{aligned} & e^{i\vartheta(t)} \frac{r_x + ir_y}{2} |0\rangle \langle 1| + \text{h.c.} \\ &= \frac{1}{2} (\cos(\vartheta(t))r_x - \sin(\vartheta(t))r_y) \hat{\sigma}_x + \frac{1}{2} (\sin(\vartheta(t))r_x + \cos(\vartheta(t))r_y) \hat{\sigma}_y \\ &= \frac{1}{2} \left[\underbrace{\begin{pmatrix} \cos(\vartheta(t)) & -\sin(\vartheta(t)) & 0 \\ \sin(\vartheta(t)) & \cos(\vartheta(t)) & 0 \\ 0 & 0 & 1 \end{pmatrix}}_{=:R(\mathbf{e}_z, \vartheta(t))} \begin{pmatrix} r_x \\ r_y \\ 0 \end{pmatrix} \right] \cdot \hat{\boldsymbol{\sigma}}. \end{aligned} \quad (\text{C.1.12})$$

Note that the phase term applied on $r_x + ir_y$ is just a rotation of the Bloch vector around the z -axis. In total we can summarize the effect of the channel by,

$$\Lambda_t [\hat{\rho}_S] = \hat{\rho}_{\text{th}}(p) + \frac{\hat{\boldsymbol{\sigma}}}{2} \cdot |A(t)| R(\mathbf{e}_z, \bar{\omega}t + \vartheta(t)) \begin{pmatrix} r_x \\ r_y \\ -|A(t)| ([1 - 2p] - r_z) \end{pmatrix} \quad (\text{C.1.13})$$

C.2. Derivation of master equation

C.2.1. Delta identity

Here, we derive an identity needed for Subsection 3.2.4,

$$\delta^{(1)} \left(\frac{\mathbf{p}^2}{2m} - \tilde{E} \right) = \begin{cases} \frac{m}{|\mathbf{p}|} \delta^{(1)} \left(|\mathbf{p}| - \sqrt{2m\tilde{E}} \right), & \text{for } \tilde{E} \geq 0 \\ 0, & \text{else} \end{cases}. \quad (\text{C.2.1})$$

This can easiest seen by going to spherical coordinates and substitute $p \, dp = m \, dE$, assume $\tilde{E} \geq 0$

$$\begin{aligned} \int d^3\mathbf{p} \, \delta^{(1)} \left(\frac{\mathbf{p}^2}{2m} - \tilde{E} \right) f(\mathbf{p}) &= m \int d\Omega \int_{E>0} dE \, \delta^{(1)} (E - \tilde{E}) \sqrt{2mE} \, f(\sqrt{2mE}\mathbf{n}(\Omega)) \\ &= \int d\Omega \int dp \, p^2 \frac{m}{p} \delta^{(1)} (p - \sqrt{2m\tilde{E}}) f(p\mathbf{n}(\Omega)) \\ &= \int d^3\mathbf{p} \, \frac{m}{|\mathbf{p}|} \delta^{(1)} (|\mathbf{p}| - \sqrt{2m\tilde{E}}) f(\mathbf{p}), \end{aligned} \quad (\text{C.2.2})$$

for any test function f . For $\tilde{E} < 0$, on the other hand, $\delta^{(1)} (E - \tilde{E})$ is effectively zero for all $E > 0$.

C.2.2. Tracing out motion

We start by calculating the matrix element,

$$\begin{aligned}
 & \langle \kappa_2, \mathbf{p} | \hat{T}^\dagger \hat{T} | \kappa_1, \mathbf{p} \rangle \\
 &= \sum_{\nu} \int d^3 \mathbf{q} \langle \kappa_2, \mathbf{p} | \hat{T}^\dagger | \nu, \mathbf{q} \rangle \langle \nu, \mathbf{q} | \hat{T} | \kappa_1, \mathbf{p} \rangle \\
 &= \sum_{\nu} \int \frac{d^3 \mathbf{q}}{(2\pi\hbar m)^2} \delta^1(E_{\nu}(\mathbf{q}) - E_{\kappa_1}(\mathbf{p})) \delta^1(E_{\nu}(\mathbf{q}) - E_{\kappa_2}(\mathbf{p})) f_{\nu\kappa_2}^*(\mathbf{q}, \mathbf{p}) f_{\nu\kappa_1}(\mathbf{q}, \mathbf{p}) \\
 &= \frac{\Delta t}{(2\pi\hbar)^3 m} \delta^{(0)}(\Delta_{\kappa_2\kappa_1}) \sum_{\nu} \int \frac{d^3 \mathbf{q}}{|\mathbf{q}|} \delta^{(1)}(|\mathbf{q}| - \sqrt{\mathbf{p}^2 - 2m\Delta_{\nu\kappa_1}}) f_{\nu\kappa_2}^*(\mathbf{q}, \mathbf{p}) f_{\nu\kappa_1}(\mathbf{q}, \mathbf{p}) \\
 &= \frac{\Delta t p}{(2\pi\hbar)^3 m} \delta^{(0)}(\Delta_{\kappa_2\kappa_1}) \sum_{\nu} \int d\Omega \frac{P_{\nu\kappa_1}(\mathbf{p})}{|\mathbf{p}|} f_{\nu\kappa_2}^*(\mathbf{P}_{\nu\kappa_2}(\Omega_{\mathbf{p}}), \mathbf{p}) f_{\nu\kappa_1}(\mathbf{P}_{\nu\kappa_1}(\Omega_{\mathbf{p}}), \mathbf{p}), \quad (\text{C.2.3})
 \end{aligned}$$

where we inserted an identity between the T operators, expressed the T operators by the scattering amplitude, used the delta identity from the main text and integrated over radial momentum in spherical coordinates.

With the matrix element at hand, we find,

$$\begin{aligned}
 & \int d^3 \mathbf{q} \left\{ \langle \mathbf{p} | \hat{T}^\dagger | \mathbf{q} \rangle \langle \mathbf{q} | \hat{T} | \mathbf{p} \rangle, \hat{\rho}_{\text{SA}} \right\} = \left\{ \langle \mathbf{p} | \hat{T}^\dagger \hat{T} | \mathbf{p} \rangle, \hat{\rho}_{\text{SA}} \right\} \quad (\text{C.2.4}) \\
 &= \frac{1}{(2\pi\hbar)^3} \frac{p\Delta t}{m} \int d\Omega \left\{ \sum_{\nu, \kappa_1, \kappa_2}^{\epsilon_{\kappa_2} = \epsilon_{\kappa_1}} \frac{P_{\nu\kappa_1}}{p} f_{\nu\kappa_2}^*(\mathbf{P}_{\nu\kappa_2}(\Omega_{\mathbf{p}}), \mathbf{p}) f_{\nu\kappa_1}(\mathbf{P}_{\nu\kappa_1}(\Omega_{\mathbf{p}}), \mathbf{p}) | \kappa_2 \rangle \langle \kappa_1 |, \hat{\rho}_{\text{SA}} \right\}
 \end{aligned}$$

A straightforward calculation verifies,

$$\sum_E \hat{L}_E(\Omega, \mathbf{p})^\dagger \hat{L}_E(\Omega, \mathbf{p}) = \sum_{\nu, \kappa_1, \kappa_2}^{\epsilon_{\kappa_2} = \epsilon_{\kappa_1}} \frac{P_{\nu\kappa_1}}{p} f_{\nu\kappa_2}^*(\mathbf{P}_{\nu\kappa_2}(\Omega_{\mathbf{p}}), \mathbf{p}) f_{\nu\kappa_1}(\mathbf{P}_{\nu\kappa_1}(\Omega, \mathbf{p}), \mathbf{p}) | \kappa_2 \rangle \langle \kappa_1 |, \quad (\text{C.2.5})$$

which completes the derivation.

C.2.3. Trace out ancilla

In order to trace out the ancilla degrees of freedom from the dissipator, we use linearity of the partial trace to focus on the trace of a single Lindblad operator,

$$\begin{aligned}
 \text{tr}_A \left\{ \mathcal{D} \left[\hat{L}_E(\Omega, \mathbf{p}) \right] \hat{\rho}_S \otimes \hat{\gamma}_A \right\} &= \sum_{k_A} p^{k_A} \text{tr}_A \left\{ \mathcal{D} \left[\hat{L}_E(\Omega, \mathbf{p}) \right] \hat{\rho}_S \otimes |k_A\rangle \langle k_A| \right\} \\
 &= \sum_{n_A, k_A} p^{k_A} \langle n_A | \left(\mathcal{D} \left[\hat{L}_E(\Omega, \mathbf{p}) \right] \hat{\rho}_S \otimes |k_A\rangle \langle k_A| \right) | n_A \rangle \\
 &= \sum_{n_A, k_A} p^{k_A} \mathcal{D} \left[\langle n_A | \hat{L}_E(\Omega, \mathbf{p}) | k_A \rangle \right] \hat{\rho}_S, \tag{C.2.6}
 \end{aligned}$$

where we expanded $\hat{\gamma}_A$ in the energy eigen basis, wrote out the partial trace in the energy eigen basis with the operators,

$$\langle n_A | \hat{L}_E(\Omega, \mathbf{p}) | k_A \rangle = \sum_{n_S, k_S}^{\Delta_{n_S, k_S}^{n_A, k_A} = E} \sqrt{\frac{P_{n_S, n_S}^{n_A, k_A}(p)}{p}} f_{n_S, k_S}^{n_A, k_A} \left(\mathbf{P}_{n_S, k_S}^{n_A, k_A}(\Omega, p), \mathbf{p} \right) |n_S\rangle \langle k_S|. \tag{C.2.7}$$

Further, we can simplify the energy conservation condition $\Delta_{n_S, k_S}^{n_A, k_A} = E$ under the $\sum_E$ sum because the same indices n_A, k_A appear in both Lindblad operators: For initial $\hat{\rho}_S = |k_S\rangle \langle k'_S|$, we find,

$$\begin{aligned}
 &\langle n_S | \left(\sum_E \langle n_A | \hat{L}_E(\Omega, \mathbf{p}) | k_A \rangle \hat{\rho}_S \langle k_A | \hat{L}_E^\dagger(\Omega, \mathbf{p}) | n_A \rangle \right) | n'_S \rangle \\
 &\propto \sum_E \delta^{(0)} \left(\Delta_{n_S, k_S}^{n_A, k_A} - E \right) \delta^{(0)} \left(\Delta_{n'_S, k'_S}^{n_A, k_A} - E \right) \\
 &= \delta^{(0)} \left(\Delta_{n_S, k_S} - \Delta_{n'_S, k'_S} \right) \underbrace{\sum_E \delta^{(0)} \left(\Delta_{n'_S, k'_S}^{n_A, k_A} - E \right)}_{=1} \\
 &= \sum_E \delta^{(0)} \left(\Delta_{n_S, k_S} - E \right) \delta^{(0)} \left(\Delta_{n'_S, k'_S} - E \right), \tag{C.2.8}
 \end{aligned}$$

where we replaced E by $\Delta_{n'_S, k'_S}^{n_A, k_A}$ in the first Kronecker delta by the virtue of the second one, noted that the remaining sum yields one, and in the end, we factorized the remaining term by introducing a new sum over energies E . The same procedure can be used in the other terms of the dissipator and we find the new Lindblad operators,

$$\hat{L}_E^{n_A, k_A}(\mathbf{p}, \Omega) = \sum_{n_S, k_S}^{\Delta_{n_S, k_S} = E} \sqrt{\frac{P_{n_S, n_S}^{n_A, k_A}(p)}{p}} f_{n_S, k_S}^{n_A, k_A} \left(\mathbf{P}_{n_S, k_S}^{n_A, k_A}(\Omega, p), \mathbf{p} \right) |n_S\rangle \langle k_S|. \tag{C.2.9}$$

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For the Hamiltonian we note,

$$\begin{aligned}
\text{tr}_A \left\{ \left[\hat{h}_{\text{eff,SA}}, \hat{\rho}_S \otimes \hat{\gamma}_A \right] \right\} &= \sum_{n_A, k_A} p^{k_A} \langle n_A | \left[\hat{h}_{\text{eff,SA}}, \hat{\rho}_S \otimes |k_A\rangle \langle k_A| \right] | n_A \rangle \\
&= \sum_{k_A} p^{k_A} \left[\langle k_A | \hat{h}_{\text{eff,SA}} | k_A \rangle, \hat{\rho}_S \right] \\
&= \left[\sum_{k_A} p^{k_A} \langle k_A | \hat{h}_{\text{eff,SA}} | k_A \rangle, \hat{\rho}_S \right]
\end{aligned} \tag{C.2.10}$$

where we expanded the trace and the thermal state in the energy eigen basis, used the linearity of commutator and trace and used a Kronecker delta that appeared in both terms of the commutator. In the last step, we absorbed the sum back in the commutator in order to find the effective Hamiltonian

$$\begin{aligned}
\hat{h}_{\text{eff}} &= \sum_{k_A} p^{k_A} \langle k_A | \hat{h}_{\text{eff,SA}} | k_A \rangle \\
&= -\frac{2\pi\hbar^2}{m} n \sum_{k_A} p^{k_A} \int d^3\mathbf{p} \mu(\mathbf{p}) \sum_{n_S, k_S}^{\epsilon_{n_S} = \epsilon_{k_S}} \frac{1}{2} \left(f_{n_S, k_S}^{k_A, k_A}(\mathbf{p}, \mathbf{p}) + f_{k_S, n_S}^{k_A, k_A}(\mathbf{p}, \mathbf{p}) \right) |n_S\rangle \langle k_S|.
\end{aligned} \tag{C.2.11}$$

C.2.4. Lindblad operators for Gaussian interaction

For $\hat{\rho}_S = |k_S\rangle \langle k'_S|$ we find,

$$\begin{aligned}
&\sum_E \int d^3\mathbf{p} \mu(\mathbf{p}) |\mathbf{j}(\mathbf{p})| \int d\Omega \hat{L}_E^{n_A, k_A} \hat{\rho}_S \hat{L}_E^{n_A, k_A \dagger} = \frac{\pi}{2} R^2 \left(\frac{V_0}{E_R} \right)^2 \sum_{n_S, n'_S}^{\Delta_{n_S, k_S} = \Delta_{n'_S, k'_S}} v_{n_S k_S}^{n_A k_A} \left(v_{n'_S k'_S}^{n_A k_A} \right)^* \\
&\underbrace{\int d^3\mathbf{p} \mu(\mathbf{p}) |\mathbf{j}(\mathbf{p})| \int d\Omega \frac{P_{n_S k_S}^{n_A k_A}(p)}{p} \exp \left(-\frac{\left(\mathbf{P}_{n_S k_S}^{n_A k_A}(\Omega, p) - \mathbf{p} \right)^2}{p_R^2} \right)}_{=\text{Int}},
\end{aligned} \tag{C.2.12}$$

where we used the Kronecker deltas, executed the sum over E and sorted the terms according to their dependence. In order to simplify the integral expression Int, we first simplify the exponential by noting,

$$\begin{aligned}
\left(\mathbf{P}_{n_S k_S}^{n_A k_A}(\Omega, p) - \mathbf{p} \right)^2 &= \left(\mathbf{p}^2 - 2m\Delta_{n_S k_S}^{n_A k_A} \right) - 2\mathbf{p} \cdot \mathbf{P}_{n_S k_S}^{n_A k_A}(\Omega, p) + \mathbf{p}^2 \\
&= 2p^2 - 2m\Delta_{n_S k_S}^{n_A k_A} - 2pP_{n_S k_S}^{n_A k_A}(p) \cos(\vartheta),
\end{aligned} \tag{C.2.13}$$

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where we assumed that the z -axis in the Ω -integration is in direction of $\mathbf{p}$. Now, we can go on by executing the solid angle integration,

$$\begin{aligned}
\text{Int} &= e^{\Delta_{n_S k_S}^{n_A k_A}/E_R} \int d^3\mathbf{p} \mu(\mathbf{p}) |\mathbf{j}(\mathbf{p})| e^{-2(p/p_R)^2} \frac{P_{n_S k_S}^{n_A k_A}(p)}{p} 2\pi \int_{-1}^1 d\cos(\vartheta) \exp\left(2 \frac{p P_{n_S k_S}^{n_A k_A}(p)}{p_R^2} \cos(\vartheta)\right) \\
&= 2\pi e^{\Delta_{n_S k_S}^{n_A k_A}/E_R} \int d^3\mathbf{p} \mu(\mathbf{p}) |\mathbf{j}(\mathbf{p})| e^{-2(p/p_R)^2} \frac{P_{n_S k_S}^{n_A k_A}(p)}{p} \frac{p_R^2}{p P_{n_S k_S}^{n_A k_A}(p)} \sinh\left(2 \frac{p P_{n_S k_S}^{n_A k_A}(p)}{p_R^2}\right) \\
&= 2\pi p_R^2 e^{\Delta_{n_S k_S}^{n_A k_A}/E_R} \int \frac{d^3\mathbf{p}}{p^2} \mu(\mathbf{p}) |\mathbf{j}(\mathbf{p})| e^{-2(p/p_R)^2} \sinh\left(2 \frac{p P_{n_S k_S}^{n_A k_A}(p)}{p_R^2}\right) \\
&= 8\pi^2 n p_R^2 e^{\Delta_{n_S k_S}^{n_A k_A}/E_R} \int_0^\infty \frac{dp}{m} \frac{p}{\sqrt{2\pi p_{\text{th}}^2}^3} e^{-(p/p_{\text{th}})^2/2} e^{-2(p/p_R)^2} \sinh\left(2 \frac{p P_{n_S k_S}^{n_A k_A}(p)}{p_R^2}\right), \tag{C.2.14}
\end{aligned}$$

where we performed the solid angle integration, canceled some terms, used $|\mathbf{j}(\mathbf{p})| = np/m$, and performed the angular part of the $\mathbf{p}$ integration. Now, we can substitute $E = p^2/(2m)$ and find,

$$\begin{aligned}
\text{Int} &= \frac{2\sqrt{2\pi} n p_R^2}{p_{\text{th}}^3} e^{\Delta_{n_S k_S}^{n_A k_A}/E_R} \int_{\min\{0, \Delta_{n_S k_S}^{n_A k_A}\}}^\infty dE \exp\left(-\left(2 + \frac{E_R}{k_B T}\right) \frac{E}{E_R}\right) \sinh\left(2\sqrt{\frac{E}{E_R}} \sqrt{\frac{E - \Delta_{n_S k_S}^{n_A k_A}}{E_R}}\right) \\
&= \frac{2\sqrt{2\pi} n p_R^2}{p_{\text{th}}^3} E_R I\left(\frac{E_R}{k_B T}, \frac{\Delta_{n_S k_S}^{n_A k_A}}{E_R}\right), \tag{C.2.15}
\end{aligned}$$

where we put the implicit condition that the pre scattering kinetic energy has to be positive and bigger than the internal energy gap $E > \max\{0, \Delta_{n_S k_S}^{n_A k_A}\}$ explicitly on the limit of the integral, substituted $z = E/E_R$, and found the parametric one-dimensional integral,

$$I(\alpha, s) = e^s \int_{\max\{0, s\}}^\infty dz e^{-(2+\alpha)z} \sinh(2\sqrt{z}\sqrt{z-s}). \tag{C.2.16}$$

With this, the original expression becomes,

$$\begin{aligned}
&\sum_E \int d^3\mathbf{p} \mu(\mathbf{p}) |\mathbf{j}(\mathbf{p})| \int d\Omega \hat{L}_E^{n_A, k_A} \hat{\rho}_S \hat{L}_E^{n_A, k_A \dagger} \\
&= \Gamma \sum_E \hat{J}_E^{n_A k_A} \hat{\rho}_S \hat{J}_E^{n_A k_A \dagger}, \tag{C.2.17}
\end{aligned}$$

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where we defined the rate Γ and the new Lindblad operators $\hat{J}_E^{n_A k_A}$,

$$\Gamma = \sqrt{2\pi^3} (nR^3) \left(\frac{V_0}{E_R} \right)^2 \left(\frac{p_R}{p_{\text{th}}} \right)^3 \frac{E_R}{\hbar} \quad (\text{C.2.18})$$

$$\hat{J}_E^{n_A k_A} = \sum_{n_S, k_S}^{\Delta_{n_S, k_S} = E} \sqrt{I \left(\frac{E_R}{k_B T}, \frac{\Delta_{n_S k_S}^{n_A k_A}}{E_R} \right)} \langle n_S, n_A | \hat{v} | n_S, n_A \rangle | n_S \rangle \langle k_S |. \quad (\text{C.2.19})$$

Similar calculation reveals that the anti-commutator part can be treated similarly such that the dissipator becomes,

$$\mathcal{C}\hat{\rho}_S = \sum_E \sum_{n_A, k_A} \mathcal{D} \left[\hat{J}_E^{n_A k_A} \right] \hat{\rho}_S. \quad (\text{C.2.20})$$

C.2.5. Properties of the integral

Here, we discuss the parametric integral,

$$I(\alpha, s) = e^s \int_{\max\{0, s\}}^{\infty} dz e^{-(2+\alpha)z} \sinh(2\sqrt{z}\sqrt{z-s}). \quad (\text{C.2.21})$$

Local detailed balance is manifested in $I(\alpha, s)$:

Observation 1 *The physical local detailed balance follows from the following mathematical symmetry,*

$$I(\alpha, -s) = e^{\alpha s} I(\alpha, s). \quad (\text{C.2.22})$$

Calculation 1 *This can be seen from the substitution $y = z + s$,*

$$\begin{aligned} I(\alpha, -s) &= e^{-s} \int_{\max\{0, -s\}}^{\infty} dz e^{-(2+\alpha)z} \sinh(2\sqrt{z}\sqrt{z+s}) \\ &= e^{-s} \int_{\max\{s, 0\}}^{\infty} dy e^{-(2+\alpha)(y-s)} \sinh(2\sqrt{y-s}\sqrt{y}) \\ &= e^{\alpha s} e^s \int_{\max\{s, 0\}}^{\infty} dy e^{-(2+\alpha)y} \sinh(2\sqrt{y}\sqrt{y-s}) \\ &= e^{\alpha s} I(\alpha, s). \end{aligned}$$

Note that this allows us to reduce all calculations to $I(\alpha, -|s|)$, which simplifies the integral by making the lower bound of the integral constant zero.

Observation 2 *For all α and s we have the inequality,*

$$\frac{2e^{-|s|}}{\alpha(\alpha+4)} \leq I(\alpha, -|s|) \leq \frac{1}{2} \left(\frac{1}{\alpha} - \frac{e^{-2|s|}}{4+\alpha} \right) \quad (\text{C.2.23})$$

Calculation 2 We use the monotonicity of the square root in order to obtain $z \leq \sqrt{z}\sqrt{z+|s|} \leq z+|s|$. With the monotonicity of the hyperbolic sine and the integral, on the other hand, we get the inequality of integrals,

$$e^{-|s|} \int_0^\infty dz e^{-(2+\alpha)z} \sinh(2z) \leq I(\alpha, -|s|) \leq e^{-|s|} \int_0^\infty dz e^{-(2+\alpha)z} \sinh(2(z+|s|)),$$

in which both bounds are elementary integrals.

This not only fixes the value of I for $s = 0$,

$$I(\alpha, 0) = \frac{2}{\alpha(\alpha+4)} \leq \frac{1}{2\alpha}, \quad (\text{C.2.24})$$

but also tells us that $I(\alpha, s)$ is suppressed exponentially for positive s .

C.2.6. Estimation of the effective rate

For $T_M = T_A = T$, we found the effective rate,

$$\tilde{\Gamma} = \Gamma = \frac{\cosh\left(\frac{\hbar\omega_S}{2k_B T}\right)}{\cosh\left(\frac{\hbar\omega_A}{2k_B T}\right)} \exp\left(\frac{\hbar(\omega_S - \omega_A)}{2k_B T}\right) I\left(\frac{E_R}{k_B T}, \frac{\hbar(\omega_S - \omega_A)}{E_R}\right). \quad (\text{C.2.25})$$

We will now show that this complicated expression fulfills,

$$\tilde{\Gamma} \leq \Gamma I\left(\frac{E_R}{k_B T}, -\frac{\hbar|\omega_S - \omega_A|}{E_R}\right). \quad (\text{C.2.26})$$

First, note that $I(\alpha, -s) = e^{\alpha s} I(\alpha, s)$ as shown in Observation (1). This implies,

$$e^{\alpha s/2} I(\alpha, s) = e^{\alpha(-s)/2} I(\alpha, -s) = e^{\alpha|s|/2} I(\alpha, |s|), \quad (\text{C.2.27})$$

which we can use to rewrite the last two terms. Further note for the first term,

$$\begin{aligned} \cosh\left(\frac{\hbar\omega_S}{2k_B T}\right) &= \cosh\left(\frac{\hbar\omega_A}{2k_B T} + \frac{\hbar(\omega_S - \omega_A)}{2k_B T}\right) \\ &= \cosh\left(\frac{\hbar\omega_A}{2k_B T}\right) \cosh\left(\frac{\hbar|\omega_S - \omega_A|}{2k_B T}\right) + \sinh\left(\frac{\hbar\omega_A}{2k_B T}\right) \sinh\left(\frac{\hbar(\omega_S - \omega_A)}{2k_B T}\right), \end{aligned} \quad (\text{C.2.28})$$

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where we used an addition theorem for hyperbolic functions. Hence, the fraction yields,

$$\begin{aligned} \frac{\cosh\left(\frac{\hbar\omega_S}{2k_B T}\right)}{\cosh\left(\frac{\hbar\omega_A}{2k_B T}\right)} &= \cosh\left(\frac{\hbar|\omega_S - \omega_A|}{2k_B T}\right) + \operatorname{sgn}(\omega_S - \omega_A) \tanh\left(\frac{\hbar\omega_A}{2k_B T}\right) \sinh\left(\frac{\hbar|\omega_S - \omega_A|}{2k_B T}\right) \\ &\leq \cosh\left(\frac{\hbar|\omega_S - \omega_A|}{2k_B T}\right) + \sinh\left(\frac{\hbar|\omega_S - \omega_A|}{2k_B T}\right) \\ &= \exp\left(\frac{\hbar|\omega_S - \omega_A|}{2k_B T}\right), \end{aligned} \quad (\text{C.2.29})$$

where used $\tanh(x) \in [0, 1)$ for $x \geq 0$ to find an upper bound. All things considered, we get,

$$\begin{aligned} \tilde{\Gamma} &= \Gamma \exp\left(\frac{\hbar|\omega_S - \omega_A|}{k_B T}\right) I\left(\frac{E_R}{k_B T}, \frac{\hbar|\omega_S - \omega_A|}{E_R}\right) \\ &= \Gamma I\left(\frac{E_R}{k_B T}, -\frac{\hbar|\omega_S - \omega_A|}{E_R}\right), \end{aligned} \quad (\text{C.2.30})$$

where we used Observation (1) again.

C.2.7. Solving the populations of the harmonic oscillator

Here, we want to proof the claim that the system of differential equations,

$$\frac{d}{d\tau} p_n(\tau) = nq p_{n-1}(\tau) + (n+1)(1-q) p_{n+1}(\tau) - [n+q] p_n(\tau), \quad n \geq 0 \quad (\text{C.2.31})$$

with initial condition $p_n(0) = \delta_{n,0}$ is solved by,

$$p_n(\tau) = e^{-\tau} (1 - e^{-\tau})^n, \quad n \geq 0. \quad (\text{C.2.32})$$

In order to show this, we first rewrite $p_n(\tau) = e^{-[n+1]\tau} (e^\tau - 1)^n$ and then find with the product rule,

$$\begin{aligned} \frac{d}{d\tau} p_n(\tau) &= -[n+1] p_n(\tau) + e^{-[n+1]\tau} \frac{d}{d\tau} (e^\tau - 1)^n \\ &= -[n+1] p_n(\tau) + n e^{-n\tau} (e^\tau - 1)^{n-1} \\ &= -[n+1] p_n(\tau) + n p_{n-1}(\tau), \end{aligned} \quad (\text{C.2.33})$$

which is what we wanted to show.

D. Appendix for Metrology

D.1. Model

D.1.1. Commutator of momentum and vector potential

Here, we will show that the momentum operator $\hat{\mathbf{p}}$ and the vector potential, defined in (4.2.2), fulfill,

$$\hat{\mathbf{p}} \cdot \hat{\mathbf{A}}(\hat{\mathbf{x}}) = \hat{\mathbf{A}}(\hat{\mathbf{x}}) \cdot \hat{\mathbf{p}}. \quad (\text{D.1.1})$$

This is best shown in index notation, where $\hat{\mathbf{A}}$ reads,

$$\hat{A}_i(\hat{\mathbf{x}}) = \frac{\mu_0}{4\pi} \frac{1}{|\hat{\mathbf{x}}|^3} \sum_{j,k} \epsilon_{ijk} \mu_j \hat{x}_k. \quad (\text{D.1.2})$$

We find

$$\begin{aligned} \hat{\mathbf{A}}(\hat{\mathbf{x}}) \cdot \hat{\mathbf{p}} - \hat{\mathbf{p}} \cdot \hat{\mathbf{A}}(\hat{\mathbf{x}}) &= \sum_i \left[\hat{A}_i(\hat{\mathbf{x}}), \hat{p}_i \right] = \frac{\mu_0}{4\pi} \sum_{i,j,k} \epsilon_{ijk} \mu_j \left[\frac{\hat{x}_k}{|\hat{\mathbf{x}}|^3}, \hat{p}_i \right] \\ &= i\hbar \frac{\mu_0}{4\pi} \sum_{i,j,k} \epsilon_{ijk} \mu_j \frac{\partial}{\partial \hat{x}_i} \left(\frac{\hat{x}_k}{|\hat{\mathbf{x}}|^3} \right) \\ &= i\hbar \frac{\mu_0}{4\pi} \sum_{j,i,k} \mu_j \epsilon_{jik} \frac{\partial}{\partial \hat{x}_i} \left(\frac{\partial}{\partial \hat{x}_k} \frac{1}{|\hat{\mathbf{x}}|} \right) \\ &= i\hbar \frac{\mu_0}{4\pi} \boldsymbol{\mu} \cdot \nabla \times \left(\nabla \frac{1}{|\hat{\mathbf{x}}|} \right) = 0, \end{aligned} \quad (\text{D.1.3})$$

where we used the commutator between functions of $\hat{\mathbf{x}}$ and the momentum operator (1.2.4), the fact that $\mathbf{x}/|\mathbf{x}|^3 = -\nabla (1/|\mathbf{x}|)$, $\epsilon_{ijk} = -\epsilon_{jik}$, and that the curl of gradient fields is zero.

The vanishing of the commutator holds true in a more general context: Consider a vector potential defined by a radial vector field $\mathbf{F}(\mathbf{x})$,

$$\hat{\mathbf{A}}(\hat{\mathbf{x}}) = \boldsymbol{\mu} \times \mathbf{F}(\hat{\mathbf{x}}). \quad (\text{D.1.4})$$

Then, we find

$$\hat{\mathbf{A}}(\hat{\mathbf{x}}) \cdot \hat{\mathbf{p}} - \hat{\mathbf{p}} \cdot \hat{\mathbf{A}}(\hat{\mathbf{x}}) \propto \boldsymbol{\mu} \cdot (\nabla \times \mathbf{F}(\hat{\mathbf{x}})) = 0, \quad (\text{D.1.5})$$

because radial vector fields have vanishing curl. The generalized formula will become important when we consider the regularized vector potential.

D.1.2. Regularized vector potential

In this part of the Appendix, we derive an integral for the regularized vector potential. The integral reads,

$$\mathbf{A}_G(\mathbf{x}) = \int d^3\mathbf{y} P_G(\mathbf{y}) \mathbf{A}(\mathbf{x} - \mathbf{y}) = \frac{\mu_0}{4\pi} \boldsymbol{\mu} \times \underbrace{\int d^3\mathbf{y} P_G(\mathbf{y}) \frac{\mathbf{x} - \mathbf{y}}{|\mathbf{x} - \mathbf{y}|^3}}_{=\mathbf{I}(\mathbf{x})}. \quad (\text{D.1.6})$$

The remaining integral can be simplified by substituting $\mathbf{z}(\mathbf{y}) = \mathbf{y} - \mathbf{x}$ and by using $\nabla|\mathbf{z}|^{-1} = -\mathbf{z}|\mathbf{z}|^{-3}$ in combination with integration by parts,

$$\begin{aligned} \mathbf{I}(\mathbf{x}) &= \int d^3\mathbf{y} P_G(\mathbf{y}) \frac{\mathbf{x} - \mathbf{y}}{|\mathbf{x} - \mathbf{y}|^3} = - \int d^3\mathbf{z} P_G(\mathbf{z} + \mathbf{x}) \frac{\mathbf{z}}{|\mathbf{z}|^3} \\ &= \int d^3\mathbf{z} P_G(\mathbf{z} + \mathbf{x}) \nabla_{\mathbf{z}} \frac{1}{|\mathbf{z}|} = - \int \frac{d^3\mathbf{z}}{|\mathbf{z}|} \nabla_{\mathbf{z}} P_G(\mathbf{z} + \mathbf{x}) = \int \frac{d^3\mathbf{z}}{\Delta^2} \left(\frac{\mathbf{z}}{|\mathbf{z}|} + \frac{\mathbf{x}}{|\mathbf{z}|} \right) P_G(\mathbf{z} + \mathbf{x}). \end{aligned} \quad (\text{D.1.7})$$

Using spherical coordinates $\mathbf{z}(r, \vartheta, \varphi)$, where the polar axis is chosen parallel to $\mathbf{x}$, we find the relation $\mathbf{z} \cdot \mathbf{x} = r|\mathbf{x}| \cos(\vartheta)$ and,

$$\int_0^{2\pi} d\varphi \mathbf{z}(r, \vartheta, \varphi) = 2\pi r \cos(\vartheta) \frac{\mathbf{x}}{|\mathbf{x}|}. \quad (\text{D.1.8})$$

The expansion of the exponential together with the φ integration yields,

$$\begin{aligned} \mathbf{I}(\mathbf{x}) &= \frac{1}{\sqrt{2\pi}\Delta^5} e^{-\frac{1}{2}\left(\frac{\mathbf{x}}{\Delta}\right)^2} \mathbf{x} \int_0^\infty dr r e^{-\frac{1}{2}\left(\frac{r}{\Delta}\right)^2} \int_{-1}^{+1} d\cos(\vartheta) \left(1 + \frac{r \cos(\vartheta)}{|\mathbf{x}|}\right) e^{-\frac{|\mathbf{x}|r \cos(\vartheta)}{\Delta^2}} \\ &= \frac{1}{\sqrt{2\pi}\Delta^5} e^{-\frac{1}{2}\left(\frac{\mathbf{x}}{\Delta}\right)^2} \mathbf{x} \left(1 - \frac{\Delta^2}{|\mathbf{x}|} \frac{\partial}{\partial|\mathbf{x}|}\right) \int_0^\infty dr r e^{-\frac{1}{2}\left(\frac{r}{\Delta}\right)^2} \int_{-1}^{+1} d\cos(\vartheta) e^{-\frac{|\mathbf{x}|r \cos(\vartheta)}{\Delta^2}} \\ &= \frac{2}{\sqrt{2\pi}\Delta^4} e^{-\frac{1}{2}\left(\frac{\mathbf{x}}{\Delta}\right)^2} \mathbf{x} \left(1 - \frac{\Delta^2}{|\mathbf{x}|} \frac{\partial}{\partial|\mathbf{x}|}\right) \frac{\Delta}{|\mathbf{x}|} \int_0^\infty dr e^{-\frac{1}{2}\left(\frac{r}{\Delta}\right)^2} \sinh\left(\frac{r|\mathbf{x}|}{\Delta^2}\right) \\ &= \frac{1}{\Delta^3} e^{-\frac{1}{2}\left(\frac{\mathbf{x}}{\Delta}\right)^2} \mathbf{x} \left(1 - \frac{\Delta^2}{|\mathbf{x}|} \frac{\partial}{\partial|\mathbf{x}|}\right) \frac{\Delta}{|\mathbf{x}|} e^{\frac{1}{2}\left(\frac{\mathbf{x}}{\Delta}\right)^2} \operatorname{erf}\left(\frac{|\mathbf{x}|}{\sqrt{2}\Delta}\right) \\ &= \frac{\mathbf{x}}{|\mathbf{x}|^3} \left(\operatorname{erf}\left(\frac{|\mathbf{x}|}{\sqrt{2}\Delta}\right) - \sqrt{\frac{2}{\pi}} \frac{|\mathbf{x}|}{\Delta} e^{-\frac{1}{2}\frac{|\mathbf{x}|^2}{\Delta^2}} \right). \end{aligned} \quad (\text{D.1.9})$$

In the second line, we used differentiation under the integral,

$$-\frac{\Delta^2}{|\mathbf{x}|} \frac{\partial}{\partial|\mathbf{x}|} e^{-|\mathbf{x}|r \cos(\vartheta)/\Delta^2} = \frac{r \cos(\vartheta)}{|\mathbf{x}|} e^{-|\mathbf{x}|r \cos(\vartheta)/\Delta^2}. \quad (\text{D.1.10})$$

Finally, we used the fundamental integral,

$$\int_0^\infty \frac{d\tau}{\sqrt{2\pi}} e^{-\tau^2/2} \sinh(b\tau) = \frac{1}{2} e^{b^2/2} \operatorname{erf}(b/\sqrt{2}), \quad (\text{D.1.11})$$

and then calculated the derivative.

D.1.3. Integral for the scattering operator without backaction

In the derivation of the scattering operator without backaction, we encountered an parameter integral,

$$I(b) = \int_{-\infty}^{+\infty} dz \frac{F(\sqrt{b^2 + z^2})}{\sqrt{b^2 + z^2}^3}. \quad (\text{D.1.12})$$

Here, F is the function regularizing our expressions,

$$F(|\mathbf{x}|) = \operatorname{erf}\left(\frac{|\mathbf{x}|}{\sqrt{2}\Delta}\right) - \sqrt{\frac{2}{\pi}} \frac{|\mathbf{x}|}{\Delta} \exp\left(-\frac{1}{2} \left(\frac{|\mathbf{x}|}{\Delta}\right)^2\right). \quad (\text{D.1.13})$$

Before we start the integration, we note the following two derivatives that will be useful in the calculation,

$$\frac{d}{dz} \frac{z}{b^2 \sqrt{b^2 + z^2}} = \frac{1}{\sqrt{b^2 + z^2}^3} \quad (\text{D.1.14})$$

$$\frac{d}{dz} F\left(\sqrt{b^2 + z^2}\right) = \sqrt{\frac{2}{\pi}} \frac{z \sqrt{b^2 + z^2}}{\Delta^3} e^{-\frac{1}{2} \frac{b^2 + z^2}{\Delta^2}}. \quad (\text{D.1.15})$$

We begin by writing the square root cubed as a derivative and apply integration by parts,

$$\begin{aligned} I(b) &= \int_{-\infty}^{+\infty} dz F\left(\sqrt{b^2 + z^2}\right) \frac{d}{dz} \frac{z}{b^2 \sqrt{b^2 + z^2}} \\ &= \frac{1}{b^2 \sqrt{1 + \left(\frac{b}{z}\right)^2}} F\left(\sqrt{b^2 + z^2}\right) \Big|_{-\infty}^{+\infty} - \int_{-\infty}^{+\infty} dz \frac{z}{b^2 \sqrt{b^2 + z^2}} \frac{d}{dz} F\left(\sqrt{b^2 + z^2}\right) \\ &= \frac{2}{b^2} - \frac{1}{b^2} e^{-\frac{b^2}{2\Delta^2}} \sqrt{\frac{2}{\pi}} \int_{-\infty}^{+\infty} dz \frac{z^2}{\Delta^3} e^{-\frac{z^2}{2\Delta^2}} \\ &= \frac{2}{b^2} \left(1 - e^{-\frac{b^2}{2\Delta^2}}\right). \end{aligned} \quad (\text{D.1.16})$$

D.1.4. Integral for the scattering operator with backaction

In preparation for the derivation of the scattering operator with backaction in Subsection 4.2.3, we derive the following integral,

$$\begin{aligned} I &= \int_{-\infty}^{+\infty} v_0 dt \frac{F(|\hat{\mathbf{x}} + v_0 t \mathbf{e}_z|) R(\mathbf{r}, -\omega_0 t)}{|\hat{\mathbf{x}} + v_0 t \mathbf{e}_z|^3} \\ &= \int_{-\infty}^{+\infty} d\tilde{z} |\tilde{z}\rangle \langle \tilde{z}| \int_{-\infty}^{+\infty} v_0 dt \frac{F(|\hat{\mathbf{x}}_{\perp} + (\tilde{z} + v_0 t) \mathbf{e}_z|) R(\mathbf{r}, -\omega_0 t)}{|\hat{\mathbf{x}}_{\perp} + (\tilde{z} + v_0 t) \mathbf{e}_z|^3}, \end{aligned} \quad (\text{D.1.17})$$

where we split the position operator $\hat{\mathbf{x}}$ in a part parallel $z \mathbf{e}_z$ and perpendicular $\hat{\mathbf{x}}_{\perp}$ to the beam axis and decomposed the $\hat{z}$ operator in its spectrum. Applying the substitution $z = \tilde{z} + v_0 t$ and factorizing the rotation matrix yields,

$$\begin{aligned} I &= \int_{-\infty}^{+\infty} d\tilde{z} |\tilde{z}\rangle \langle \tilde{z}| \int_{-\infty}^{+\infty} dz \frac{F(|\hat{\mathbf{x}}_{\perp} + z \mathbf{e}_z|) R\left(\mathbf{r}, \frac{\omega_0}{v_0}(\tilde{z} - z)\right)}{|\hat{\mathbf{x}}_{\perp} + z \mathbf{e}_z|^3} \\ &= \int_{-\infty}^{+\infty} dz \frac{F(|\hat{\mathbf{x}}_{\perp} + z \mathbf{e}_z|) R\left(\mathbf{r}, \frac{\omega_0}{v_0}(\hat{z} - z)\right)}{|\hat{\mathbf{x}}_{\perp} + z \mathbf{e}_z|^3} \\ &= R\left(\mathbf{r}, \frac{\omega_0}{v_0} \hat{z}\right) \int_{-\infty}^{+\infty} dz \frac{F(|\hat{\mathbf{x}}_{\perp} + z \mathbf{e}_z|) R\left(\mathbf{r}, \frac{\omega_0}{v_0}(-z)\right)}{|\hat{\mathbf{x}}_{\perp} + z \mathbf{e}_z|^3}. \end{aligned} \quad (\text{D.1.18})$$

D.1.5. Expanding the exponential in the backaction case

In this part of the appendix, we expand the exponential of the scattering operator, we encountered in Subsection 4.2.3. As already mentioned in the main text, the square of the angular part yields,

$$\left[e^{-i\hat{\varphi}} \hat{\sigma}_+ - e^{i\hat{\varphi}} \hat{\sigma}_- \right]^2 = e^{-2i} \underbrace{\hat{\sigma}_+^2}_{=0} - \underbrace{\{\hat{\sigma}_+, \hat{\sigma}_-\}}_{=\mathbb{1}} + e^{+2i} \underbrace{\hat{\sigma}_-^2}_{=0} = -\mathbb{1}. \quad (\text{D.1.19})$$

By induction, this allows to express any power,

$$\begin{aligned} \left[e^{-i\hat{\varphi}} \hat{\sigma}_+ - e^{i\hat{\varphi}} \hat{\sigma}_- \right]^{2n} &= (-1)^n \mathbb{1} \\ \left[e^{-i\hat{\varphi}} \hat{\sigma}_+ - e^{i\hat{\varphi}} \hat{\sigma}_- \right]^{2n+1} &= (-1)^n \left[e^{-i\hat{\varphi}} \hat{\sigma}_+ - e^{i\hat{\varphi}} \hat{\sigma}_- \right]. \end{aligned} \quad (\text{D.1.20})$$

Plugging the general powers into the power series of the exponential function yields

the desired result,

$$\begin{aligned}
\hat{S}_\vartheta &= \sum_{n=0}^{\infty} \frac{[\vartheta g(\hat{r})]^n}{n!} [e^{-i\hat{\varphi}} \hat{\sigma}_+ - e^{i\hat{\varphi}} \hat{\sigma}_-]^n \\
&= \sum_{n=0}^{\infty} \frac{[\vartheta g(\hat{r})]^{2n}}{(2n)!} [e^{-i\hat{\varphi}} \hat{\sigma}_+ - e^{i\hat{\varphi}} \hat{\sigma}_-]^{2n} + \sum_{n=0}^{\infty} \frac{[\vartheta g(\hat{r})]^{2n+1}}{(2n+1)!} [e^{-i\hat{\varphi}} \hat{\sigma}_+ - e^{i\hat{\varphi}} \hat{\sigma}_-]^{2n+1} \\
&= \sum_{n=0}^{\infty} (-1)^n \frac{[\vartheta g(\hat{r})]^{2n}}{(2n)!} \mathbb{1} + \sum_{n=0}^{\infty} (-1)^n \frac{[\vartheta g(\hat{r})]^{2n+1}}{(2n+1)!} [e^{-i\hat{\varphi}} \hat{\sigma}_+ - e^{i\hat{\varphi}} \hat{\sigma}_-] \\
&= \cos(\vartheta g(\hat{r})) \mathbb{1} + \sin(\vartheta g(\hat{r})) [e^{-i\hat{\varphi}} \hat{\sigma}_+ - e^{i\hat{\varphi}} \hat{\sigma}_-].
\end{aligned} \tag{D.1.21}$$

D.2. Sensing without backaction

D.2.1. QFI of a centered Gaussian

We solve an integral encountered in Subsection 4.3.1,

$$\frac{|\mathbf{n}_\perp|^2}{2\chi^2} \int_0^\infty \frac{d\tau}{\tau} e^{-(\tau/\chi)^2/2} \left(1 - e^{-\tau^2/2}\right)^2. \tag{D.2.1}$$

Note that the convergence of the integral at $\tau = 0$ is ensured by the fact the zeroth order term in the exponential is canceled,

$$1 - e^{-\tau^2/2} = \frac{\tau^2}{2} - \frac{\tau^4}{8} + \dots = \tau^2 \left(\frac{1}{2} - \frac{\tau^2}{8} + \dots \right). \tag{D.2.2}$$

Hence, we can not simply integrate term by term. Fortunately, we can use the following trick to simplify the integral,

$$\left(1 - e^{-\tau^2/2}\right)^2 = 1 - 2e^{-\tau^2/2} + e^{-\tau^2} = 2 \left(1 - e^{-\tau^2/2}\right) - \left(1 - e^{-\tau^2}\right), \tag{D.2.3}$$

where we added and subtracted a 1 to obtain two terms that converge under the integral. With the definition,

$$I(\chi) = \int_0^\infty \frac{dt}{t} e^{-(t/\chi)^2/2} \left(1 - e^{-t^2/2}\right), \tag{D.2.4}$$

and a substitution, we see

$$\mathcal{J}(\chi) = \frac{2|\mathbf{n}_\perp|^2}{\chi^2} \left(2I(\chi) - I(\sqrt{2}\chi) \right). \tag{D.2.5}$$

It remains to find a solution to $I(\chi)$, which can be done with the formula,

$$\int_0^\infty \frac{dx}{x} (e^{-\nu x^p} - e^{-\mu x^p}) = \frac{1}{p} \ln \left(\frac{\mu}{\nu} \right), \quad \mu, \nu > 0, \tag{D.2.6}$$

found in [183]. In our case, this yields,

$$I(\chi) = \frac{1}{2} \ln(1 + \chi^2) \quad (\text{D.2.7})$$

and hence,

$$\mathcal{J} = \frac{2|\mathbf{n}_\perp|^2}{\chi^2} \ln \left(\frac{1 + \chi^2}{\sqrt{1 + 2\chi^2}} \right). \quad (\text{D.2.8})$$

D.2.2. Angular momentum basis

In Subsection 4.2.4 of the main text, we introduced the basis of (semi-improper) eigenstates $(|r, k\rangle)$. Here, we will present the orthogonality and completeness of this basis. We do this by considering the cartesian components of an arbitrary state $\psi(\mathbf{x}_\perp) = \langle \mathbf{x}_\perp | \psi \rangle$. Clearly, these components can also be expressed as functions of the polar coordinates, $\psi(\mathbf{x}_\perp) = \tilde{\psi}(r, \varphi)$. Employing the Fourier transform in the angle φ for fixed radius r , yields an alternative representation of the wave function,

$$\psi(\mathbf{x}_\perp) = \tilde{\psi}(r, \varphi) = \sum_{k \in \mathbb{Z}} A_k(r) \frac{1}{\sqrt{2\pi}} e^{ik\varphi}. \quad (\text{D.2.9})$$

By demanding $\langle r, k | \psi \rangle = A_k(r)$, we find the position space representation of $|r, k\rangle$,

$$\langle \mathbf{x}_\perp | r, k \rangle = \frac{1}{r} \delta^{(1)}(|\mathbf{x}_\perp| - r) \frac{1}{\sqrt{2\pi}} e^{ik\varphi_x}. \quad (\text{D.2.10})$$

For the completeness, we note

$$\begin{aligned} \langle \mathbf{x}_\perp | \left(\int_0^\infty dr \, r \sum_{k \in \mathbb{Z}} |r, k\rangle \langle r, k| \right) | \psi \rangle &= \int_0^\infty dr \, r \sum_{k \in \mathbb{Z}} A_k(r) \langle \mathbf{x}_\perp | r, k \rangle \\ &= \sum_{k \in \mathbb{Z}} A_k(|\mathbf{x}|) \frac{1}{\sqrt{2\pi}} e^{ik\varphi} \\ &= \psi(\mathbf{x}_\perp) = \langle \mathbf{x}_\perp | \psi \rangle, \quad \forall \mathbf{x}_\perp, |\psi\rangle. \end{aligned} \quad (\text{D.2.11})$$

Hence, we find,

$$\int_0^\infty dr \, r \sum_{k \in \mathbb{Z}} |r, k\rangle \langle r, k| = \mathbb{1}. \quad (\text{D.2.12})$$

For the orthogonality, we use the completeness of the cartesian basis,

$$\begin{aligned}
 \langle r', k' | r, k \rangle &= \int d^2 \mathbf{x}_\perp \langle r', k' | \mathbf{x}_\perp \rangle \langle \mathbf{x}_\perp | r, k \rangle \\
 &= \int_0^\infty dr_x \, r_x \frac{1}{r'} \delta^{(1)}(r' - r_x) \frac{1}{r} \delta^{(1)}(r - r_x) \int_0^{2\pi} \frac{d\varphi_x}{2\pi} e^{i(k-k')\varphi_x} \\
 &= \frac{1}{r} \delta^{(1)}(r' - r) \delta_{k'k}.
 \end{aligned} \tag{D.2.13}$$

D.3. Sensing with backaction

D.3.1. Derivation of the scattering channel

In Subsection 4.4.2, we encounter the scattering channel,

$$\Lambda_{\mathbf{c}, \vartheta} = \text{tr}_S \left\{ \hat{S}_\vartheta (\hat{\rho}_M \otimes \hat{\rho}_S) \hat{S}_\vartheta^\dagger \right\}. \tag{D.3.1}$$

Here, we will take the trace for a general spin state, which can be characterized by the Bloch vector $\mathbf{c}$,

$$\hat{\rho}_S = \frac{1}{2}(1 + c_z) |\uparrow\rangle \langle \uparrow| + \frac{1}{2}(c_x - ic_y) |\uparrow\rangle \langle \downarrow| + \frac{1}{2}(c_x + ic_y) |\downarrow\rangle \langle \uparrow| + \frac{1}{2}(1 - c_z) |\downarrow\rangle \langle \downarrow|. \tag{D.3.2}$$

Due to the linearity of the trace, we can split $\hat{S}_\vartheta (\hat{\rho}_M \otimes \hat{\rho}_S) \hat{S}_\vartheta^\dagger$ into four terms,

$$\begin{aligned}
 \hat{\rho}_M \otimes |\uparrow\rangle \langle \uparrow| &\mapsto \hat{K}_0 \hat{\rho}_M \hat{K}_0^\dagger \otimes |\uparrow\rangle \langle \uparrow| + \hat{K}_+ \hat{\rho}_M \hat{K}_+^\dagger \otimes |\downarrow\rangle \langle \downarrow| \\
 \hat{\rho}_M \otimes |\downarrow\rangle \langle \downarrow| &\mapsto \hat{K}_0 \hat{\rho}_M \hat{K}_0^\dagger \otimes |\downarrow\rangle \langle \downarrow| + \hat{K}_- \hat{\rho}_M \hat{K}_-^\dagger \otimes |\uparrow\rangle \langle \uparrow| \\
 \hat{\rho}_M \otimes |\uparrow\rangle \langle \downarrow| &\mapsto \hat{K}_0 \hat{\rho}_M \hat{K}_-^\dagger \otimes |\uparrow\rangle \langle \uparrow| - \hat{K}_+ \hat{\rho}_M \hat{K}_0^\dagger \otimes |\downarrow\rangle \langle \downarrow| \\
 \hat{\rho}_M \otimes |\downarrow\rangle \langle \uparrow| &\mapsto \hat{K}_- \hat{\rho}_M \hat{K}_0^\dagger \otimes |\uparrow\rangle \langle \uparrow| - \hat{K}_0 \hat{\rho}_M \hat{K}_+^\dagger \otimes |\downarrow\rangle \langle \downarrow|,
 \end{aligned} \tag{D.3.3}$$

where we already left out terms with non-zero trace on the right hand side. Taking the

partial trace and combining the results with the entries of the state $\hat{\rho}_S$ yields,

$$\begin{aligned}
 \Lambda_{\mathbf{c},\vartheta} [\hat{\rho}_M] & \quad (D.3.4) \\
 &= \frac{1+c_z}{2} \left(\hat{K}_0 \hat{\rho}_M \hat{K}_0^\dagger + \hat{K}_+ \hat{\rho}_M \hat{K}_+^\dagger \right) + \frac{1-c_z}{2} \left(\hat{K}_0 \hat{\rho}_M \hat{K}_0^\dagger + \hat{K}_- \hat{\rho}_M \hat{K}_-^\dagger \right) \\
 & \quad + \frac{c_x - i c_y}{2} \left(\hat{K}_0 \hat{\rho}_M \hat{K}_-^\dagger - \hat{K}_+ \hat{\rho}_M \hat{K}_0^\dagger \right) + \frac{c_x + i c_y}{2} \left(\hat{K}_- \hat{\rho}_M \hat{K}_0^\dagger - \hat{K}_0 \hat{\rho}_M \hat{K}_+^\dagger \right) \\
 &= \hat{K}_0 \hat{\rho}_M \hat{K}_0^\dagger + \frac{1}{2} \left(\hat{K}_+ \hat{\rho}_M \hat{K}_+^\dagger + \hat{K}_- \hat{\rho}_M \hat{K}_-^\dagger \right) + \frac{c_z}{2} \left(\hat{K}_+ \hat{\rho}_M \hat{K}_+^\dagger - \hat{K}_- \hat{\rho}_M \hat{K}_-^\dagger \right) \\
 & \quad + \frac{c_x}{2} \left(\hat{K}_0 \hat{\rho}_M \left[\hat{K}_- - \hat{K}_+ \right]^\dagger + \left[\hat{K}_- - \hat{K}_+ \right] \hat{\rho}_M \hat{K}_0^\dagger \right) \\
 & \quad + \frac{i c_y}{2} \left(\left[\hat{K}_+ + \hat{K}_- \right] \hat{\rho}_M \hat{K}_0^\dagger - \hat{K}_0 \hat{\rho}_M \left[\hat{K}_+ + \hat{K}_- \right]^\dagger \right).
 \end{aligned}$$

Introducing a complex number that characterizes the coherences $c_x + i c_y = |\mathbf{c}_\perp| e^{i\varphi_c}$, the second two lines can be rephrased as,

$$\begin{aligned}
 & \frac{c_x}{2} \left(\hat{K}_0 \hat{\rho}_M \left[\hat{K}_- - \hat{K}_+ \right]^\dagger + \left[\hat{K}_- - \hat{K}_+ \right] \hat{\rho}_M \hat{K}_0^\dagger \right) \\
 & + \frac{i c_y}{2} \left(\left[\hat{K}_+ + \hat{K}_- \right] \hat{\rho}_M \hat{K}_0^\dagger - \hat{K}_0 \hat{\rho}_M \left[\hat{K}_+ + \hat{K}_- \right]^\dagger \right) \\
 &= +i |\mathbf{c}_\perp| \hat{K}_0 \hat{\rho}_M \sin(\vartheta g(\hat{r})) [\sin(\hat{\varphi}) \cos(\varphi_c) - \cos(\hat{\varphi}) \sin(\varphi_c)] \\
 & \quad - i |\mathbf{c}_\perp| \sin(\vartheta g(\hat{r})) [\sin(\hat{\varphi}) \cos(\varphi_c) - \cos(\hat{\varphi}) \sin(\varphi_c)] \\
 &= i |\mathbf{c}_\perp| \left(\hat{K}_0 \hat{\rho}_M \sin(\vartheta g(\hat{r})) \sin(\hat{\varphi} - \varphi_c) - \sin(\vartheta g(\hat{r})) \sin(\hat{\varphi} - \varphi_c) \hat{\rho}_M \hat{K}_0^\dagger \right) \quad (D.3.5)
 \end{aligned}$$

All things considered, we find the channel,

$$\begin{aligned}
 \Lambda_{\mathbf{c},\vartheta} [\hat{\rho}_M] &= \hat{K}_0 \hat{\rho}_M \hat{K}_0^\dagger + \frac{1}{2} \left(\hat{K}_+ \hat{\rho}_M \hat{K}_+^\dagger + \hat{K}_- \hat{\rho}_M \hat{K}_-^\dagger \right) + \frac{c_z}{2} \left(\hat{K}_+ \hat{\rho}_M \hat{K}_+^\dagger - \hat{K}_- \hat{\rho}_M \hat{K}_-^\dagger \right) \\
 & \quad + i |\mathbf{c}_\perp| \left(\hat{K}_0 \hat{\rho}_M \sin(\vartheta g(\hat{r})) \sin(\hat{\varphi} - \varphi_c) - \sin(\vartheta g(\hat{r})) \sin(\hat{\varphi} - \varphi_c) \hat{\rho}_M \hat{K}_0^\dagger \right). \quad (D.3.6)
 \end{aligned}$$

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List of publications

The results discussed in this thesis are based on the following published works:

- A Michael Gaida and Stefan Nimmrichter. Otto cycles with a quantum planar rotor. *Phys. Rev. E*, 110(3):034109, 2024
- B Michael Gaida, Giulio Gasbarri and Stefan Nimmrichter. Thermodynamically consistent collisional master equation in a low-density gas with internal structure. *arXiv:2506.21394v1*
- C Santiago Steven Beltrán Romero, Michael Gaida, Philipp Haslinger, Stefan Nimmrichter and Dennis Rätzel. Metrology for magnetic moments in electron microscopes. [In preparation]

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