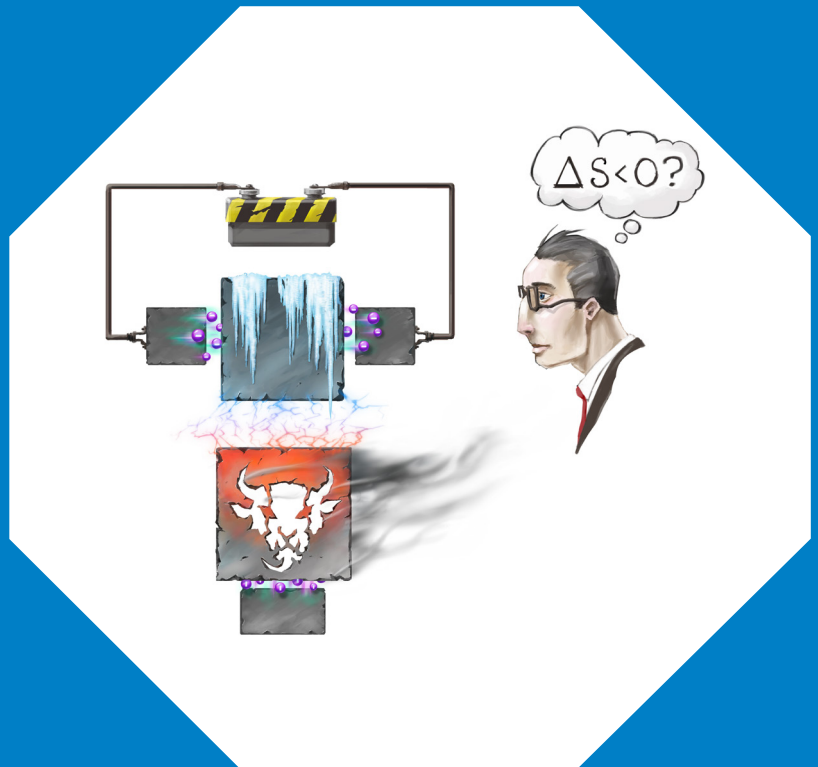


# Non-Markovian environments and information exchange in stochastic thermodynamics

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Aki Kutvonen



# Non-Markovian environments and information exchange in stochastic thermodynamics

**Aki Kutvonen**

A doctoral dissertation completed for the degree of Doctor of Science (Technology) to be defended, with the permission of the Aalto University School of Science, at a public examination held at the lecture hall M1 (Otakaari 1, Espoo) of the school on 5th of February 2016 at noon.

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The current challenge in the field of thermodynamics is to extend the fundamental laws of thermodynamics to small non-equilibrium systems. The framework of stochastic thermodynamics has proven useful in studying small systems, which often are fluctuating and easily driven out of equilibrium. In stochastic thermodynamics the fundamental laws are formulated using fluctuating and trajectory dependent variables such as entropy, heat and work evolving under stochastic equations of motion. More recently, the possibility to manipulate small systems by performing measurements and feedbacks has also been included in the thermodynamic framework.

In this thesis, stochastic thermodynamics and thermodynamics of information are studied using physically feasible model systems based on single electron tunneling at low temperatures. We study dissipation, entropy production, and thermodynamics of information in these setups analytically and by using Monte Carlo simulations and numerically solving master equations. We develop a model of non-Markovian dynamics, in which the system and the environmental degrees of freedom are correlated during the process. This model explains the sources of additional entropy production, not included in the standard stochastic thermodynamics description, and it can be realized in the operation of a voltage driven single electron box. Furthermore, we introduce two different model setups, where dissipation can be made negative, and thus the setups operate as Maxwell's demons. In both setups, the role of information is quantitatively characterized.

The research presented in the thesis develops the field of thermodynamics at small scales into a more quantitative and accurate direction, where interaction energies and non-equilibrium excitations in the environment are taken into account. Furthermore, the results provide a step towards more precise modeling, understanding, and design of small scale devices, where dissipation and fluctuations play a major role.

**Keywords** Stochastic thermodynamics, information thermodynamics, small systems, fluctuations

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**Tekijä**

Aki Kutvonen

**Väitöskirjan nimi**

Ei-markoviset ympäristöt ja informaatio stokastisessa termodynamiikassa

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Stokastinen termodynamiikka on perinteisen termodynamiikan laajennus, joka selittää ja mallintaa pienien epätasapainossa olevien systeemien fysiikkaa. Stokastisen termodynamiikan teoria pohjautuu yksittäisten realisaatioiden avulla määritettyihin termodynamiisiin suureisiin, kuten entropia, työ ja lämpö. Informaation termodynamiikka tutkii näiden pienien systeemien dissipaatiota kun systeemejä manipuloidaan käyttämällä tietoa systeemin mikroskooppisesta tilasta.

Tässä väitöskirjassa tutkitaan stokastista termodynamiikkaa ja informaation termodynamiikkaa käyttäen apuna matalien lämpötilojen elektronisia systeemejä. Tutkimus on tehty monte carlo -simulaatiomenetelmillä, sekä liikeyhtälöiden numeerisella ja analyyttisellä ratkaisemisella.

Työssä kehitetään malli, joka selittää ei-markovisen systeemin dynamiikkaa ja dissipaatiota prosessissa, jossa systeemi ja sen ympäristö ovat korreloituneita. Tällä mallilla voidaan selittää ylimääräinen entropiantuotanto, jota normaali stokastinen termodynamiikka ei pysty selittämään, ja joka voitaisiin havaita niin sanotun yksielektronilaatikon toiminnassa. Työssä esitetään myös kaksi fysikaalista mallisysteemiä, jossa dissipaatio voidaan tehdä negatiiviseksi. Nämä systeemit ovat esimerkkejä Maxwellin demoneista, joiden toiminta voidaan selittää informaation termodynamiikan avulla.

Tämän väitöskirjan tulokset tarkentavat stokastisen termodynamiikan ja informaation termodynamiikan teoriaa suuntaan, jossa vuorovaikutusenergiat ovat otettu huomioon entistä tarkemmin. Lisäksi tulokset kehittävät teoreettisia metodeja entistä hyödyllisemmiksi pienen mittakaavan laitteiden suunnittelussa ja mallintamisessa.

**Avainsanat** Stokastinen termodynamiikka, informaation termodynamiikka, pienet systeemit, fluktuaatiot

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# Preface

The work presented in this thesis was carried out in the Multiscale Statistical Physics group (MSP) at the Department of Applied Physics, Aalto University School of Science during the years 2011-2015.

During my years at Aalto University I have been supported by many great people who I would like to thank. First of all, I would like to thank my PhD and M. Sc. works supervisor Prof. Tapio Ala-Nissila. I started to work with Tapsa in 2009 and under his supervision I have learned the things I know about academic world. I have always felt encouraged, supported and I have been given a change to choose my research topic to my liking. I'm grateful for that.

I want to thank the present and the previous members of the MSP group. It has been a pleasure to share thoughts with you, both related to science and life in general. Many thanks to you all, especially to Dr. Timo Ikonen, Dr. Paolo Solinas, M. Sc. Vili Heinonen, M. Sc. Harri Mökkönen and M. Sc. Samu Suomela. Thanks also to Dr. Emmi Ruokokoski and M. Sc. Topi Siro for listening my whining about work and providing me pleasant lunch company over the years at Otaniemi. Special thanks to Dr. Giulia Rossi for patience in teaching me scientific writing and many other things about science in my early years as a masters student in MSP group.

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Last, I want to thank my family and friends for support and encouragement. Making a PhD thesis is a lot of fun, but sometimes it's good to have some other things in life as well.

January 11, 2016,

Aki Kutvonen

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# List of Publications

This thesis consists of an overview and of the following publications which are referred to in the text by their Roman numerals.

**I** J. P. Pekola, A. Kutvonen, and T. Ala-Nissila. Dissipated work and fluctuation relations for non-equilibrium single-electron transitions. *Journal of Statistical Mechanics: Theory and Experiment*, P02033 (5 Pages), February 2013.

**II** A. Kutvonen, T. Ala-Nissila, and J. P. Pekola. Entropy production in a non-Markovian environment. *Physical Review E*, **92**, 012107 (7 pages), July 2015.

**III** A. Kutvonen, J. V. Koski, and T. Ala-Nissila. Thermodynamics and efficiency of an autonomous on-chip Maxwell's demon. Accepted for publication in *Scientific Reports*, October 2015.

**IV** A. Kutvonen, T. Sagawa, and T. Ala-Nissila. Thermodynamics of information exchange between two coupled quantum dots. Submitted to *Physical Review E*, December 2015.



# Author's Contribution

## **Publication I: “Dissipated work and fluctuation relations for non-equilibrium single-electron transitions”**

The author performed the numerical simulations, derived part of the analytical results, wrote the parts of the manuscript concerning the simulations and took actively part in writing and revising the manuscript.

## **Publication II: “Entropy production in a non-Markovian environment”**

The author proposed the model, carried out the analysis and calculations, and wrote the manuscript.

## **Publication III: “Thermodynamics and efficiency of an autonomous on-chip Maxwell’s demon”**

The author carried out the calculations and numerical results including the analysis of the results, and mainly wrote the manuscript.

## **Publication IV: “Thermodynamics of information exchange between two coupled quantum dots”**

The author developed the model with the co-authors and prof. Jukka Pekola, carried out the analysis and calculations, and wrote the manuscript.

During the work described in this thesis, the author has also contributed to additional related articles [1,2], not included in the thesis.



# 1. Introduction

"It is the only physical theory of universal content which I am convinced will never be overthrown, within the framework of applicability of its basic concepts." - *Albert Einstein*. Einstein correctly predicted that the laws of thermodynamics have kept their place among the most fundamental laws in physics. The generality of the concepts in thermodynamics makes it particularly useful, explaining a variety of phenomena, from basic heat machines to abstract quantum systems. As modern nanotechnology has developed, small devices and fundamental science at small scales have become increasingly accessible and important from the perspective of technology and fundamental science. The current challenge in the field is to extend the laws of thermodynamics to these small systems, which are often fluctuating and easily driven outside of equilibrium.

The standard theory of thermodynamics is a theory of averages over large, well defined ensembles containing large degrees of freedom [3]. Outside equilibrium, and when the number of degrees of freedom in the problem is small, no universal theory of applicability comparable to the thermodynamics is known. Motivated by recent developments in nanotechnology, a great effort has been made to extend thermodynamics to microscopically small systems and to non-equilibrium processes [4–9]. The common fluctuation relations, formulated, for example, in Refs. [10–14], introduce remarkable equalities to describe non-equilibrium (irreversible) processes. These relations formulate thermodynamics by using fluctuating variables such as entropy, heat and work, and connect those to equilibrium variables, such as the Helmholtz free energy.

Recently, researchers have struggled to create a unifying framework for different fluctuation theories. In setups where the dynamics are described by stochastic equations of motion, such as Langevin equations or master equations, a framework of stochastic thermodynamics has proven useful



[15–18]. Stochastic thermodynamics resembles the traditional thermodynamics but describes the processes in a single realization framework, where the thermodynamic variables fluctuate and are not described by averages only. This framework is particularly useful in systems in which the average values are not sufficient to characterize the system due to fluctuations. Typically, this happens in small systems where the scales of energies are the order of the thermal fluctuations  $k_B T$ .

In small systems, entropy production may be negative for single realizations. These realizations are sometimes referred to as microscopic violations of the second law of thermodynamics, however they do not exactly break the second law, since the second law holds for the average entropy production only [5]. Recently, a great interest has developed towards creating setups where the entropy production could also be negative on average [9, 19–33]. In these setups part of the system may produce negative entropy, however, by noting the entropy production in the surroundings, the overall entropy production is positive, and thus the violation of the second law is only apparent.

The modern setups where a part of the total system produces negative entropy remind the cases of the famous information-to-energy thought experiments by Maxwell [34] and Szilard [35]. In both of these setups an intelligent being measures the microscopic state of the system and later utilizes this information in a manner apparently contradicting the second law. Recently, various theoretical [19–29], as well as experimental [9, 30–33] realizations of modern information machines, where information is utilized, have been proposed. The modern theoretical framework describing these information machines, formulated by using stochastic thermodynamics and information theory, is called information thermodynamics [27, 36–41]. The main area of interest in information thermodynamics is measurement and feedback, which can be used to control the microscopic degrees of freedom and thus the fluctuating thermodynamic quantities.

Fluctuations and dissipation at small scales are not only scientifically important, but to control and typically to minimize those in the operation of nanoscale devices, is a crucial task in pushing the boundaries of nanotechnology. In addition, a better understanding of measurements and feedback in small systems, and the role of information in the operation of small systems in general, may help to develop new, unconventional small scale devices.

## 1.1 Historical overview on stochastic thermodynamics

The theory of thermodynamics dates back to 19th century, to pre-industrial times, when Kelvin, Carnot and others developed means to better understand the functioning of heat machines [3]. Later in the 19th century and in early 20th century the microscopic foundations for thermodynamics were laid by Maxwell, Boltzmann, Planck, Clausius, Gibbs and various other physicists and mathematicians creating the framework of statistical mechanics. In this framework the thermodynamic quantities were explained from microscopic dynamics and properties of the system using equilibrium ensembles [42].

While statistical mechanics and thermodynamics were, and still are useful, it was clear that these theories are not able to describe majority of the processes occurring in nature. A typical feature in nature is that processes rarely take place in equilibrium conditions. Thus it became relevant to extend the theory to describe non-equilibrium conditions. This was done by using linear response theories, formalized for example in Onsager reciprocal relations [43] and in Green-Kubo formulae [44,45]. However, although these relations are often called non-equilibrium statistical physics, they still rely on the equilibrium ensembles and should be thus considered really as near-equilibrium statistical physics. While equilibrium behavior of various systems share general features, the non-equilibrium behavior is usually system specific, which makes a general theoretical framework hard to develop.

The modern era of thermodynamics at small scales began in the late 1970's when Bochkov and Kuzolev derived an equality describing fluctuating values of work [10, 46]. Later, in 1990's, the fluctuation theorem, Jarzynski equality and the Crooks fluctuation relations were introduced [11, 12, 47–50]. In this framework the thermodynamics of fluctuating thermodynamic variables, defined for single trajectories, were formalized using equalities. In the thermodynamic limit these equalities yield known thermodynamic inequalities. The fluctuation theories were studied in the context of both Hamiltonian and stochastic evolution of motion. In 2000's Sekimoto, Seifert and others extended the thermodynamics of fluctuating variables under stochastic evolution of motion, now commonly referred to as stochastic thermodynamics [14–16, 18, 51].

The major development in the thermodynamic description of small systems in the recent decade has been to include the possibility to manipu-

late the system by performing measurements and feedbacks. This field, developed by Sagawa, Ueda and others, called information thermodynamics, consider correlations between systems and formulate them using concepts such as mutual information [21, 36–39, 52]. As a result, various theoretical as well as experimental realizations of information machines [9, 19–26, 29–33] have been studied.

While stochastic thermodynamics and information thermodynamics have broadened our knowledge on non-equilibrium thermodynamics, especially at small scales, many open questions still remain. Most of the results rely on neglecting of interaction energies between the system and its environment, and on clear separation of timescales. In particular, in small systems it is not always so clear that these assumptions can be made and how they change the descriptions of thermodynamic quantities. Information thermodynamics in general is still a developing field and, for example, it is not always clear how the measurement and feedback should be interpreted in a specific model system.

## 1.2 Objectives of the thesis

The accurate testing of various fluctuation theorems in a well defined interacting system, satisfying requirements of physical feasibility and where large statistics can be obtained, is a cumbersome task. In this thesis we use systems based on single electron tunneling at low temperatures, which satisfy the above requirements. We use Monte Carlo simulations, numerical solving of master equations and analytical methods based on stochastic thermodynamics and information thermodynamics to study stochastic thermodynamics with system-environment and system-system correlations.

The topic of the first part of the thesis is work and entropy fluctuations in a system, where the environment and the system are correlated to each other so that the environment becomes time dependent during the driving of the system. In Publications I and II, to our knowledge for the first time, we take the time dependence in the environment into account in a stochastic model. We show that this results into an additional non-Markovian entropy generation terms, which modifies the work fluctuation relations. In addition, by building a model which is realized in a driven single electron box system, we are able to identify the non-Markovian terms in the setup.

The second part of the thesis is devoted to studying correlations in stochastic thermodynamics by using information thermodynamics. In Publications III and IV we study two different realizations of Maxwell's demon devices. In both the setups the energetics of the total system, consisting of the system of interest and the demon, are accessible. Thus the thermodynamics of the whole system-demon compound can be accurately studied. Publication III is the first experimentally feasible setup where the measurement and the feedback are performed separately and the setup contains both the microscopic demon and the setup itself. Publication IV studies thermodynamics and efficiency of information production and utilization in an autonomous Maxwell's demon setup, where the demon acts independently, measuring and performing feedback without any external control.

### 1.3 Organization of the thesis

The first chapter of the thesis is devoted to a short introduction to stochastic thermodynamics and information thermodynamics including the main topics of the thesis. In Chapter 2, we introduce the basic concepts in stochastic thermodynamics and derive the most common fluctuation theorems. In Chapter 3, we introduce the stochastic thermodynamics of single electron tunneling, used especially in Publications I-III, but also partly relevant for Publication IV. Chapter 4 explains the results of Publications I and II: the fluctuations of work and entropy in a non-Markovian environment. Chapter 5 is devoted to the information thermodynamic description of stochastic thermodynamics including the review of the results of Publications III and IV. Chapter 6 presents the summary and conclusions of the results presented in this thesis including future research directions.



## 2. Stochastic thermodynamics

In this section, we briefly review the basics of stochastic thermodynamics. We derive the thermodynamic quantities for single realizations using stochastic evolution of motion. We also derive the most important fluctuation relations, namely the integral fluctuation relation for entropy production, the Jarzynski equality and the Crooks fluctuation theorem. For those interested in further details, numerous articles on this matter exists, see e.g. [15, 16] and references therein.

### 2.1 Energetics in Markovian dynamics

Let us denote the state of a system at time  $t$  by  $x(t)$ . We assume that the state of the system belongs to a discrete set of states,  $x(t) \in \{1, 2, \dots, k\}$  and has an energy  $E_{x(t)}$ . Furthermore, we assume that the system is manipulated using external time dependent control parameter  $\lambda(t)$ , which could be realized for example by an external time dependent magnetic field. As a result, the energies of the states depends on time due to the time dependence of the external control, i.e. the energy of the system is given by  $E_{x(t)}(t)$ . We assume that the dynamics are stochastic, the process takes place in discrete time splitting  $t_{i+1} = t_i + \Delta t$ , and denote  $\omega_{x_i, x_{i-1}}(t_i)$  as the transition rate from  $x_{i-1} \equiv x(t_{i-1})$  to  $x_i$  at control parameter value  $\lambda_i = \lambda(t_i)$ .

Assume that the system state changes from  $x_{i-1}$  to  $x_i$  at time  $t_i$ . The energy released from the system in this transition is given by  $Q_i = E_{x_{i-1}}(t_i) - E_{x_i}(t_i)$ . Since we are dealing with an interacting system, the energy released is deposited to the environment and thus  $Q_i$  is the heat dissipated to the environment. During a trajectory  $\mathbf{x}$ , from  $x_0 = x(0)$  to  $x_f = x(t_f)$ , consisting of  $n$  transitions, evolving through states  $\mathbf{x} = \{x_0, x_1, x_2, \dots, x_n\}$  and driven by  $\lambda(t)$ , the total dissipated heat is given by  $Q[\mathbf{x}] = \sum_{i=1}^n Q_i$ .

In addition to the transitions driven by the interaction with the environment, the energy of the system changes due to the drive  $\lambda(t)$ . That is to say, even if the state does not change between  $t \in ]t_{i-1}, t_i[$ , the energy of the system changes by  $W_i = E_{x_{i-1}}(t_i) - E_{x_{i-1}}(t_{i-1})$ . Since during this interval there is no energy escape to the environment, the energy change  $W_i$  is the work done to the system. The total energy change during a trajectory  $x$  is then given by

$$\begin{aligned} \Delta E[\mathbf{x}] &= E_{x_f}(t_f) - E_{x_0}(t_0) \\ &= \sum_{i=1}^n \{ [E_{x_i}(t_i) - E_{x_{i-1}}(t_i)] + [E_{x_{i-1}}(t_i) - E_{x_{i-1}}(t_{i-1})] \} = -Q[\mathbf{x}] + W[\mathbf{x}], \end{aligned} \quad (2.1)$$

where  $W$  is the work done during the trajectory. Equation (2.1) is the first law of thermodynamics for trajectory  $\mathbf{x}$ .

## 2.2 Entropy production

We proceed to study the entropy generated in a single realization. By defining the time reverse process by  $\tilde{x}(t) = x(\tilde{t}) = x(t_f - t)$  under control parameter protocol  $\tilde{\lambda}(t) = \lambda(\tilde{t}) = \lambda(t_f - t)$ , the total entropy generation of trajectory  $\mathbf{x}$  is defined as

$$\Delta S_T = \Delta S_S + \Delta S_F = \ln \frac{p_I[x_0]}{p_F[\tilde{x}_0]} + \ln \frac{p[\mathbf{x}|x_0]}{\tilde{p}[\tilde{\mathbf{x}}|\tilde{x}_0]}, \quad (2.2)$$

where  $\Delta S_S$  is the change in the system entropy and  $\Delta S_F$  is the entropy flow to the environment,  $p[\mathbf{x}|x_0]$  and  $\tilde{p}[\tilde{\mathbf{x}}|\tilde{x}_0]$  are the probabilities of the forward and reverse path  $\tilde{\mathbf{x}} = \{\tilde{x}_0, \tilde{x}_1, \tilde{x}_2, \dots, \tilde{x}_n\} = \{x_n, x_{n-1}, x_{n-2}, \dots, x_0\}$ , and  $p_I[x_0]$  and  $p_F[\tilde{x}_0]$  are the probabilities of the initial and final states, respectively [14, 15, 53]. If the dynamics are stochastic and governed by a Markovian master equation, the probability of a path  $\mathbf{x}$  is defined as the product of the probabilities of the path constituents:

$$p[\mathbf{x}|x_0] = e^{-\int_{t_0}^{t_1} \omega_{x_0, x_0}(t') dt'} \prod_{i=1}^n \omega_{x_i, x_{i-1}}(t_i) e^{-\int_{t_i}^{t_{i+1}} \omega_{x_i, x_i}(t') dt'}, \quad (2.3)$$

where  $\omega_{x_i, x_i} = 1 - \sum_{j \neq i} \omega_{x_j, x_i}$  is the diagonal element of the rate matrix  $\omega$ , corresponding to the rate of no transitions and  $t_{n+1} = t_f$ . Thus  $e^{-\int_{t_i}^{t_{i+1}} \omega_{x_i, x_i}(t') dt'}$  corresponds to the probability of not making a transition from state  $x_i$  during the time  $t \in [t_i, t_{i+1}]$ . Similarly the backward path probability of a realization  $\tilde{\mathbf{x}}$  is defined as

$$\tilde{p}[\tilde{\mathbf{x}}|\tilde{x}_0] = e^{-\int_{\tilde{t}_0}^{\tilde{t}_1} \tilde{\omega}_{\tilde{x}_0, \tilde{x}_0}(t') dt'} \prod_{i=1}^n \tilde{\omega}_{\tilde{x}_i, \tilde{x}_{i-1}}(\tilde{t}_i) e^{-\int_{\tilde{t}_i}^{\tilde{t}_{i+1}} \tilde{\omega}_{\tilde{x}_i, \tilde{x}_i}(t') dt'}, \quad (2.4)$$

where  $\tilde{\omega}(\tilde{t}) = \omega(t_f - t)$  is the time reversed rate. By making a change of variables  $t' \rightarrow t_f - t'$ , the backward path probability is given by

$$\tilde{p}[\tilde{\mathbf{x}}|\tilde{x}_0] = e^{-\int_{t_n}^{t_f} \omega_{x_n, x_n}(t') dt'} \prod_{i=1}^n \omega_{\tilde{x}_{n-i}, \tilde{x}_{n-i+1}}(t_{n-i}) e^{-\int_{t_{n-i}}^{t_{n-i+1}} \omega_{x_{n-i}, x_{n-i}}(t') dt'}. \quad (2.5)$$

By making a change of variables  $i \rightarrow n - i$ , we note that the ratio of the path probabilities, the entropy flow of Eq. (2.2) is given by

$$\Delta S_F[\mathbf{x}] = \ln \prod_{i=1}^n \frac{\omega_{x_i, x_{i-1}}(t_i)}{\omega_{x_{i-1}, x_i}(t_i)}, \quad (2.6)$$

where  $\omega_{x_i, x_{i-1}}(t_i)$  is the transition rate from  $x_{i-1}$  to  $x_i$  at control parameter value  $\lambda(t_i)$ .

If the stationary distribution of the process with a fixed value of  $\lambda(t_i)$  is the canonical equilibrium distribution  $p(x_i) \propto \exp[-\beta E_{x_i}(t_i)]$ , rates are connected by the detailed balance condition (DB):

$$\frac{\omega_{x_i, x_{i-1}}(t_i)}{\omega_{x_{i-1}, x_i}(t_i)} = \frac{e^{-\beta E_{x_i}(t_i)}}{e^{-\beta E_{x_{i-1}}(t_i)}} = e^{\beta Q_i}. \quad (2.7)$$

We note that the condition above does not require that the process takes place in equilibrium [48]. Rather, it is an constraint placed on the ratio of rates which assumes that the environment is an ideal heat bath at an inverse temperature  $\beta = 1/k_B T_B$ . Often the condition of Eq. (2.7) is referred to as the local detailed balance condition (LDB). However, LDB is the observation that the constraint of Eq. (2.7) continues to hold for individual reservoir induced transitions, even when system is coupled to multiple reservoirs. By applying the LDB condition to Eq. (2.6), the entropy flow becomes

$$\Delta S_F[\mathbf{x}] = \beta Q[\mathbf{x}], \quad (2.8)$$

and the total entropy production is given by

$$\Delta S_T = \Delta S_S + \Delta S_F = \ln \frac{p_I[x_0]}{p_F[\tilde{x}_0]} + \beta Q[\mathbf{x}]. \quad (2.9)$$

Outside of equilibrium no well established definition for a single realization of entropy production exists. The total entropy production  $\Delta S_T$  in a trajectory was defined through Eq. (2.2). However, the trajectory dependent system entropy part  $S_S$  as a Shannon entropy  $S_S = -k_B \ln p(x)$  is justified by the fact that at least its average  $\langle S_S \rangle = -k_B \sum p(x) \ln p(x)$  in equilibrium is the thermodynamic entropy of the system. Furthermore, by assuming that the environment is an ideal heat bath (DB condition), the entropy flow becomes the entropy increase caused by the heat transfer to



the environment (Eq. (2.8)). In this case the entropy flow is often referred to as medium entropy production or reservoir entropy production and the mathematical definition of Eq. (2.2) coincides with classical definitions of entropy. Furthermore, the non-equilibrium entropy  $S_T$  has been shown to satisfy the non-equilibrium second law, connecting entropy to work and to the non-equilibrium free energy [54].

Another justification for calling  $\Delta S_T$ , defined as in Eq. (2.2), as the entropy comes from the fact that  $\Delta S_T$  satisfies the detailed fluctuation theorem (DFT):

$$\frac{p_I[x_0]p[\mathbf{x}|x_0]}{p_F[\tilde{x}_0]\tilde{p}[\tilde{\mathbf{x}}|\tilde{x}_0]} = e^{\Delta S_T}, \quad (2.10)$$

which due to conservation of probability implies the integral fluctuation relation (IFT):

$$\langle e^{-\Delta S_T} \rangle = \int p_I[x_0]p[\mathbf{x}|x_0]e^{-\Delta S_T}D[\mathbf{x}] = \int p_F[\tilde{x}_0]\tilde{p}[\tilde{\mathbf{x}}|\tilde{x}_0]D[\tilde{\mathbf{x}}] = 1, \quad (2.11)$$

where the integration is over all the possible paths. The equations above have been derived using other type of equations of motion, including hamiltonian dynamics [13, 17, 49, 50]. Furthermore, IFT and DFT have been experimentally verified with various different setups [1, 4, 6, 31, 55].

The DFT (Eq. (2.10)) states that the probability of observing a realization with negative entropy production  $\Delta S_T[\mathbf{x}] < 0$ , is exponentially suppressed compared to its positive entropy producing counterpart  $\Delta S_T[\tilde{\mathbf{x}}] = -\Delta S_T[\mathbf{x}]$ . Thus it's extremely rare, and impossible in the thermodynamic limit, to observe a macroscopic violation of the second law. In order to observe realizations with negative entropy production with a reasonable probability, the entropy production must be small in the process. Furthermore, using Jensen's inequality, the IFT (Eq. (2.11)) implies that on average the total entropy must be positive

$$\langle \Delta S_T \rangle \geq 0. \quad (2.12)$$

Remarkably, the second law of thermodynamics can thus be derived using stochastic microscopic equations of motion. However, one could argue that the second law was built in from the very beginning by defining the entropy as in Eq. (2.2).

### 2.3 Work fluctuation relations

In this section, we derive two famous fluctuation relations, namely the Crooks fluctuation relation, which is an special case of the detailed fluctu-

ation relation (Eq. (2.10)), and its integral version, the Jarzynski equality [11]. While these fluctuation relations can be considered merely as a special cases of the entropy fluctuation relations, due to historical reasons they deserve special attention in this thesis. Furthermore, these fluctuation relations, often called as work fluctuation relations, are particularly useful as they connect non-equilibrium quantities (work) to equilibrium ones (free energy). In Publication I we study the Jarzynski equality using the single electron box introduced in the next Chapter.

First we derive the Crooks fluctuation relation by assuming the microscopic reversibility condition

$$\frac{p[\mathbf{x}|x_0]}{\tilde{p}[\mathbf{x}|\tilde{x}_0]} = e^{\beta Q[\mathbf{x}]}, \quad (2.13)$$

which follows from Eqs. (2.2) and (2.8). The microscopic reversibility condition can be also derived using a slightly more general assumption of balance condition instead of the local detailed balance condition [48]. Furthermore, for the time being we assume that the process starts and ends in canonical equilibrium, even though in the end it turns out that the final state can be arbitrary. In equilibrium, the probability of sampling a state  $x(t)$  is given by  $p[x(t)] \propto \exp[\beta(F(t) - E_{x(t)}(t))]$ , where  $F(t) = -\beta^{-1} \ln \sum_x E_{x(t)}(t)$  is the Helmholtz free energy. We note that equilibrium quantities are time independent, but we explicitly write the time in  $x(t)$ ,  $E(t)$  and  $F(t)$  which refers to the value of the external control parameter  $\lambda(t)$ . By combining the microscopic reversibility condition (Eq. (2.13)) and the assumption on equilibrium initial and final states, we obtain

$$\frac{p_I[x_0]p[\mathbf{x}|x_0]}{p_F[\tilde{x}_0]\tilde{p}[\mathbf{x}|\tilde{x}_0]} = e^{\beta[Q+\Delta E]-\beta\Delta F} = e^{\beta(W-\Delta F)}, \quad (2.14)$$

where we used the first law of thermodynamics (Eq. (2.1)). The equation above is known as the Crooks fluctuation relation [12]. Its integrated version

$$\begin{aligned} \langle e^{\beta(W-\Delta F)} \rangle &= \int p_I[x_0]p[\mathbf{x}|x_0]e^{\beta(W-\Delta F)}D[\mathbf{x}] \\ &= \int p_F[\tilde{x}_0]\tilde{p}[\mathbf{x}|\tilde{x}_0]D[\mathbf{x}] = 1 \end{aligned} \quad (2.15)$$

is known as the Jarzynski equality. During the derivation we assumed that the final state at time  $t_f$  has to be an equilibrium state of the process. This, however, is not a necessary assumption. Assume that there exists an additional relaxation phase at  $t > t_f$ , during which the system is allowed to relax to equilibrium. The work and free energy would not change during the relaxation because work and free energy are functions of the control parameter  $\lambda(t)$ . Thus the work and the free energy, and

therefore the value of the dissipated work  $W - \Delta F$ , is the same whether the process ended in equilibrium or not. Further discussion on this issue and Jarzynski equality in general can be found in Ref. [5].

Both the Crooks fluctuation relation and the Jarzynski equality are remarkable because they connect equilibrium quantities ( $\Delta F$ ) to trajectory dependent non-equilibrium quantities ( $W$ ). Furthermore, Eqs. (2.14) and (2.15) are both equalities describing non-equilibrium processes. On a macroscopic level these processes are described by inequalities, of which the second law of thermodynamics is the most famous one.

### 3. Stochastic thermodynamics in single electron tunneling

Quantitative testing of fluctuation theorems is a cumbersome task. Statistically significant realizations with negative values of dissipation are rare, but it is exactly these values that have large contribution to the exponential averages, like those in the Jarzynski equality or in the integral fluctuation theorem. Therefore, one typically needs a large number of realizations to make statistically reliable estimates on the exponential average. In addition, one needs an interacting system that is experimentally feasible and in which the theoretical framework for dissipation is well defined.

The first experimental verifications of the fluctuation theorems were performed using polymer pulling experiments [4–6]. However, the problem with these systems is the limited number of realizations one is able to obtain. Furthermore, the analytical and numerical modeling of these systems is cumbersome.

The systems used in this thesis are based on single electron tunneling at low temperatures, like the single electron box which we introduce briefly in this chapter [56–59]. The single-electron devices satisfy the requirements of a limited number of relevant degrees of freedom, and clear separation of time and energy scales, making them particularly suitable for studying thermodynamics at small scales. Furthermore, single-electron devices are robust so that a large number of repetitions can be obtained in a short time, and the processes are relatively easy to model analytically and numerically [1, 60–62]. In this section, we will also briefly outline the basics of semi-classical single-electron tunneling at low temperatures, used in Publications I-IV.

### 3.1 Tunneling rates

The tunneling rates we use in this thesis can be formally obtained using a Fermi golden rule calculation of voltage biased elastic tunneling. As a starting point, we assume that the Hamiltonians of the two sides of the tunnel junction, the left and the right side, are given by  $H_L = \sum_{k,\sigma} \epsilon_k c_{k,\sigma}^\dagger c_{k,\sigma}$  and  $H_R = \sum_{q,\sigma} \epsilon_q c_{q,\sigma}^\dagger c_{q,\sigma}$ , respectively. Operators  $c_{k,\sigma}^\dagger$  and  $c_{k,\sigma}$  are the standard creation and annihilation operators of electron with spin  $\sigma$  and wave vector  $k$  or  $q$  with energies  $\epsilon_k$  and  $\epsilon_q$ , respectively. The charge transfer process from the left to the right electrode is described by the tunneling Hamiltonian  $H_{T,L} = \sum_{k,q,\sigma} T_{k,q} c_{k,\sigma}^\dagger c_{q,\sigma}$ , where  $T_{k,q}$  is the tunneling matrix element between states  $k$  and  $q$ . By carrying out a standard Fermi's golden rule calculation of the transition rate from an initial state  $i$  to a final state  $f$  in the perturbing Hamiltonian  $H_{T,L}$ , one obtains a tunneling rate

$$\gamma_{f \leftarrow i} = \frac{2\pi}{\hbar} |\langle f | H_{T,L} | i \rangle|^2 \delta(E_f - E_i), \quad (3.1)$$

where  $E_f$  and  $E_i$  are the energies of the final and initial states. We do the standard assumptions of elastic theory of tunneling, namely that the tunneling matrix elements do not depend on the wave vector,  $T_{k,q} = T$ . Furthermore, we assume that the timescales involved are longer than the electron-electron relaxation timescale and thus the electrons on the leads can be considered to form a Fermi gas. The tunneling rate is then given by:

$$\Gamma_{R \leftarrow L} = \sum_{i,f} \gamma_{f \leftarrow i} = \frac{2\pi |T|^2}{e^2 \hbar} \int_{-\infty}^{\infty} D_L(\epsilon) D_R(\epsilon) f_L(\epsilon, \mu_L) [1 - f_R(\epsilon, \mu_R)] d\epsilon, \quad (3.2)$$

where  $D_L$  and  $D_R$  are the densities of states on the left and right junctions, and  $f_L(\epsilon, \mu_L)$  and  $f_R(\epsilon, \mu_R)$  are the Fermi functions of the left and right electrodes describing the occupation probability of a state with energy  $\epsilon$  in chemical potential  $\mu$ . Term  $f_L(\epsilon, \mu_L)$  is the occupation probability of the state with energy  $\epsilon$  in the left electrode at chemical potential  $\mu_L$  and inverse temperature  $\beta_L$ . Term  $1 - f_R(\epsilon, \mu_R)$  is the probability of not occupying the state with energy  $\epsilon$  in the right electrode at chemical potential  $\mu_R$  and inverse temperature  $\beta_R$ . Since the product of the Fermi function almost vanishes outside small energy window  $\mu_R - \mu_L$ , the densities of states  $D_L$  and  $D_R$  can be considered constants and thus taken outside of the integral. The total tunneling rate then becomes

$$\Gamma_{R \leftarrow L} = \sum_{i,f} \gamma_{f \leftarrow i} = \frac{1}{e^2 R_T} \int_{-\infty}^{\infty} d\epsilon f_L(\epsilon, \mu_L) [1 - f_R(\epsilon, \mu_R)], \quad (3.3)$$

where the constant  $R_T = \hbar/(2\pi|T|^2 D_L D_R)$  is the tunneling resistance.

Let us now denote the difference in the chemical potentials as  $\Delta U = \mu_L - \mu_R$ , which is often called the “charging energy difference”.  $\Delta U$  corresponds to the electrostatic work done in transferring the electron charge  $e$  over the bias voltage  $V$ , thus  $\Delta U = eV$ . By changing variables  $E = \epsilon - \mu_L$ , and by measuring the energy from the Fermi level of the left electrode, the tunneling rate is given by

$$\begin{aligned} \Gamma_{R \leftarrow L} &= \frac{1}{e^2 R_T} \int_{-\infty}^{\infty} d\epsilon \frac{1}{e^{\beta(\epsilon - \mu_L)} + 1} \left[ 1 - \frac{1}{e^{\beta(\epsilon - \mu_R)} + 1} \right] \\ &= \frac{1}{e^2 R_T} \int_{-\infty}^{\infty} dE \frac{1}{e^{\beta_L E} + 1} \left[ 1 - \frac{1}{e^{\beta_R(E + \Delta U)} + 1} \right] \\ &\equiv \frac{1}{e^2 R_T} \int_{-\infty}^{\infty} dE f_L(E) [1 - f_R(E + \Delta U)], \end{aligned} \quad (3.4)$$

where we have dropped the chemical potentials from the notation and use  $\Delta U$  instead. In the case of quantum dots, used in Publication IV, the tunneling rate takes a slightly different form, since the continuous Fermi distribution of states is replaced by a single state with energy  $\epsilon_d$ . For example, if the right lead is replaced by a quantum dot at the same temperature as the left lead  $\beta_L = \beta$ , the number of empty states in the dot is given by  $\delta(\epsilon_d - E)$  (cf.  $1 - f_R(\epsilon, \mu_R)$ ) and thus the tunneling rate is given by

$$\Gamma_{D \leftarrow L} = \Gamma \frac{1}{e^{\beta(\epsilon_d - \mu_R)} + 1}, \quad (3.5)$$

where the coupling strength is given by  $\Gamma = (2\pi|T|^2 D_L)/\hbar$ .

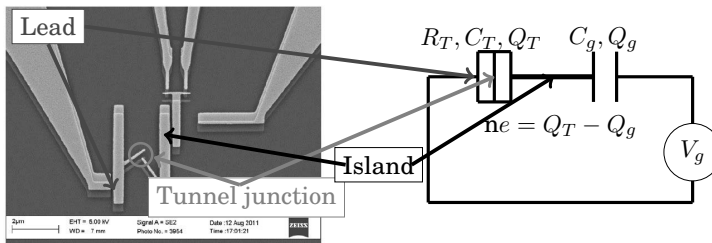
A more accurate treatment of tunneling phenomena would include the electromagnetic environment of the tunnel junction in the calculations [63]. However at sufficiently high temperatures, the simple model is in good agreement with experimental results [64].

### 3.2 Dissipation in voltage driven tunneling

In this section, we discuss fluctuation theorems in a more concrete context by introducing a simplistic single-electron tunneling device, the single electron box (SEB), used in Publications I-II and shown in Fig. 3.1. The SEB system consists of a metallic island, which is coupled to a metallic electrode (the lead) by a tunnel junction and to another electrode by a capacitor. The lead and the capacitor are attached to a voltage source that biases the tunnel junction with voltage  $V_g(t)$ . The electrons may tunnel between the lead and the island, but not between the capacitor and the

island. By controlling the bias voltage, we can control the rate of tunneling between the lead and the island in which the tunnel junction allows electrons to enter or leave the island.

We perform a certain *protocol*, a schedule in which the bias voltage  $V_g(t)$  is changed in time, multiple times, and gather the value of dissipation from each realization of the stochastic tunneling process. The distribution of dissipation obtained can then be used, for example, in testing fluctuation relations. Here, we focus on the normal metal version of the SEB, while in the experimental realizations, it is convenient to have lower tunneling rates and thus use tunnel junctions between normal metals and superconductors or semiconducting quantum dots [1, 62, 64].



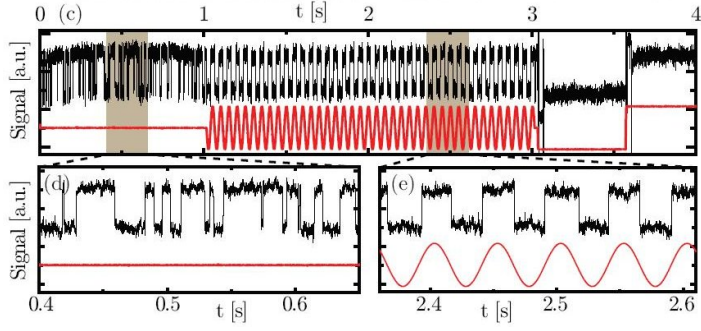
**Figure 3.1.** On the right a schematic picture of an SEB. On the left a photograph of the SEB and a charge measurement device (on the upper right side of the island), manufactured at O. V. Lounasmaa Laboratory, Aalto University.

The key feature of the single electron box is that at low temperatures, when the charging energy of the box  $E_C$  is much larger than the scale of the thermal fluctuations  $k_B T$ , typically order of 10 to 100  $k_B T$ , the island has a well defined discrete number  $n$  of excess electrons at all times. In a metallic SEB, the total equilibrium number of electrons is large, of the order of  $10^9$ , whereas  $n$  describes the deviation from that number. The number of excess electrons  $n$  on the island, corresponding to the state of the system, can be experimentally observed in real time by capacitively monitoring the charge of the island, as shown in Figure 3.2.

The energetics of the system are given by its Coulomb energy, represented by the Hamiltonian

$$H_S(n, n_g) = E_C(n - n_g)^2, \quad (3.6)$$

where  $E_C = e^2/(2C_\Sigma)$  is the charging energy,  $e$  is the electron charge,  $n_g = C_g V_g/e$  is the scaled voltage, and  $C_\Sigma = C_T + C_g + C_0$  is the total capacitance, which is given by the sum of those of the tunnel junction  $C_T$ , the gate capacitance  $C_g$ , and the self capacitance  $C_0$  of the island.

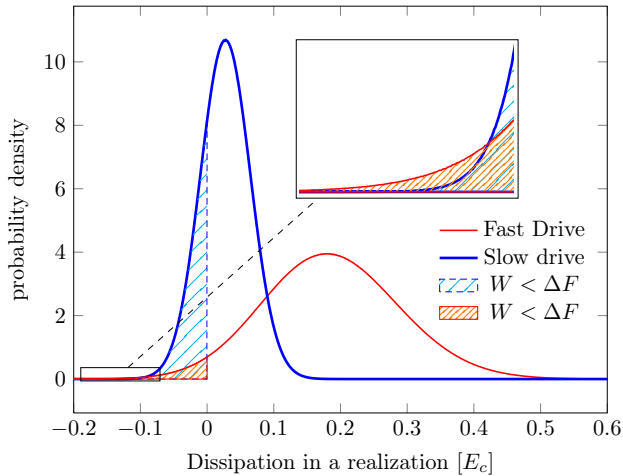


**Figure 3.2.** Experimental data showing the controllability of the SEB. The black line describes the state of the island and the red line the gate voltage. The signal represented by the black line has two clear distinct values, the upper one corresponding to the excited state  $n = 1$ , and the lower one corresponding to the state  $n = 0$ . When the bias voltage is in its minimum,  $n_g = 0$ , and the state  $n = 0$  is favored. In the maximum value of bias,  $n_g = 1$ , and the state  $n = 1$  is favored. By ramping the bias, the state  $n$  changes, as shown in the lower right panel. By fixing the value to  $n_g = 0.5$ , the occupation probability of both states  $n = 0$  and  $n = 1$  are equally probable, as shown in the lower left panel. The top panel shows data from a 4-second-long process. The lower panels show parts of the process in more detail. The figure is taken from Ref. [64].

The constant  $E_C$  gives the unit of energy to charge the box by an extra electron. To simplify the notation, we scale the gate voltage  $V_g$  and refer to  $n_g = C_g V_g / e$  as the control parameter in the setup. In a well-controlled experiment, the state of the SEB can have only two possible values, say  $n = 0$  and  $n = 1$ . To achieve this regime, the SEB needs to be operated at low enough temperatures,  $k_B T < E_C$ , and the values of the control parameter  $n_g$  may vary within one period (amplitude  $< 1$ ) only.

Depending on the value of  $n_g$ , either state  $n = 0$  or  $n = 1$  can be made energetically favorable, as can be seen from Eq. (3.6). Protocols we study in Publication I start from equilibrium at control parameter value  $n_g(0) = 0$  and end to  $n_g(t_f) = 1$ . Almost certainly in these processes, the initial state is  $n = 0$  since the probability of excited state  $n = 1$  is exponentially suppressed by  $\exp[-\beta E_C] \approx \exp[-40]$ . In the end of the process at control parameter value  $n_g(t_f) = 1$ , the state of the system is almost certainly  $n = 1$  and thus at least one tunneling event has occurred somewhere between  $t \in [0, t_f]$ . Each of the tunneling events dissipate heat by  $Q^\pm(t) = H_S(n^i, n_g(t)) - H_S(n^f, n_g(t)) = \pm E_C[2n_g(t) - 1]$ , where the  $+$  sign corresponds to a tunneling in ( $n^i \rightarrow n^f : 0 \rightarrow 1$ ) event and  $-$  to tunneling out ( $n^i \rightarrow n^f : 1 \rightarrow 0$ ) event. The dissipated heat  $Q^\pm(t) = \mp \Delta U$  corresponds to the electrostatic work  $\pm \Delta U(t) = \pm(\mu_L(t) - \mu_I(t))$  needed to





**Figure 3.3.** Probability distribution of dissipated work  $W - \Delta F$  for periodically driven SEB ( $\Delta F = 0$ ) for two different process speeds. Since it is extremely rare to observe a realization with a change in internal energy of the SEB, the work done equals the heat dissipated within the resolution accuracy of the figure. Thus the dissipated work in this case equals the dissipated heat. The faster process dissipates more, and for both process speeds, we can obtain negative values of dissipation. However for both speeds, the second law of thermodynamics is satisfied since dissipation is positive on average. Dissipation unit  $E_c$  (set to  $40 k_B T$  in the simulations) is the charging energy of the island. Data comes from Monte Carlo simulations with frequencies  $10^6 \text{s}^{-1}$  and  $10^7 \text{s}^{-1}$  taken from Publication I.

transfer the charge over the potential barrier, with a - sign. Here,  $\mu_I$  and  $\mu_L$  are the chemical potentials of the island and the lead, respectively.

The total dissipation in the process is given by the sum of the individual dissipating events. By repeating the protocol multiple times, we obtain statistics from the dissipation. Fig. 3.3 shows typical dissipation results from Monte Carlo simulations used in Publication I. More discussion on the energetics and different thermodynamic variables in the context of the SEB can be found in Refs. [60, 61].

## 4. Non-Markovian stochastic thermodynamics

Markovianity in general means that state of the system  $x(t + \Delta t)$  at time  $t + \Delta t > t$ , or at least its probability, can be known from the state  $x(t)$ . Interacting system is Markovian if all the degrees of freedom in the environment relax to equilibrium faster than that of the system. Thus, there is neither coupling nor back-action, often referred to as memory effects, caused by the coupling of degrees of freedom between the system and the environment. In this case, the environmental degrees of freedom can be coarse grained away, and the system evolution can be described using the system degrees of freedom alone. However by definition, an interacting system always has coupling of degrees of freedom between the system and the environment through an interaction Hamiltonian. Therefore, Markovian evolution is always, strictly speaking, an approximation for an interacting system. Especially for small systems and when interested in single realizations of a process, the non-Markovian effects may become important.

Some recent works have investigated entropy production under non-Markovian dynamics by considering different types of memory or by using thermodynamic arguments [8, 65–69]. However, the crucial assumption behind these models is that the environment is modeled by an ideal equilibrium environment and thus there cannot be non-equilibrium excitations in the environment during the process [67].

In this section, we address systems with a non-Markovian environment using stochastic thermodynamics and simultaneously review Publications I and II. In these setups, the environment is not static, but the dissipation from the system will affect the environment, causing back-action to the system. To study single realization thermodynamics, we must extend the framework to include the environmental degrees of freedom, which characterize the environment's time-dependent deviation from equilibrium. In

particular, we use the SEB system in a non-Markovian regime as a model system in which the results can be realized.

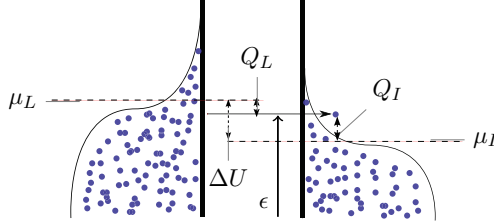
#### 4.1 Work fluctuation theorems

In this section, we consider the SEB in an experimentally feasible situation, where the collective electrons on the island are driven away from equilibrium by energy deposition from the tunneling electron. The realization of this regime requires a small heat capacity of the island. The relaxation of non-equilibrium excitations (the electron-electron relaxation rate), created by energy deposition of a tunneling electron, is the fastest time scale in the setup. In experiments, the internal relaxation time is of the order of  $\tau_{e-e} = 10^{-9}$  s, while the relaxation time back to the equilibrium by the coupling to the phonon bath of the substrate on which the SEB is fabricated, is of the order of  $\tau_{e-p} = 10^{-4}$  s. If the process timescale  $\tau_d$  satisfies  $\tau_{e-e} \ll \tau_d \ll \tau_{e-p}$ , the electrons on the small metallic island of the tunnel junction adopt a Fermi distribution at an effective temperature during the drive, which can be seen as a change in the tunneling rates [55].

We note that the effective temperature is a result of the stochastic energy deposition from tunneling electrons due to the driving of the control parameter. As a result the tunneling rates change as if the temperature of the system would have changed. Thus, the effective temperature is a result of system dynamics and is not controlled externally by another heat bath. In typical experiments, the heat capacity of the small metallic island is of the order of  $1000k_B$ , and the operating temperature around 100 mK, resulting in equilibrium temperature fluctuations in the order of a few mK. Depending on the drive protocol, the speed of the drive in particular, the heat dissipation per tunneling electron is of the order of  $k_B T$  resulting in temperature fluctuations of the order of 0.1 mK per tunneling event.

Let us consider that an electron with energy  $E$  tunnels from the lead at chemical potential  $\mu_L$  to the island at chemical potential  $\mu_I$ . The energy of the system  $H_S(n, n_g)$  (Eq. (3.6)) changes by the heat dissipated  $\mu_I - \mu_L$ . The dissipated heat is divided between two sides of the tunnel junction, the island and the lead. The heat deposited to the lead is given by  $Q_L = \mu_L - E$  and the heat to the island by  $Q_I = E - \mu_I$ , as illustrated in Fig. 4.1. We note that the Fermi gas of electrons in both the

lead and the island are thus reservoirs for the system and the changes in their energies,  $Q_I$  and  $Q_L$ , are to be considered as heat. The system in turn is represented by the electrostatic energy of Eq. (3.6). The energy of the system  $H_S(n, n_g)$  thus excludes the energies of the electrons in the Fermi gases, which are irrelevant for the electrostatic description of the system. The total dissipated heat in the tunneling event is given by  $Q_T = (\mu_L - E) + (E - \mu_I) = \mu_L - \mu_I = \Delta U$ .



**Figure 4.1.** When an electron with energy  $E$  tunnels from  $\mu_L$  to  $\mu_I$ , the total dissipated heat in the process equals the difference in the chemical potentials  $\Delta U = \mu_L - \mu_I$ . From the total dissipated heat, the left electrode receives  $Q_L = \mu_L - E$  and the right electrode  $Q_R = E - \mu_R$ . The blue dots and the curves on the lead and the island sides describe Fermi occupation probabilities of the electrons.

The processes start from an initial equilibrium at time  $t = 0$ . The electron energy distribution on the island is given by the equilibrium value  $f_{T(0)}$ , where  $T(0) = T_B$  is the temperature of the phonon bath. Assume that at time  $t$ , an electron with energy  $E$  tunnels from the lead to the island, depositing a heat amount of  $Q_I$  into the island. Due to the fast relaxation of the non-equilibrium excitation inside the island, the temperature of the island changes rapidly to  $T(t) = T_B + Q_I/C$ , where  $C$  is the heat capacity of the island. Thus, the electron energy distribution on the island after the tunneling event is given by  $f_{T(t)}$  [61, 64]. Due to the heating, the next tunneling event is driven by the tunneling rate

$$\Gamma^\pm(t)_{T(t)} = \frac{1}{e^2 R_T} \int_{-\infty}^{\infty} f_{T_B}(\pm E) \{1 - f_{T(t)}(\pm[E + \Delta U(t)])\} dE, \quad (4.1)$$

which depends on the previous tunneling events due to the temperature  $T(t)$ . Here + denotes for tunneling in and - for tunneling out. Thus, there is memory of the previous states implying that the process is non-Markovian.

In equilibrium, the rates  $\Gamma^\pm$  are local detailed balance (LDB) connected, but if  $T(t) \neq T_B$ , the LDB condition is broken. In publication I, we show that by expanding the tunneling rates with respect to the inverse temperature difference  $\Delta\beta = 1/k_B T - 1/k_B T_B$ , we obtain  $\Gamma^-/\Gamma^+ = \exp(-\beta\Delta U)[1 -$

$\frac{1}{2}\Delta U\Delta\beta]$ , which yields the standard LDB in the case of  $\Delta\beta = 0$ . Using the expansion in  $\Delta\beta$  into first order, the work fluctuation theory for the dissipated work  $W_d = W - \Delta F$  is given by

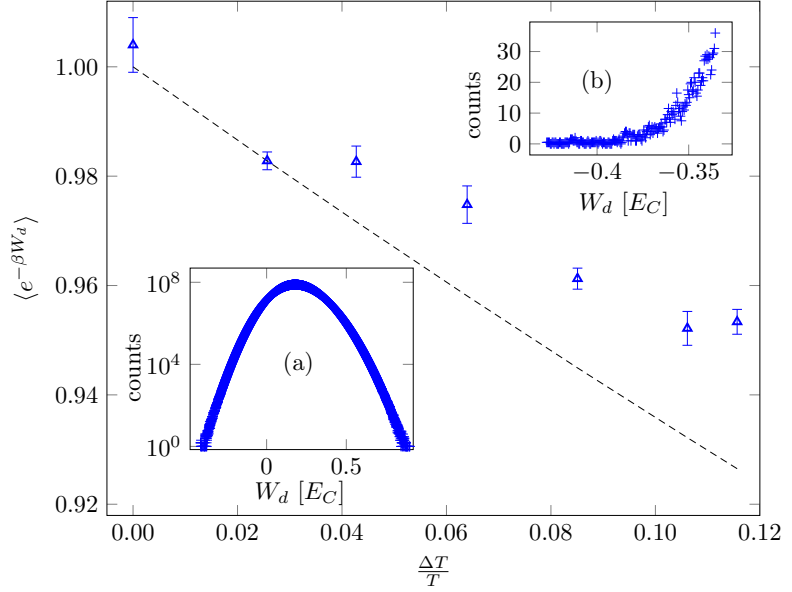
$$\langle e^{-\beta W_d} \rangle = 1 - \frac{1}{2}\langle Q \rangle \Delta\beta. \quad (4.2)$$

In the derivation of the result above, we assume that the protocol consists of a linearly increasing part  $t \in [0, t_f/2] : n_g(t) = 2t/t_f$  and a linearly decreasing one  $t \in [t_f/2, t_f] : n_g(t) = 2 - 2t/t_f$ . Furthermore, we assume that during the increasing part of the protocol, only one tunneling in event occurs, and in the decreasing part only one tunneling out event occurs. Experimentally this regime, where the number of transitions is restricted to one per increasing and decreasing leg is obtained with a fast change of  $n_g(t)$ , that is, making  $t_f$  short. In addition, we assume that during the protocol, the temperature of the island does not relax to its equilibrium temperature, which is reasonable given that the process is fast enough,  $t_f \ll \tau_{e-p}$ , so that the electron-phonon relaxation rate plays no role during the drive.

In Publication I, we study dissipation in the SEB system numerically, using Monte Carlo simulations. The numerical results are also shown in Fig. 4.2. The theoretical result of Eq. (4.2) and the results from the Monte Carlo simulations using different heat capacity values  $C$  are in good agreement in small values of  $\Delta\beta$ . Deviation between the numerical results and the theoretical prediction is to be expected in smaller heat capacity values, corresponding larger deviation in  $\Delta\beta$ , since the result of Eq. (4.2) is based on expansion in  $\Delta\beta$ . Furthermore in the numerical simulations, the number of transitions was not restricted to one per increasing and decreasing leg.

The result of Eq. (4.2) states that the dissipated work  $W - \Delta F$  does not satisfy the integral fluctuation relation  $\langle \exp[-\beta(W - \Delta F)] \rangle = 1$  in the SEB system if overheating is present. In Publication I, we show that the total entropy production  $S_T$ , defined in Eq. (2.2), satisfies the integral fluctuation relation  $\langle \exp[-S_T] \rangle = 1$  as we already expect from its mathematical construction. Since the process starts from equilibrium, meaning that the initial states are sampled from equilibrium distribution,  $W_d = W - \Delta F$  should coincide with the total entropy production  $S_T$ , but clearly this is not so, since  $S_T$  satisfies the integral fluctuation relation and  $W_d$  does not.

The difference between  $S_T$  and  $W_d$  can be understood as a consequence of the additional entropy production in the environment. This entropy



**Figure 4.2.** Simulation results (triangles) of  $\langle e^{-\beta(W-\Delta F)} \rangle$  and the corresponding theoretical approximation of the single jump and small temperature increase  $\Delta\beta$  of Eq. (4.2) (dashed line). The horizontal axis shows the average temperature increase  $\Delta T/T$  after the first half sweep. The corresponding values of the heat capacities from left to right are:  $2000k_B$ ,  $1200k_B$ ,  $8000k_B$ ,  $600k_B$ ,  $480k_B$  and  $440k_B$ . Smaller heat capacity leads to a bigger change in the temperature. The error bars are the standard error of the mean of the corresponding data. Each data point is obtained from  $1.75 \times 10^{11}$  independent realizations of the tunneling process. The insets (a) and (b) show the sampling from simulation with  $C = 480k_B$  with  $1.75 \times 10^{11}$  realizations, (b) demonstrating explicitly that even the tails of the distribution are well sampled. The charging energy  $E_C$  was set to  $40 k_B T$ .

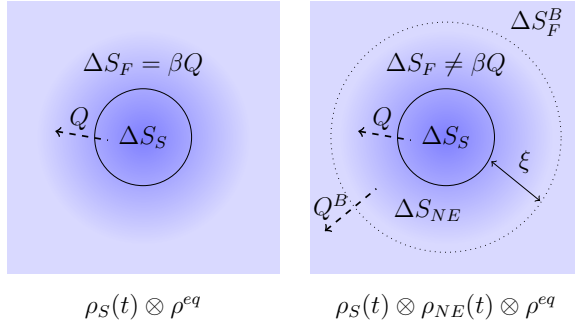
is not measured from the work done from the system degrees of freedom alone. Therefore, to determine whether the work fluctuation relation should hold or not, one should in principle measure the environment as well and make sure no deviation from equilibrium is present. From the point of view of experiments and accurate theoretical models, this is challenging. Furthermore, as discussed in the beginning of this section, non-Markovian effects are always present when the system interacts with its environment.

## 4.2 Entropy production

We have shown that the fluctuations of dissipated work  $W_d = W - \Delta F$  do not satisfy the integral fluctuation relation in the SEB system if the electron population on the island is driven outside of equilibrium during the process. However, the total entropy production as defined in Eq. (2.2) satisfies the integral fluctuation relation. In this section, we study what other components of entropy production exist in addition to the Markovian entropy  $S^m \equiv \beta(W - \Delta F)$ . This additional *non-Markovian entropy production* needs to be included in the entropy production to obtain the full entropy production. The content of this section is based on Publication II, in which additional details can be found.

From a more general perspective and not limited to the SEB system alone, we study entropy fluctuations in a system coupled to an environment that is not an ideal heat bath during the process. We assume that there exists a subset of degrees of freedom of the environment that are correlated with the dynamics of the system and do not relax to equilibrium defined by  $T_B$  on a time scale faster than that of the system. We call these degrees of freedom the *non-equilibrium subsystem* (NE) of the environment, which cannot be described by the bath's thermal equilibrium distribution during the drive and must therefore be explicitly taken into account. In the case of the SEB, the NE is realized by the  $10^9$  electrons on the island.

In general, a rigorous study of such non-equilibrium fluctuations in the environment requires a complete knowledge of the time evolution of all the degrees of freedom inside the NE. However, if the NE has an internal relaxation time  $\tau_{NE}$  much shorter than the characteristic relaxation time scale of the system  $\tau_S$ , we can simplify the problem. In addition, we assume that the NE is weakly coupled to the rest of the environment



**Figure 4.3.** A schematic of sources of additional entropy generation caused by non-equilibrium excitations in the environment. The left panel illustrates a purely Markovian system, in which the environment is assumed to stay in equilibrium during the process. The right panel shows the case of an NE, with a measure  $\xi$  that now constitutes a subset of the bath degrees of freedom. The NE can be driven away from equilibrium due to the coupling to the system leading to an additional entropy production (see text for details).

such that all the degrees of freedom in the NE mutually relax to a (non-equilibrium) state at time scales  $\tau_{NE} \ll \tau_S$ . We note that even if the relaxation inside the NE is the fastest time scale, Markovian evolution does not follow here. The NE is finite and due to energy deposition from the system during the trajectory  $x$ , the NE will evolve through quasi-equilibrium states different from the equilibrium state of the rest of the environment. That is to say, the transition  $x_i \rightarrow x_{i+1}$  is affected by the earlier transitions  $\{x_j \rightarrow x_{j+1}\}$ , where  $j < i$ , meaning that the evolution is non-Markovian.

In Markovian dynamics, entropy production can be split into entropy flow  $\Delta S_F$  and the system entropy generation  $\Delta S_S$  (cf. Eq. (2.2)). The entropy flow is, due to the equilibrium property of the surrounding heat bath, given by  $\beta Q$ , where  $Q$  is the dissipated heat. In the case of the NE setup, the entropy flow  $\Delta S_F$  is no longer given by  $\beta Q$  since the heat dissipated from the system does not transfer into an equilibrium environment. Furthermore, the NE of the setup is now time dependent, so its Shannon entropy will change during the process ( $\Delta S_{NE}$ ). In general, there can be heat transfer from the NE to the rest of the heat bath ( $Q^B$ ) and associated entropy flow generated by it ( $\Delta S_F^B$ ). In the case of the SEB example, the latter terms will be neglected. In Fig. 4.3, we illustrate the different entropic terms in the Markovian and the NE dynamics.

In the case of the non-Markovian SEB, the internal relaxation time of the NE,  $\tau_{NE}$ , corresponds to the electron-electron relaxation time  $\tau_{e-e}$ , which is the fastest time scale in the problem. Therefore, the electron



population on the island forms a well-defined NE in the setup. Next, we go through the modification to entropy production caused by the NE and studied in Publication II in more detail. First, since the system (the number of excess electrons  $n \in \{0, 1\}$  on the island) is not driven by the equilibrium temperature bath, the Markovian entropy flow, Eq. (2.8) is replaced by

$$\Delta S_F = \beta Q_T + \Delta \beta Q_I, \quad (4.3)$$

where the latter term is the additional contribution from the NE of the setup, the small island. The term  $Q_I$  is the heat dissipated to the island in a transition. Equation (4.3) results from the fact that the system is effectively driven by a different temperature heat bath during the drive.

Furthermore, another contribution to the non-Markovian sources is the entropy of the NE, that is, the electron population of the island, given by  $S_I = -\ln f_T$ , where  $f_T$  is the probability distribution of the degrees of freedom inside the NE ( $\rho_{NE}$ ), i.e. the Fermi function of the island at temperature  $T$ . In a tunneling event at time  $t$ , an electron with energy  $E$ , which changes the temperature of the island from  $T_i$  to  $T_{i+1}$  and the electron population distribution from  $f_{T_i}$  to  $f_{T_{i+1}}$ , induces an entropy change of

$$S_I^\pm = \log \frac{f_{T_i}[\pm(E + \Delta U(t))]}{f_{T_{i+1}}[\pm(E + \Delta U(t))]}, \quad (4.4)$$

where the + sign corresponds to a tunneling in event and - to tunneling out. As shown in the appendix of Publication II in detail, the rates are not LDB connected ( $\ln[\Gamma^+/\Gamma^-] = \beta Q_T$ ), but connected through entropies  $\Delta S_F$  and  $S_I$ .

Initially, the NE is coupled to the bath, and the initial temperature  $T_I$  is sampled from distribution  $p_I(T_I)$ , which in equilibrium is normally distributed with a variance  $k_B T^2/C$ , where  $C$  is the heat capacity of the island. This is due to the fact that the island is small and has a finite heat capacity. Thus in addition to the tunneling events,  $S_I$  can change due to the heat transfer from the bath. The associated entropy is given by

$$\Delta S_I^T = \ln \frac{p_I^T(T_I)}{p_F^T(T_F)}, \quad (4.5)$$

where  $p_F(T_F)$  is the probability to sample the final temperature  $T_F$ . We neglected the entropic terms resulting from the interaction between the NE (the island of the SEB) and the rest of the environment (the phonon bath). This is justified by the fact that the electron-phonon relaxation time  $\tau_{e-p}$  is orders of magnitude longer than the process time. Thus, we

can assume that there is no energy transfer between the NE and its environment during the drive. In Publication II, we show that by adding the correction to the entropy flow,  $\Delta\beta Q_I$ , and the change in the Shannon entropy of the NE due to the heat transfer from the system ( $S_I$ ) and from the outer equilibrium bath ( $S_I^T$ ), defined in Eqs. (4.3), (4.4) and (4.5), respectively, we obtain the full entropy production. This entropy satisfies the integral fluctuation theorem

$$\langle e^{-S_T} \rangle = \langle e^{-\beta(W-\Delta F)-\Delta\beta Q_I-\Delta S_I-\Delta S_I^T} \rangle = 1. \quad (4.6)$$

To summarize, there is always additional entropy production along with the Markovian sources of entropy if the system is interacting. In general, the identification of these additional terms is difficult and requires measurement on the environmental degrees of freedom. However, using additional relaxation time assumptions, the problem can be simplified to the NE model, in which the non-equilibrium conditions in the environment can be explicitly taken into account. The overheated SEB system is a physical model system in which the conditions for a well-defined NE are realized.

To study dissipation in single realizations with a non-equilibrium environment, one needs to include the relevant environmental degrees of freedom into the equations of motion. If one coarse grained the environmental degrees away, one could in principle write a coarse grained description, for example, a generalized Langevin equation for the system. This would be enough to study the dynamics of the system alone but not enough to obtain knowledge about the entropy production or fluctuations of work.

A possible approach to study entropy production under non-equilibrium environments is to include the non-equilibrium part of the environment as a part the system. In that approach, the environment of this extended system is in equilibrium and thus the dynamics of the extended system are Markovian. The common fluctuation relations are expected to be satisfied in the setup without additional modifications. The difficulty however is then that the thermodynamic variables, such as heat and internal energy are defined for the extended system. If one eventually is interested on the thermodynamics of the system alone, one needs trace back the system energetics from the extended system energetics. This might turn out difficult, as one needs to know for example how the source of heat dissipated from the extended system was divided between the system and the non-equilibrium subsystem.



## 5. Stochastic thermodynamics of information

In this chapter, we study thermodynamics of setups consisting of two subsystems. The two subsystems are coupled to each other and to a thermal environment. We study correlations between the subsystems by formalizing them using information thermodynamics. By adjusting parameters in these setups, one subsystem can be made to function in a manner apparently contradicting the second law. The second law is retained in these setups by taking the dissipation of the other subsystem into account. However, without knowing the existence of the other subsystem, the first subsystem would seem to violate the second law of thermodynamics. Therefore, these setups are also commonly referred to as Maxwell's demons.

In the previous chapter, we discussed system-environment correlations from a point of view of non-Markovian dynamics. Also such setups, and correlations there, could have been mathematically formalized using information thermodynamics. For example one could have formalized the change of temperature of the NE in the non-markovian SEB as a result of measurements and feedbacks on the tunneling electrons. Later on in this section, we discuss a generalized Jarzynski equality, where correlations between the system and its environment are taken into account in this manner.

In information thermodynamics, the correlated environment of the system of interest, meaning the other subsystem, consists of preferably limited number of degrees of freedom. This enables to quantitatively characterize different information thermodynamic quantities, making the framework of information thermodynamics more suitable than the non-Markovian approach. In addition to creating different Maxwell's demon setups and studying the operation of the system-demon compound, we are interested in how the role of information can be implemented in the thermody-

namic description of the setups.

In publication III, we study thermodynamics of measurement using a setup which can be also made to work as a Maxwell's demon device. In Publication IV, we study various thermodynamic properties of an autonomous information machine, which can be made to function as a cooler.

## 5.1 Information in thermodynamics of small systems

In the original Maxwell's demon thought experiment, an unknown intelligent being, a demon, controls the positions of atoms in a box. The box is divided in half and the Demon controls the door between the two halves of the box, based on measurements on the atoms. By measuring the velocity of each atom, the demon opens and closes the door so that in the end the fast and the slow atoms are on different sides of the box. As a result, the demon has created a temperature difference which could be used as a source of work. The second law is retained by noting that work is needed for the operation of the demon.

By using information about the microscopic state of the system, the demon could apparently violate the second law of thermodynamics. However, while the entropy of the system decreased in the process, in order to retain the second law, the entropy of the surroundings must increase by at least the same amount. Nevertheless, the Maxwell's demon thought experiment shows how the restrictions defined by the second law can be loosened if there is utilization of information.

The modern information thermodynamics of small systems sees the action of the demon divided in two parts. In the first part, the demon measures the system to obtain information about the current microscopic state of the system. In the second part, the Demon performs feedback to lower the system entropy.

From an information theoretical point of view, the information content of an event  $x$  is  $-\ln[p(x)]$ . Thus the rarer the event, the more information it carries. The average information content is the Shannon information (entropy)

$$H(X) = - \sum_x p(x) \ln p(x), \quad (5.1)$$

which measures the uncertainty of the variable  $x$  [70]. The less we know about  $x$ , the greater the Shannon information  $H(X)$ . Now consider that  $x$  is not an independent variable, but there exists a correlated variable  $y$

and their joint probability distribution function is given by  $p(x, y)$ . Mutual information between  $x$  and  $y$  is defined as

$$I(x, y) = \ln \frac{p(x, y)}{p_X(x)p_Y(y)}, \quad (5.2)$$

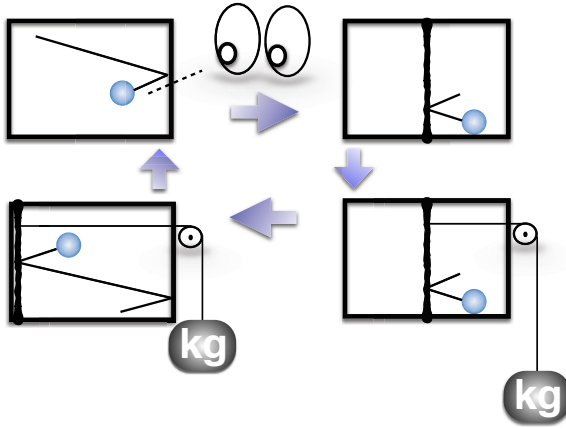
where  $p_X(x) = \sum_y p(x, y)$  and  $p_Y(y) = \sum_x p(x, y)$  are the marginal probability distributions of  $x$  and  $y$ , respectively. The quantity  $I(x, y)$  measures how much the states  $x$  and  $y$  are correlated, or equivalently how well the state  $x$  or  $y$  can be known by knowing the other one. The average of mutual information,  $\langle I \rangle = \sum_{x,y} p(x, y) I(x, y)$  tells how much the two subsystems are correlated on average. For example, if the variables  $x$  and  $y$  are completely independent,  $p(x, y) = p_X(x)p_Y(y)$  and the average mutual information  $\langle I \rangle = 0$ . Furthermore, the mutual information is given by the Shannon entropies as

$$\langle I \rangle = H(X) + H(Y) - H(X, Y), \quad (5.3)$$

where  $H(X, Y) = -\sum_{x,y} p(x, y) \ln[p(x, y)]$ . Since the Shannon entropy is always non-negative and  $H(X, Y) \geq \max[H(X), H(Y)]$ , the maximum mutual information between  $X$  and  $Y$  is given by  $\min[H(X), H(Y)]$ .

Suppose we have measured the system state  $x$  with some kind of a measurement device and obtained a measurement outcome  $y$ . If our measurement is perfect, we know that  $x = y$ , i.e.  $p(x, y) = 0$  if  $x \neq y$ . However, in general there can be sources of error and we could obtain a measurement outcome  $y$  even if the state of the system  $x$  differs from it. In this case, the mutual information  $\langle I(x, y) \rangle$  describes the accuracy of our measurement and how much information we have on the system. This information could be exploited to extract usable work out of it.

A simple example of a setup, in which information can be used to extract work is the famous example of a Szilard engine illustrated in Fig. 5.1. Initially an ideal gas particle moves freely inside a box in volume  $V$  in thermal equilibrium at temperature  $T$ . We measure the location of the particle and obtain, for example, that it is on the right side of the box. After that we insert a movable barrier in the middle of the box, which divides the volume of the box in half. Since the particle is now trapped in a smaller volume on the right side of the box, the pressure has increased. By allowing the barrier to move, and for example attaching a weight to it, we can extract work out of the system and restore it to the potential energy of the weight. Isothermally moving the barrier to the location of the left wall and using the ideal gas equation of state, the work extracted



**Figure 5.1.** Illustration of the Szilard engine. The position of an ideal gas atom is measured followed by an installation of a movable barrier which divides the volume of the box in half. The atom is then allowed to relax isothermally to its original equilibrium volume. In the process the barrier lifts a weight against gravity performing  $k_B T \ln 2$  of work in the weight.

is given by

$$W_{ext} = \int_{V/2}^V p dV = k_B T \ln 2. \quad (5.4)$$

In this example, we were able to extract work because we had information on the position of the particle before the insertion of the barrier. We can now be more quantitative on this by studying the role of mutual information in the operation of the Szilard engine. Since we assumed that the measurement is perfect, the joint probability distribution is given by  $p(\text{left}, \text{left}) = p(\text{right}, \text{right}) = 1/2$  and  $p(\text{left}, \text{right}) = p(\text{right}, \text{left}) = 0$ , where the states are the system state and the measurement outcome as defined earlier in this section. The system Shannon entropy is given by  $H(X) = -\sum_x p_X(x) \ln p_X(x) = \ln 2$ , where  $x \in \{\text{left}, \text{right}\}$ . Similarly, we obtain for  $H(Y) = \ln 2$  and  $H(X, Y) = \ln 2$ . Therefore, by using Eq. (5.3) or the fact that in a perfect measurement the mutual information is given by  $\min[H(X), H(Y)]$ , we obtain  $\langle I \rangle = \ln 2$ . Since the work extracted was given by  $k_B T \ln 2$ , the work and the mutual information are related in this example by  $W_{ext} = k_B T \langle I \rangle$ .

From a more general point of view, in the case of the Szilard engine the measurement was perfect, the process was quasi-static and the system was a simplistic ideal gas system. In a more complex setup, the relation between the information and work extraction may not be so simple. However, it can be shown that a general feedback system obeys the following

generalized Jarzynski equality [38–40]

$$\langle e^{\beta(\Delta F - W) - I} \rangle = 1. \quad (5.5)$$

The equation above implies, using Jensen’s inequality,  $\langle \beta(\Delta F - W) - I \rangle \geq 0$ , which results to a fundamental bound for extracted work in the presence of information:

$$W_{ext} \leq -\Delta F + k_B T \langle I \rangle. \quad (5.6)$$

Thus, if we have  $\langle I \rangle$  of mutual information about the system, we can extract  $-\Delta F + k_B T \langle I \rangle$  of work out of the system. Since without the information the maximum available work is given by  $-\Delta F$ , the information we have on the system allows us to extract more work. Equation (5.6) can be seen as a generalization of the second law of thermodynamics to feedback systems, where information is a resource of work. If the initial and the final states are the same, such as in the case of the Szilard engine, the process becomes cyclic and the free energy difference  $\Delta F$  vanishes. As a result, we have made a cyclic process which extracts usable energy out of a single heat reservoir. This is a violation of the second law and thus a Maxwell’s demon mechanism, producing additional entropy somewhere, should be present.

Returning to the original Maxwell’s demon, it obtains information  $I$  about the individual atoms by measuring their velocities. Depending on the measurement outcome, the demon either opens or closes the door between the two sides of the box. Because the demon had information  $I$  in its use, it was able to perform the feedback by controlling the door and creating a temperature gradient in the box. Both the Szilard engine and the Maxwell’s demon can thus be referred to as information machines. In modern literature Maxwell’s demon is often characterized as any feedback controller that utilizes information about the microscopic state of the system at the level of thermal fluctuations. Due to the developments in manipulation of microscopic systems, various modern realizations of Maxwell’s demon setups exists, where both Eqs. (5.5) and (5.6) have been experimentally studied. A selection of these setups are listed in the Introduction chapter.

## 5.2 Information between two subsystems

In the previous section we studied the role of information between the demon and the system to which the feedback was performed. However,



since the demon was not included in the model, thermodynamics of the demon itself, and that of the whole system-demon compound, were not accessible. In this section, we include the demon into the general theoretical framework and thus we will be able to discuss also the measurement phase during which the demon obtains information about the system.

In the case of the original Maxwell's demon, the measurement and the feedback were both performed by the same entity, the demon. However, in the Szilard engine it is not clear exactly how the measurement is done and whether or not the feedback is carried out by the same entity. The actual physical implementation of the entity measuring and performing the feedback could consist of a measurement device, a memory and an entity reading the measurement outcome from the memory and acting accordingly. For the time being, let us assume that the measurement was carried out by the demon which has an internal memory, and thus the measurement device, the memory and the demon refer to the same entity. These different terms could be interchanged depending on the point of view, and for example in Publication III, the use of all these three terms could be justified for the other correlated subsystem. The framework we discuss next commonly refers to the outside entity as "memory", and thus we will use this term [36, 39, 41].

By  $X$  we denote the system whose state  $x$  we wish to measure. Furthermore, by  $Y$  we denote the memory, which stores the measurement outcome  $y$  of state  $x$ . We assume that the memory is initially detached from the system and during the measurement the memory interacts with the measured system storing the measurement outcome. Finally, the memory is reset to its initial state, making the whole process cyclic. We study thermodynamics of information exchange in each step of the process. In Publication IV, we study an explicit model which is based on coupled quantum dots, in which the results of this section are applied.

We consider the total system, including the system to be measured and the memory, as one combined system, which helps us to write down the thermodynamics from a more global perspective. The Shannon entropy of the total combined system,  $\langle S_{X+Y} \rangle$ , is given by  $\langle S_{X+Y} \rangle = -k_B H(X, Y)$ , where the only difference to the earlier Shannon information (Eq. (5.1)) is the scaling with the physical constant  $k_B$ . Thus the entropies of the systems  $X$  and  $Y$  are given by  $\langle S_X \rangle = -k_B H(X)$  and  $\langle S_Y \rangle = -k_B H(Y)$ , respectively. By straightforward use of the definitions, the entropy  $\langle S_{X+Y} \rangle$

can be written as

$$\langle S_{X+Y} \rangle = \langle S_X \rangle + \langle S_Y \rangle - k_B \langle I \rangle. \quad (5.7)$$

Analogous to the splitting of the entropy, we split the total energy  $\langle E_{X+Y} \rangle = \sum_{x,y} p(x,y) E_{X+Y}$  into the system contribution  $\langle E_X \rangle = \sum_x p_X(x) E_X(x)$ , the memory contribution  $\langle E_Y \rangle = \sum_y p_Y(y) E_Y(y)$ , and the interaction part  $\langle E_I \rangle = \sum_{x,y} p(x,y) E_I(x,y)$ . Here  $E_X$ ,  $E_Y$  and  $E_I$  are the parts of the total combined system Hamiltonian, which depend on the degrees of freedom  $x$ ,  $y$  and the terms containing both, respectively. Using the above splitting of the energy and entropy, the non-equilibrium free energy

$$\mathcal{F} = \sum_{x,y} p(x,y) E(x,y) + k_B T \sum_{x,y} p(x,y) \ln p(x,y) \quad (5.8)$$

for the total combined system can be written as

$$\mathcal{F}_{X+Y} = \mathcal{F}_X + \mathcal{F}_Y + \langle E_I \rangle + \beta^{-1} \langle I \rangle, \quad (5.9)$$

where  $\mathcal{F}_X = \langle E_X \rangle + \beta^{-1} \sum_x p_X(x) \ln p_X(x)$  is the free energy of the system and  $\mathcal{F}_Y = \langle E_Y \rangle + \beta^{-1} \sum_y p_Y(y) \ln p_Y(y)$  is the free energy of the memory.

Outside of equilibrium no well established definition of free energy exists. The extension of the equilibrium Helmholtz free energy to non-equilibrium by a straightforward application of the same formula, Eq. (5.8), may seem too careless. However, the non-equilibrium free energy  $\mathcal{F}$  coincides with the known equilibrium free energy  $F$  in equilibrium. Furthermore, it has been shown that the equilibrium free energy  $F$  is the minimum of  $\mathcal{F}$ , i.e.  $\mathcal{F}[p(t)] \geq F[p^{eq}(t)]$ , and the minimum of  $\mathcal{F}[p(t)]$  is obtained for the equilibrium Boltzmann distribution  $p^{eq}(t) \propto \exp[-\beta E_X(x)]$ . In addition, it has been shown that the entropy production  $\Delta S_T$  between two non-equilibrium states satisfies the non-equilibrium version of the second law  $\Delta S_T \geq W - \Delta \mathcal{F}$ . These facts together support the extension of the free energy outside of equilibrium as  $\mathcal{F}$  (Eq. (5.8)) [54].

Assume that the measurement is performed by manipulating an external parameter, which brings the system and the memory in contact. During this measurement the memory interacts with the measured system and stores the measurement outcome of  $x$  into the variable  $y$ . The external work  $W_{meas}$ , required to perform the measurement, is bounded by  $W_{meas} \geq \Delta \mathcal{F}_{X+Y}$ , because the entropy generated in the measurement must be positive according to the second law, that is  $\langle S_M \rangle = W_{meas} - \Delta_m \mathcal{F}_{Y+X} \geq 0$ , where  $\Delta_m$  refers to the change in the measurement phase.

If we further assume, as typically done in the literature, that the measurement does not change the state of the system  $x$  during the measurement, the measurement work satisfies

$$W_{meas} \geq \Delta_m \mathcal{F}_Y + \beta^{-1} \Delta_m \langle I \rangle + \Delta_m \langle E_I \rangle, \quad (5.10)$$

where we used the splitting of the total free energy, Eq. (5.9). In a measurement, the measurement outcome  $y$  should become more correlated to the state of the system  $x$ . Thus the mutual information  $\langle I \rangle$  should increase in the measurement. We note that Eq. (5.10) also allows for the state  $x$  to change if its probability  $p_X(x)$  during the measurement does not change. Furthermore, we keep the interaction energy  $E_I$  in the treatment, even though it is usually assumed to be zero before and after the measurement interaction. In many cases the interaction energy can be considered small, especially when dealing with averages. However, when considering single realizations and fluctuation theorems, the influence of the interaction part may become significant.

In the feedback phase, the correlations built are used to extract work, as we already saw in the example of the Szilard engine. We will assume that during the feedback the memory is not altered. That is to say, the free energy of the memory,  $F_Y$ , does not change in the feedback. The entropy generated in the feedback,  $\langle S_{FB} \rangle = -W_{ext} - \Delta_{fb} \mathcal{F}_{X+Y} \geq 0$ , must be positive, meaning that the work extracted is bounded by

$$W_{ext} \leq -\Delta_{fb} \mathcal{F}_{X+Y} = -\Delta_{fb} \mathcal{F}_X - \beta^{-1} \Delta_{fb} \langle I \rangle - \Delta_{fb} \langle E_I \rangle, \quad (5.11)$$

where  $\Delta_{fb}$  denotes the change in the feedback. If the interaction energy is once again neglected, the equation above coincides with the second law of thermodynamics, Eq. (5.6), for feedback. The decrease of mutual information in the feedback,  $\Delta_{fb} \langle I \rangle$  of Eq. (5.11), should be understood as  $\Delta_{fb} \langle I \rangle = -\langle I \rangle$  of Eq. (5.6), where  $\langle I \rangle$  is the amount of mutual information we have about the system.

In order to perform cyclic measurement-feedback operations, we need to restore the memory and the system back to their initial states. This erasure process is associated with a positive generation of entropy  $\langle S_{ER} \rangle = W_{er} - \Delta_{er} \mathcal{F}_{X+Y} \geq 0$ , where  $\Delta_{er}$  denotes the change in the erasure process. The erasure work is then bounded by

$$W_{er} \geq \Delta_{er} \mathcal{F}_{X+Y} = \Delta_{er} \mathcal{F}_X + \Delta_{er} \mathcal{F}_Y + \beta^{-1} \Delta_{er} \langle I \rangle + \Delta_{er} \langle E_I \rangle. \quad (5.12)$$

Since the measurement-feedback-erasure process is cyclic,  $\Delta_m \mathcal{F}_{X+Y} +$

$\Delta_{fb}\mathcal{F}_{X+Y} + \Delta_{er}\mathcal{F}_{X+Y} = 0$ , and thus

$$W_{er} + W_{meas} - W_{ext} \geq 0. \quad (5.13)$$

The equation above states what we already expect, namely that the work needed for the measurement and the erasure processes exceeds the amount of work which can be extracted in the feedback. Thus an information machine working in contact to a single heat reservoir cannot work in a way that would result to a positive net energy gain.

There has been a long debate about what compensates for the work one is able to extract using information, for example in the Szilard engine. Szilard and Brillouin argued that the work is needed for the measurement while Bennett and Landauer argued that the compensating work is needed for the feedback [35, 71–73]. Bennett and Landauer considered the case where the interaction energy  $E_I$  is neglected. Furthermore, they assumed that the memory is symmetric in the sense of an equivalent energy  $E$  for all states, i.e.  $E_Y(y) = E$  for all  $y$ . In addition, they assumed a perfect measurement, where the mutual information obtained in the measurement equals the entropy of the memory,  $\Delta_m\langle I \rangle = \Delta_m\langle S_Y \rangle$ , and therefore the RHS of Eq. (5.10) reduces to zero, i.e.  $W_{meas} \geq 0$ . By constructing a reversible measurement, one could then obtain a situation where no work is needed for the measurement. In this kind of device, the erasure of the memory would be then the compensating part of the process. Furthermore, Landauer and Bennett considered an erasure process of the memory alone. That is to say, they assumed that the memory is not connected to the system during the erasure and thus  $\Delta_{er}\mathcal{F}_{X+Y} = \Delta\mathcal{F}_Y$ . Furthermore, since the memory was assumed symmetric, only the entropic part changes and thus we arrive at the result  $W_{er} \geq k_B T H(Y)$ , which for 2-state (1-bit memory) yields the famous Landauer principle [74]

$$W_{er} \geq k_B T \ln 2, \quad (5.14)$$

which states that the erasure of one bit of information requires a minimum of  $k_B T \ln 2$  of work.

The famous Landauer's principle is obtained in a special case of a symmetric memory and negligible interaction energy between the memory and the system during the erasure. In general it is difficult to say what part of the total process compensates for the extracted work. If the interaction energies are taken into account, the laws of thermodynamics do not immanently forbid a process, where the lower bound of  $W_{er}$  in Eq. (5.12)

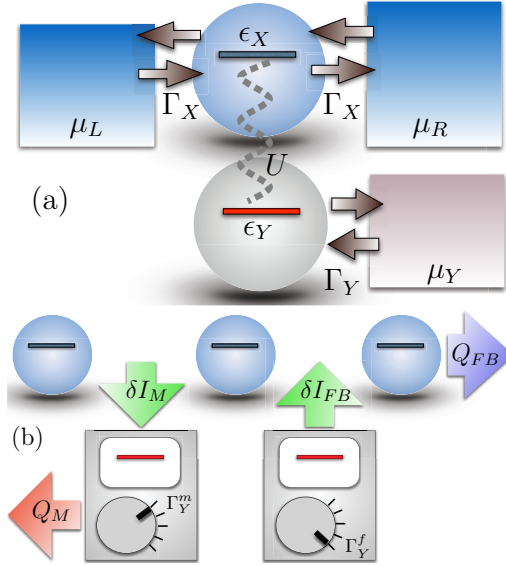
is zero. In this case, there could be a reversible erasure process, which requires no work. In this kind of setup, the work extracted in the feedback would be compensated in some other part of the process.

In Publication III, we study a physical model system, demonstrated and explained in Fig 5.2. The setup is based on coupled quantum dots, where the measurement and feedback are performed by tuning the coupling strength between the measurement dot and its reservoir. Because the marginal distributions  $p_X$  and  $p_Y$  are symmetric in the setup,  $p_X(x) = p_Y(y) = 1/2$  for all  $x$  and  $y$ , the free energies of the system and the measurement dots do not change in the measurement and feedback, i.e.  $\Delta_m \mathcal{F}_Y = \Delta_{fb} \mathcal{F}_X = 0$ . Thus the sole effect of the measurement is to increase correlation (mutual information) between the dots, which is used in the feedback phase. The feedback phase can restore the setup back to its initial state if the initial and final coupling strengths  $\Gamma^Y$  are set to the same value. In this case there is no distinct erasure phase, but negative entropy production in the system is compensated by positive dissipation in other parts of the total system.

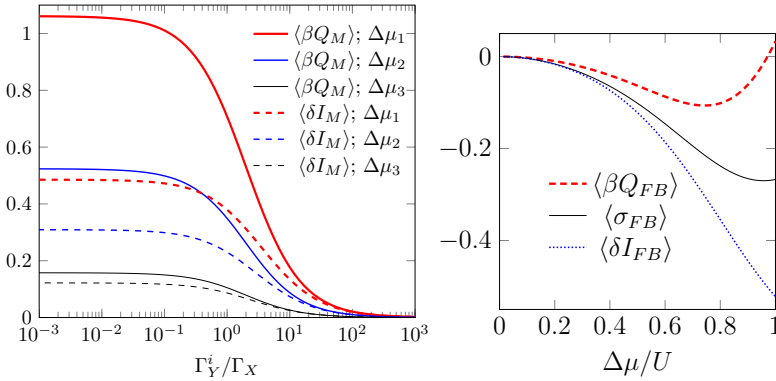
In Publication III and in Fig. 5.3, we show numerical results on the measurement and feedback phases. In the measurement, a positive amount of mutual information is obtained while the second law for measurement,  $\langle S_M \rangle \geq 0$ , is satisfied. In the feedback, both the bare entropy production  $\beta \dot{Q}_{FB}$  and the coarse grained entropy production  $\sigma_{FB}$ , studied in the Publication in detail, are negative. However, the total entropy production is positive and the second law for feedback is satisfied. Furthermore, in Publication III, we show that the integral fluctuation relations, for both the measurement and the feedback entropies,  $\langle e^{-\Delta S_m} \rangle = 1$  and  $\langle e^{-\Delta S_{fb}} \rangle = 1$ , are satisfied in the setup.

### 5.3 Steady state autonomous Maxwell's Demon

In the previous section, we studied correlations between two subsystems, which were manipulated by controlling an external parameter. With suitable protocols these setups could be made to function as a Maxwell's demon device. In these setups, the measurement and feedback were controlled and performed separately. In this section, we discuss setups, where the demon acts independently, measuring and performing feedback without any external control. These kind of setups are commonly referred to as autonomous Maxwell's demons [2, 21–23, 26, 37, 75]. Since there is



**Figure 5.2.** (a): The setup of coupled two-level quantum dots, the system dot  $X$  with energy  $\epsilon_X$  and the measurement dot  $Y$  with energy  $\epsilon_Y$ , coupled to each other so that the state  $(x, y) = (1, 1)$  has energy  $\epsilon_X + \epsilon_Y + U$ . State  $x$  of the dot  $X$  can change due to tunneling in or out from the left (L) and right (R) reservoirs, which are in chemical potentials  $\mu_L$  and  $\mu_R = \mu_L + \Delta\mu$ , respectively, and the coupling strengths are fixed to  $\Gamma_X$ . Dot  $Y$  can change electrons with its reservoir at a chemical potential  $\mu_Y$ . (b): In the measurement the coupling strength  $\Gamma_Y$  is increased from  $\Gamma_Y^i$  to  $\Gamma_Y^m$ , increasing mutual information between the dots by  $\delta I_M$  and dissipating  $\beta Q_M \geq \delta I_M$  as heat. In the feedback the coupling strength  $\Gamma_Y^m$  is decreased to  $\Gamma_Y^f$  and  $\delta I_{FB}$  of mutual information is consumed, resulting to negative dissipation  $\beta Q_{FB} < 0$ , in an apparent violation of the second law. Thus the measurement dot can be made to function as a Maxwell's demon.



**Figure 5.3.** On the left: Average dissipation  $\langle \beta Q_M \rangle$  and the change in mutual information  $\langle \delta I_M \rangle$  in the measurement as a function of the initial coupling strength  $\Gamma_Y^i$  with three different biases,  $\Delta\mu_1 = 0.95U$ ,  $\Delta\mu_2 = 0.75U$  and  $\Delta\mu_3 = 0.5U$ , assuming that the post measurement state is thermalized. The mutual information obtained in the measurement is positive,  $\langle \delta I_M \rangle > 0$ , showing that correlations are built. The entropy production through dissipated heat,  $\langle \beta Q_M \rangle$ , is larger than the information obtained, ensuring the positivity of the total entropy production,  $\langle S_M \rangle = \beta \langle Q_M \rangle - \langle \delta I_M \rangle \geq 0$ . On the right: bare entropy  $\langle \beta Q_{FB} \rangle$ , coarse-grained entropy  $\langle \sigma_{FB} \rangle$  and mutual information  $\langle \delta I_{FB} \rangle$  production in the feedback at  $T = 0.1 k_B^{-1}U$ . Since  $\langle \beta Q_{FB} \rangle \geq \langle \sigma_{FB} \rangle \geq \langle \delta I_{FB} \rangle$ , the negative entropy production is bounded by the change in the mutual information  $-\beta^{-1} \langle \delta I_{FB} \rangle$ . Further details are presented in Publication III.

no external control, these setups could be realized by considering either a relaxing or a steady state system. Here we are interested in a device working in steady state non-equilibrium conditions, like the one we consider in Publication IV.

We assume that the system of interest evolves under Markovian dynamics and use stochastic thermodynamics introduced in Chapter 2. The probability distribution of state  $(x_i, y_i)$ ,  $p_i \equiv p(x_i, y_i)$ , follows the master equation

$$\dot{p}_i = - \sum_f J_{i \rightarrow f}, \quad (5.15)$$

where

$$J_{i \rightarrow f} = \omega_{f \leftarrow i} p_i - \omega_{i \leftarrow f} p_f, \quad (5.16)$$

is the current from  $(x_i, y_i)$  to  $(x_f, y_f)$  and  $\omega_{f \leftarrow i}$  is the transition rate from  $(x_i, y_i)$  to  $(x_f, y_f)$ . The state  $x$  is the state of the system and  $y$  is the state of the demon. The total entropy  $S_{\text{tot}}$  can be written as the sum of the total Shannon entropy,  $S = -k_B \sum_i p_i \ln p_i$ , and the entropy flow,  $S_r = \ln \{ \prod \omega_{f \leftarrow i} / \omega_{i \leftarrow f} \}$  [53]. We assume that the system and the demon reservoirs are both at inverse temperature  $\beta$ . If the rates  $\omega$  are local detailed balance connected, the entropy flow is given by  $S_r = \beta Q_T$ , where  $Q_T$  is the heat dissipated by the total combined system.

The entropy flow  $S_r$  is often called the reservoir entropy, due to the connection to the entropy increase of an equilibrium bath ( $\beta Q_T$ ). However, this nomenclature may be misleading since if the rates are not LDB connected, the entropies  $S_r$  and  $\beta Q_T$  differ in general. This will be exactly the case later on, thus we refer to  $S_r$  as the *coarse grained entropy* and to  $\beta Q_T$  as the *bare entropy*.

Standard stochastic thermodynamics approach gives the entropy production rate as

$$\dot{S}_{\text{tot}} = \frac{1}{2} \sum_{i,f} J_{i \rightarrow f} \ln \left( \frac{p_i \omega_{f \leftarrow i}}{p_f \omega_{i \leftarrow f}} \right) = \dot{S} + \dot{S}_r, \quad (5.17)$$

which is always non-negative by its mathematical construction [15,21,54]. As discussed in more detail, for example in Ref. [21], the total entropy production of Eq. (5.17) can be divided into two non-negative contributions: One describes the entropy production in the system

$$\dot{S}_{\text{tot}}^X = \frac{1}{2} \sum_{i,f} J_{i \rightarrow f}^X \ln \left( \frac{p_i \omega_{f \leftarrow i}}{p_f \omega_{i \leftarrow f}} \right), \quad (5.18)$$

and the other entropy production in the demon:

$$\dot{S}_{\text{tot}}^Y = \frac{1}{2} \sum_{i,f} J_{i \rightarrow f}^Y \ln \left( \frac{p_i \omega_{f \leftarrow i}}{p_f \omega_{i \leftarrow f}} \right). \quad (5.19)$$

Terms  $J_{i \rightarrow f}^X = J_{(x_i, y_i) \rightarrow (x_f, y_i)}$  and  $J_{i \rightarrow f}^Y = J_{(x_i, y_i) \rightarrow (x_i, y_f)}$  are the system and demon currents, induced by the transitions in the system and the demon, respectively. Furthermore, we assumed that the dynamics are bipartitive, i.e. the system and the demon states cannot change simultaneously:  $\omega_{i \leftarrow f} = 0$  if  $x_i \neq x_f$  and  $y_i \neq y_f$ .

The system entropy production rate of Eq. (5.18) can be further divided into

$$\begin{aligned} \dot{S}_{\text{tot}}^X &= \frac{1}{2} \sum_{i,f} J_{i \rightarrow f}^X \left\{ \ln \left( \frac{\omega_{f \leftarrow i}}{\omega_{i \leftarrow f}} \right) + \ln \left( \frac{p_{X,i}}{p_{X,f}} \right) - \ln \left( \frac{p(y_i|x_f)}{p(y_i|x_i)} \right) \right\} \\ &= \dot{S}_r^X + \dot{S}^X - \dot{I}^X \geq 0, \end{aligned} \quad (5.20)$$

where  $\dot{S}_r^X$  is the entropy flow from the system,  $\dot{S}^X$  is the change of the system Shannon entropy,  $S^X = -k_B \ln p_X$ , where  $p_X = \sum_y p(x, y)$  is the marginal distribution, and  $\dot{I}^X$  is the change in mutual information  $I$  (Eq. (5.3)) due to tunneling events in the system.

Similarly, the demon entropy production rate splits into

$$\begin{aligned} \dot{S}_{\text{tot}}^Y &= \frac{1}{2} \sum_{i,f} J_{i \rightarrow f}^Y \left\{ \ln \left( \frac{\omega_{f \leftarrow i}}{\omega_{i \leftarrow f}} \right) + \ln \left( \frac{p_{Y,i}}{p_{Y,f}} \right) - \ln \left( \frac{p(x_i|y_f)}{p(x_i|y_i)} \right) \right\} \\ &= \dot{S}_r^Y + \dot{S}^Y - \dot{I}^Y \geq 0, \end{aligned} \quad (5.21)$$



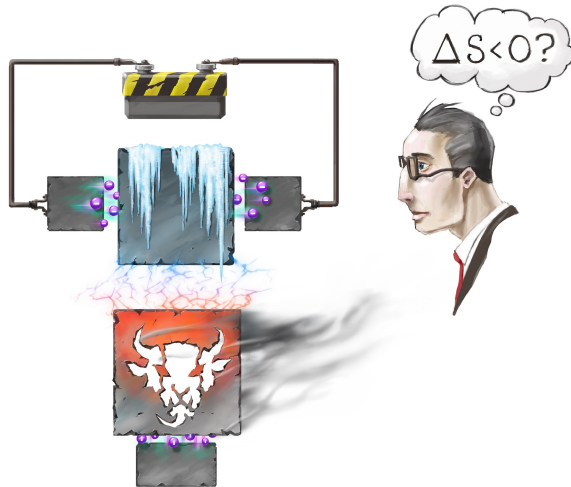
where  $\dot{S}_r^Y$  is the entropy flow from the demon,  $\dot{S}^Y$  is the change of the demon Shannon entropy,  $S^Y = -k_B \ln p_Y$ ,  $\dot{I}^X$  is the change in mutual information  $I$  induced by tunneling events in the demon. Terms  $\dot{I}^Y$  and  $\dot{I}^X$  are the flows of mutual information produced by the system and the demon, respectively, and quantify how much transitions in  $y$  and  $x$  increase the correlation between  $x$  and  $y$  [21, 36]. The entropy flows  $\dot{S}_r^X$  and  $\dot{S}_r^Y$  are given by the dissipated heats,  $\beta\dot{Q}_X$  and  $\beta\dot{Q}_Y$ , from the system and the demon, respectively, if the rates  $\omega$  are local detailed balance connected.

In Publication IV, we study an electronic setup, in which the system is biased by two reservoirs (the left and the right leads) at different chemical potentials. The setup is illustrated in Fig. 5.4. More precisely, a single electron transistor [76] acts as the system to be measured and a single electron box acts as the demon, performing measurements and feedbacks. The state  $x$  may change due to a tunneling event from both the reservoirs, and thus the effective rate at which the state  $x$  changes is the sum of the individual tunneling rates. This effective rate is not detailed balance connected and thus the entropy flow  $\dot{S}_r^X$  of Eq. (5.20) is not given by the dissipated heat  $\beta\dot{Q}_X$ .

The difference between the entropy flow  $\dot{S}_r^X$  (the coarse grained entropy) and the bare entropy production  $\beta\dot{Q}_X$  could be expected in all systems which are biased by a chemical potential difference. If the reservoirs are at different temperatures, i.e. the system is temperature biased, the sum rate will again break the local detailed balance connection, since there is no uniform temperature for the LDB condition. In Publication IV, we show that if the rates  $\omega$  are not local detailed balance connected, the coarse grained entropy  $S_r$  (denoted as  $\sigma^X$  in the Publication) is related to the bare entropy through exponential averaging:  $\langle e^{S_r - \beta Q_X} \rangle = 1$ .

Equations (5.20) and (5.21) describe the entropy production in the system and the demon and the dynamic flow of mutual information between them. This equations could be applied to any bipartitive system evolving under Markovian dynamics. However, here we are interested in steady state dynamics, thus  $\dot{p}_i = 0$  (Eq. (5.15)). In steady state, the joint distribution function  $p(x, y)$  do not change in time and thus  $S^X$  and  $S^Y$  do not change in the process. Furthermore, the total derivative of mutual information vanishes and thus  $\dot{I}^X = -\dot{I}^Y$ . Equations (5.20) and (5.21) are then given by

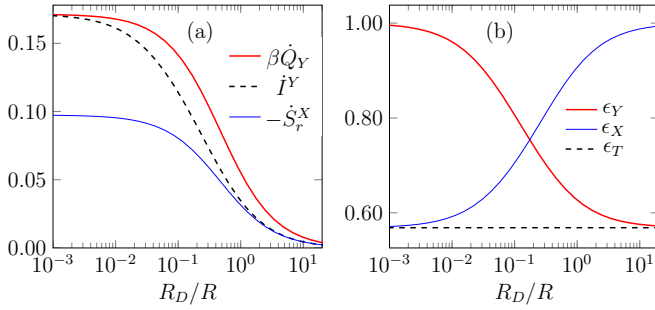
$$\begin{aligned}\dot{S}_{\text{tot}}^X &= \dot{S}_r^X + \dot{I}^Y \geq 0; \\ \dot{S}_{\text{tot}}^Y &= \dot{S}_r^Y - \dot{I}^Y \geq 0.\end{aligned}\tag{5.22}$$



**Figure 5.4.** A schematic picture of a voltage biased single electron transistor (SET) (the upper plate) capacitively coupled to a SEB detector (the lower plate), which acts as the Demon in the setup. Without seeing the Demon, the observer sees the SET system cooling even though the current runs through it. This would be a violation of Joule's law and the second law of thermodynamics. However, the second law is retained by the heat dissipation in the Demon.

In publication IV, we show that by adjusting the tunneling resistance ( $R_D$ ) between the demon and its reservoir, the flow of information  $\dot{I}^Y$  between the system and the demon can be tuned. If the resistance  $R_D$  is made small, corresponding to a fast measurement and feedback, the information flow  $\dot{I}^Y \geq 0$  increases. The entropic cost of sustaining the flow of information is the dissipation of heat  $\dot{Q}_Y$  (denoted as  $\dot{Q}_D$  in the Publication) in the demon. Therefore, the efficiency  $\epsilon_Y = \dot{I}^Y / \beta \dot{Q}_Y$  characterizes the demon's ability to produce information. Figure 5.5 shows that a lower resistance  $R_D$ , corresponding to a faster demon, results to a larger dissipation  $\dot{Q}_Y$  and flow of information  $\dot{I}^Y$  as well as to a higher efficiency  $\epsilon_D$ . If the demon resistance is made so low that the demon thermalizes, the information flow coincides with the dissipation  $\dot{Q}_Y$ , corresponding to the maximum efficiency  $\epsilon_Y = 1$ . This result is shown also analytically in Publication IV.

The essential feature for the setup to work as a Maxwell's demon is the ability to produce negative entropy. Within a relatively large parameter range, the system can produce negative coarse grained entropy  $\dot{S}_r^X$  and also physically observable negative entropy in the form of cooling ( $\beta \dot{Q}^X < 0$ ). Furthermore, the system's efficiency to utilize the flow of information,  $\epsilon_X = -\dot{S}_r^X / \dot{I}^Y$ , decreases in increasing the demon speed (increasing  $R_D$ ), as shown in Fig. 5.5. In the publication we show that the efficiency of the



**Figure 5.5.** (a): Entropy production rate in the demon  $\beta\dot{Q}_Y$ , the flow of information  $\dot{I}^Y$ , and the coarse grained entropy production rate  $\dot{S}_r^X$  in the system as a function of the demon tunneling resistance ( $R_D$ ). Smaller resistance makes the demon faster. In agreement with Eq. (5.22), the entropy production in the demon exceeds the flow of information, which in turn is the upper bound for the negative entropy production in the system,  $\beta\dot{Q}_Y \geq \dot{I}^Y \geq -\dot{S}_r^X$ . (b): The efficiency of information production,  $\epsilon_Y$ , its utilization,  $\epsilon_X$ , and that of the whole production-utilization,  $\epsilon_T$ . In the fast Demon limit ( $R_D \ll R$ ), the flow of information in the Demon equals the heat dissipation rate ( $\epsilon_Y = 1$ ), while in the slow limit the utilization of information flow becomes efficient ( $\epsilon_X = 1$ ). Parameters in both (a) and (b) are those optimal for the maximum cooling power,  $T = 0.08\kappa k_B^{-1}$  and  $eV/\kappa = 0.72$ , discussed in Publication IV

whole measurement-feedback -cycle is given by

$$\epsilon_T = 2/(\beta\kappa)s_r^X, \quad (5.23)$$

where  $\kappa$  is the coupling energy between the system and the demon and  $s_r^X = \ln[\omega_{(1,0) \leftarrow (0,0)}/\omega_{(0,0) \leftarrow (1,0)}]$  is the coarse grained entropy production in the relaxation from state  $(0, 0)$  to  $(1, 0)$  or equivalently from  $(1, 1)$  to  $(0, 1)$ . The efficiency of the whole measurement-feedback -cycle is thus independent of the demon speed, that is to say that  $\epsilon_T$  does not depend on the resistance  $R_D$ . Therefore, the demon's ability to produce information more efficiently trades off to a decrease in the utilization of the information.

We note that the operation of the demon could have been studied using the notion of an information reservoir, as done for example in Refs. [22, 27–29, 37]. In this section the demon, which plays the role of information reservoir for the system, was autonomous. For these kinds of setups, the framework which we used, based on dynamic flow of mutual information, is more transparent [21, 36]. In the previous section, we wanted to focus in the thermodynamics of the measurement and feedback, which are better defined by considering the thermodynamic splitting of the total system into subsystems. In both sections, the information reservoir point of view would have changed some interpretations, however the important physics would have remained the same. The framework we used based on entropic flows and mutual information and the notion of information reservoirs are

compared for example in Refs. [27–29].



## 6. Summary and conclusions

The main focus of this work has been correlations in non-equilibrium thermodynamics of small systems. In particular, we have studied systems with non-Markovian environments and information exchange in stochastic thermodynamics. In Chapter 4, we extended the framework of stochastic thermodynamics to non-Markovian dynamics, where the system and the environment evolution are correlated. In Chapter 5, we formulated influence of correlations between two systems coupled to each other and to a thermal environment using information thermodynamics. We have studied systems, in which the correlations were built using external parameters as well as systems which were correlated to each other at all times and evolving under steady state non-equilibrium conditions.

In Publication I, we studied the dissipated work  $W_d = W - \Delta F$  using a single electron box, which was manipulated externally by controlling the bias voltage of the box. Due to the small heat capacity of the island, the energy dissipated by the tunneling electrons leads to temperature fluctuations during the drive. This overheating effect is a stochastic process and not controlled by an external control parameter. By performing numerical simulations and expanding the tunneling rates in temperature deviation from the equilibrium temperature, we have shown that the work fluctuation theory  $\langle \exp[-\beta W_d] \rangle = 1$  is not satisfied in the setup, since the environment is not described by an ideal heat bath during the drive. However, the integral fluctuation relation for the total entropy production,  $\langle \exp[-S_T] \rangle = 1$  is satisfied. We concluded that there exists additional entropy production, which is not given by the dissipated work  $W_d$ , even if the process starts from equilibrium.

In Publication II, we studied entropy production under non-Markovian dynamics, in which the system and the environmental degrees of freedom are correlated during the process. By using a simplified model, which is

based on separation of time scales, we identified the sources of additional entropy production caused by the non-equilibrium excitations in the environment. The additional non-Markovian entropy production was studied in the operation of the overheating single electron box, in which the relaxation time arguments can be realized. We showed analytically that the additional entropy terms need to be included into the entropy production in order to obtain the full entropy production which satisfies the integral fluctuation relation. These additional terms require measurements of the environmental degrees of freedom.

In Publication III we studied a setup of two coupled quantum dots, where the correlation between the dots can be adjusted by changing the coupling strength  $\Gamma_Y$  between the measurement dot and its reservoir. By increasing the coupling strength  $\Gamma_Y$ , we can measure the state of the system dot with the measurement dot. We studied analytically and numerically information gain and dissipation in the measurement. We have also shown that the same system can work as a Maxwell's demon setup, where the information obtained in the measurement is utilized resulting to negative dissipation. The setup contains both the microscopic system and the demon, and the measurement and feedback are performed separately, as in the case of the Szilard engine.

In Publication IV, we studied an autonomous Maxwell's demon setup, in which a capacitively coupled single electron box, the demon of the setup, measures and performs feedback on a single electron transistor. The measurement and feedback are performed independently by the demon itself, without any external control. The dynamic flow of information in this non-equilibrium steady state setup can be formalized using information thermodynamics. By changing the speed of the demon (tunneling resistance between the single electron box and its reservoir), the amount of information produced and utilized, as well as their efficiency can be tuned. In this way the properties of the demon can be used to control dissipation in the setup.

In summary, stochastic thermodynamics and information thermodynamics were studied analytically and numerically using physically feasible model systems. The results presented in this thesis help to understand the correlations in microscopic dynamics, the microscopic origin of thermodynamics, and especially the origin of entropy production. Furthermore, the results provide a step towards more accurate modeling, understanding and design of small scale devices, where dissipation and fluctu-

ations play a major role.

Interesting research directions for the future include the setup of Publication IV in a quantum regime, where quantum effects create additional correlations. In addition, the coupled quantum dot system could be used to study non-Markovian dynamics, including different definitions of non-Markovianity, using a setup in which the memory of dynamics can be tuned. Work in these directions is in progress.





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Stochastic thermodynamics is a modern theoretical framework which extends the traditional thermodynamics to non-equilibrium systems and processes, and is particularly useful in studying systems at microscopic scales. Information thermodynamics combines the developments in stochastic thermodynamics and information theory. It is used to study thermodynamics of measurement and feedback operations. In this thesis we study both stochastic and information thermodynamics using physically feasible model systems, based on single electron tunneling at low temperatures. The results provide a step towards more accurate modeling, understanding, and design of small scale devices, where dissipation and fluctuations play a major role.



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