Steady state fluctuations of the dissipated heat for a quantum stochastic model

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Abstract: We introduce a quantum stochastic dynamics for heat conduction. A multi-level subsystem is coupled to reservoirs at different temperatures. Energy quanta are detected in the reservoirs allowing the study of steady state fluctuations of the entropy production. Our main result states a symmetry in its large deviation rate function.

KEY WORDS: entropy production, fluctuation theorem, quantum stochastic calculus

1 Introduction

Steady state statistical mechanics wants to construct and to characterize the stationary distribution of a subsystem in contact with several reservoirs. By nature the required scenario is an idealization as some essential specifications of the reservoirs must be kept constant. For example, intensive quantities such as temperature or (electro-)chemical potential of the different reservoirs are defined and unchanged for an extensive amount of time, ideally ad infinitum. Reservoirs do not interact directly with each other but only via the subsystem; they remain at their same spatial location and can be identified at all times. That does not mean that nothing happens to the reservoirs; flows of energy or matter reach them and they are like sinks and sources of currents that flow through the subsystem. Concrete realizations and models of steady states vary widely depending on the type of substances and on the nature of the driving mechanism.

An old and standard problem takes the subsystem as a solid in contact at its ends with two heat reservoirs and wants to investigate properties of the energy flow. Beloved by many is a classical model consisting of a chain or an array of coupled anharmonic oscillators connected to thermal noises at the boundaries. The reservoirs are there effectively modeled by Langevin forces while the bulk of the subsystem undergoes a Hamiltonian dynamics, see e.g. [RT, EPR, MNV]. Our model to be specified below is a quantum analogue of that scenario in the sense that we also consider a combination of Hamiltonian

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dynamics and Markovian thermal forces.
We imagine a chain of coupled two(or multi)-level systems. The dynamics of the isolated subsystem is unitary with Hamiltonian H_S. Quanta of energy ω are associated to the elementary transitions between energy levels. Two physical reservoirs at inverse temperatures $\beta_k, k = 1, 2$, are now attached to the subsystem. The total dynamics is described by a quantum stochastic differential equation through which we can observe the number $N_{\omega,k}$ of quanta with energy ω that are piled up in the $k-$th reservoir. The total energy $N \equiv H_S + \sum_{\omega,k} \omega N_{\omega,k}$ is conserved under the dynamics (Proposition 2.4). The change in the second term corresponds to the flow of energy quanta in and out of the reservoirs and specifies the dissipated heat. Our main result is obtaining a symmetry in the fluctuations of that dissipated heat that extends the so called steady state fluctuation theorem to a quantum regime (Proposition 2.5).
The quantum stochastic evolution that defines the model is a particular dilation of a semigroup dynamics that describes the weak coupling regime of our subsystem coupled to quasi-free boson fields. The dilation, a sort of quantum Langevin equation, is much richer and enables the introduction of a natural path space measure. One should remember here that a major conceptual difficulty in coming to terms with the notion of a variable entropy production for quantum steady states is to understand its path-dependence. One option is to interrupt the unitary dynamics with collapses, see e.g. [CDJMN]. Others have proposed an entropy production operator, avoiding the problem of path-dependence. Our set-up follows a procedure that is well-known in quantum optics with thermal noises formally replacing photon detectors, see [Bo, BMK]. In the resulting picture we record each energy quantum that is transferred between subsystem and reservoirs. It induces a stochastic process on quanta transferrals and there remains no problem to interpret the fluctuations of the entropy production. From the mathematical point of view, the model can be analyzed via standard probabilistic techniques.

1.1 Related Results

In the past decade, a lot of interest has been going to the Gallavotti-Cohen fluctuation relation [GC, ECM], see [M2] for more recent references. In its simplest form that relation states that the steady state probability of observing a total entropy decrease $w_T = -wT$ in a time T, is exponentially damped with respect to the probability of observing an increase of w_T as

$$\frac{\boldsymbol{Prob}(w_T = wT)}{\boldsymbol{Prob}(w_T = -wT)} \approx e^{+wT} \tag{1.1}$$

at least for very large time spans T. The relation (1.1) is known as the steady state fluctuation theorem (SSFT) and states a symmetry in the fluctuations of the entropy dissipation in a stationary nonequilibrium steady state. The symmetry was first discovered in the context of dynamical systems and was applied to the phase space contraction rate in strongly chaotic dynamical systems, see [R2, ECM, GC]. There is also a transient version of that symmetry, sometimes exactly verified for finite T and known as the transient fluctuation theorem (TFT), see [SEC]. The basic underlying mechanism and general unifying principles connecting SSFT and TFT with statistical mechanical entropy have been explained in [MN, M2].

In the present paper we derive a quantum steady state fluctuation theorem. Monnai and Tasaki [MT] have investigated an exactly solvable harmonic system and found quantum corrections to both SSFT and TFT. Matsui and Tasaki [MaT] prove a quantum TFT in a general C^*-algebraic setting. It is however unclear what is the meaning of their entropy production operator. A related quantum Jarzynski relation was studied in [DM].

Besides the fluctuation theorem, we also describe a new approach to the study of heat conduction in the quantum weak coupling limit. In [LS2] Lebowitz and Spohn studied the thermodynamics of the weak-coupling generator. They identified the mean currents, and they proved a Green-Kubo relation. At that time it was however impossible to conclude that these expressions are the first non-zero contributions to their counterparts at finite coupling λ. That has recently been shown in a series of papers by Jaksic and Pillet [JP1, JP2, JP3, JP4], who used spectral techniques to study the system at finite coupling λ. It was also shown that the stationary state of the weak coupling generator is the zeroth order contribution to the system part of the so called NESS, the natural nonequilibrium steady state. The current fluctuations we define in our model, agree with the expressions of [LS2] as far as the mean currents and the Green-Kubo formula is concerned. Our entropy production operator is however new, it differs for example from the proposal of [MaT].

The approach taken here also differs from the more standard route that has been followed and that was outlined by Ruelle in [R1]. Recently and within that approach and context of heat conduction, new results have been obtained in [AS, JP3, JP4, AbF]. To us it remains however very much unclear how to define and study in that scenario a fluctuating entropy; in contrast, that is exactly one of the things we can easily achieve via our approach but we remain in the weak coupling limit.

1.2 Basic strategies

1.2.1 Microscopic approach

In general one would like to start from a microscopic quantum dynamics. The system is then represented by a finite dimensional Hilbert space $\mathcal{H}$ and system Hamiltonian H_S. The environment is made from m thermal reservoirs, infinitely extended quantum mechanical systems, with environment Hamiltonian, formally,

$$H_R \equiv H_1^R + \ldots + H_m^R$$

The coupling between system and reservoirs is local and via some bounded interaction term λH^{SR} so that the total Hamiltonian takes the form

$$H_\lambda \equiv H_S \otimes 1 + 1 \otimes H_R + \lambda \sum_{k=1}^{m} V_k \otimes R_k$$

where we have already inserted a specific form for the coupling H^{SR} using self-adjoint reservoir operators R_k and V_k acting on $\mathcal{H}$. On the same formal level, which can however easily be made precise, the total quantum dynamics is then just

$$U_t^\lambda \equiv e^{-iH_\lambda t} \cdot e^{+iH_\lambda t}$$

We will not follow the beautiful spectral or scattering approach that has recently been exploited for that nonequilibrium problem. We refer the reader to the specialized references such as [JP3, R1, AS] and we only outline the main steps totally ignoring essential assumptions and technicalities:
One starts the dynamics from an initial state

$$\omega \equiv \mathrm{tr} \otimes \omega^1 \otimes \ldots \otimes \omega^m$$

where tr stands for the normalized trace-state in the system and the ω^k are equilibrium KMS states at inverse temperature β_k for the k-th reservoir, $k = 1, \ldots, m$. The quantum dynamics takes that initial state to the new (now coupled) state ω_t at time $t > 0$. The NESS is obtained via an ergodic average

$$\bar{\rho} \equiv \lim_{T \uparrow +\infty} \frac{1}{T} \int_0^T dt\, \omega_t$$

One of the first questions (and partially solved elsewhere, see e.g. [JP3, R1, AS]) is then to derive the natural conditions under which the mean entropy production rate

$$\dot{S} \equiv \sum_{k=1}^{m} \beta_k\, \bar{\rho}(i[H_\lambda, H_k^R])$$

is strictly positive. While that mean entropy production certainly coincides with conventional wisdom, we do not however believe that the operator

$$i[H_\lambda, H_k^R]$$

or equivalent expressions, is the physically correct candidate for the study of current fluctuations which would obey the SSFT. That is not even the case for the simplest (classical) stochastic dynamics; one needs to go to path-space and study current fluctuations in terms of (fluctuating) trajectories.

1.2.2 Weak coupling approach

Starting from the microscopic dynamics above, we can of course always look at the reduced dynamics Λ_t^λ on the system

$$\Lambda_t^\lambda \rho = \mathrm{Tr}_R[U_t^\lambda(\rho \otimes \omega^1 \otimes \ldots \otimes \omega^m)]$$

for a density matrix ρ on the system. Obviously, the microscopic evolution couples the system with the environment and the product form of the state will in general not be preserved. One can however attempt a Boltzmann-type Ansatz or projection technique to enforce a repeated randomization. That can be made rigorous in the so called weak coupling limit. For that one needs the interaction picture and one keeps $\lambda^2 t = \tau$ fixed. That is the Van Hove-Davies-limit [H, D2]:

$$\lim_{\lambda \to 0} \Lambda_t^0 \, \Lambda_t^\lambda \rho \equiv e^{\tau L} \rho$$

where L is a linear operator acting on density matrices for the system. The generator will be written out more explicitly in Section 2.1 but its dual $\mathcal{L}$ acting on $\mathcal{B}(\mathcal{H})$ is of the form, see (2.5),

$$\mathcal{L}(\cdot) = -i[H_f, \cdot] + \sum_k \mathcal{L}_k(\cdot)$$

where the $\mathcal{L}_k$ can be identified with the contribution to the dissipation from the kth reservoir. H_f is an effective, renormalized Hamiltonian depending on details of the reservoirs and the coupling.
From now on we write ρ for the (assumed) unique invariant state (see [LS2] for sufficient conditions to have a unique invariant state):

$$e^{\tau L} \rho = \rho, \quad \tau \geq 0$$

Again one can study here the mean entropy production, as for example done in [LS2] and argue that

$$\mathrm{Tr}[\rho\,\mathcal{L}_k(H^S)]$$

represents the stationary heat flow into the k'th reservoir, at least in the weak coupling regime. Nothing tells us here however about the physical fluctuations in the heat current for which higher moments should be considered. In fact, the reservoirs are no longer visible as the weak coupling dynamics is really a jump process on the energy levels of the system Hamiltonian, see further in Section 3.1. The heat flow and the energy changes in the individual reservoirs cannot be reconstructed from the changes in the system. The present paper will use a new idea for the study of the fluctuations of the heat dissipation in a reservoir.

1.2.3 Dilation

While the weak coupling dynamics is very useful for problems of thermal relaxation (one reservoir) and for identifying the conditions of microscopic reversibility (detailed balance) characterizing an equilibrium dynamics, not sufficient information is left in the weak coupling limit to identify the variable heat dissipated in the various reservoirs. Heat is path-dependent and we need at least a notion of energy-trajectories.[3] The good news is that we can obtain such a representation at the same time as we obtain a particular dilation of the weak coupling dynamics. The representation is basically achieved via an unravelling of the weak coupling generator $\mathcal{L}$ and the corresponding Dyson-expansion of the semigroup dynamics. That will be explained under 2.2.

There are many possible dilations of a quantum dissipation. It turns out that there is a dilation whose restriction to the system coincides with the Dyson-representation in terms of energy-trajectories of the weak coupling dynamics. That dilation is well studied and goes under the name of a quantum stochastic dynamics. The associated quantum stochastic calculus was invented by Hudson and Parathasaraty, [HP, P]. It has been extensively employed for the purpose of quantum counting processes, see e.g. [BMK, BGM]. Various representations and simplifications have been added, such as in [AF] where a (classical) Brownian motion extends the weak coupling dynamics.

[3] At least, if one has a stochastic or effective description of the system dynamics, as is the case in the weak-coupling limit. We do **not** claim at all that the trajectory-picture is microscopically fundamental.

1.2.4 Results

We prove a symmetry in the large deviation generating function (Proposition 2.5). In the case of non-degenerated free subsystem Hamiltonians H_S, this rate function is analytic and this implies the large deviation principle. The symmetry is recongnized as the fluctuation theorem for the entropy production. For degenerate Hamiltonians, we can only establish analyticity in a neighbourhood of 0, this implies the central limit theorem for the currents (but we do not stress this point). Here the symmetry does not translate in a standard fluctuation theorem. However, in both cases, the symmetry implies a Green-Kubo relation and (modified) Onsager reciprocity (Proposition 2.7).

1.3 Outline of the paper

In section 2, we introduce the quantum stochastic model and state the result. In section 3 follows a discussion where the main points and novelties are emphasized. Proofs are postponed to section 4.

2 The Model

2.1 Weak Coupling

We briefly introduce here the weak-coupling dynamics without speaking about its derivation, which is not relevant for the discussion here. Some of that was briefly adressed in Sections 1.2.1-1.2.2.
Let $\mathcal{H}$ be a d-dimensional Hilbert space assigned to a small subsystem, called system in what follows. Let H_S be a Hamiltonian on $\mathcal{H}$ with eigenvalues $\epsilon_x, \ldots, \epsilon_y$ labelled via $x, y \in \Lambda$, some finite set and spectral projections $P_x, P_y, \ldots$.

$$F \equiv \{\omega \in \mathbb{R} | \quad \exists x, y \in \Lambda : \omega = \epsilon_x - \epsilon_y\} \tag{2.1}$$

is the set of eigenvalues of the derivation $-i[H_S, \cdot]$. We label by $k = 1, \ldots, m$ different heat reservoirs at inverse temperatures β_k. To each reservoir k is assigned a self-adjoint operator V_k[4] on $\mathcal{H}$ and for each $\omega \in F$, we put

$$V_{\omega,k} = e^{\frac{\beta_k \omega}{4}} \psi_{\omega,k} P_y V_k P_x, \qquad \epsilon_x - \epsilon_y = \omega \tag{2.2}$$

with $\psi_{\omega,k} \in \mathbb{C}$ arbitrary, but obeying the constraint $\bar{\psi}_{\omega,k} = \psi_{-\omega,k}$.
We further consider an effective self-adjoint Hamiltonian H_f satisfying

$$[H_f, H_S] = 0 \tag{2.3}$$

[4] One can take more couplings per reservoir, and they need not be self-adjoint, but for notational clarity, we restrict to that case.

We work with the following weak coupling generator $\mathcal{L}$ on $\mathcal{B}(\mathcal{H})$

$$\mathcal{L}(\cdot) = -i[H_f, \cdot] + \sum_{\omega,k} \left(V^*_{\omega,k} \cdot V_{\omega,k} - \frac{1}{2}\{V^*_{\omega,k}V_{\omega,k}, \cdot\}\right) \tag{2.4}$$

By grouping all terms with the same k, we can write

$$\mathcal{L}(\cdot) = -i[H_f, \cdot] + \sum_k \mathcal{L}_k(\cdot) \tag{2.5}$$

For $\mathcal{L}$ to be really the generator derived in [D1], we would need to relate the factors $\phi_{\omega,k}$ to correlation functions in the reservoir and we should fix the renormalized hamiltonian H_f. However, since this is not necessary for our discussion, we omit these specifications.
We will need the following assumption **A1**
There is an open set $\mathcal{V} \subset \mathbb{R}^m$ such that for all $\overline{\beta} = (\beta_1, \dots, \beta_m) \in \mathcal{V}$, the generator $\mathcal{L} \equiv \mathcal{L}_{\overline{\beta}}$ has a unique invariant steady state $\rho = \rho_{\overline{\beta}}$. This implies that 0 is a non-degenerate eigenvalue (corresponding to $\mathbb{1}$) of $\mathcal{L}_{\overline{\beta}}$. Moreover, none of the $V_{\omega,k}$ should be zero [5].
Remark that if $\overline{\beta} = (\beta, \dots, \beta)$, then $\rho_{\overline{\beta}} = \exp[-\beta H_S]/\mathrm{tr}(\exp[-\beta H_S]) \equiv \rho_\beta$
Furthermore, one can easily see that

$$\sum_{x \in \Lambda} P_x \rho P_x = \rho \tag{2.6}$$

2.2 Unravelling of the generator

We associate to that semigroup dynamics a path space measure by a procedure which is known as "unravelling of the generator."
Write the weak coupling generator as

$$\mathcal{L} = \mathcal{L}_0 + \sum_{\omega,k} \mathcal{J}_{\omega,k}$$

with

$$\mathcal{J}_{\omega,k}(\cdot) = V^*_{\omega,k} \cdot V_{\omega,k} \tag{2.7}$$

and

$$\mathcal{L}_0(\cdot) = -i[H_f, \cdot] - \frac{1}{2}\sum_{\omega,k}\{V^*_{\omega,k}V_{\omega,k}, \cdot\})$$

Let $\Omega^1_{t,n}$ be the set of ordered n-tupels $\{(t_1, \dots, t_n) \big| t_1 \le t_2 \le \dots \le t_n \le t\}$. Put $\Omega^1_t = \bigcup_{n \in \mathbb{N}} \Omega^1_{t,n}$. One can make Ω^1_t into a measure space $(\Omega^1_t, d\sigma)$ by

[5] This ensures also that every part of the system is effectively coupled to all reservoirs.

embedding $\Omega^1_{t,n}$ in $([0,t]^n, \mathcal{A}^1_t, dx)$ with dx the Lesbegue measure and $\mathcal{A}^1_t$ the σ-algebra. In the same way, one defines $\Omega^1 \equiv \Omega^1_{t=+\infty}$ and $\mathcal{A}^1 \equiv \mathcal{A}^1_{t=+\infty}$. Finally let $\Omega \equiv (\Omega^1)^\ell = \prod_{k,\omega} \Omega^1_{k,\omega}$ with $\Omega^1_{k,\omega}$ denoting identical copies of Ω^1, similarly we introduce $\mathcal{A} \equiv (\mathcal{A}^1)^\ell$ and $\Omega_t, \mathcal{A}_t$ as products of $\Omega^1_t, \mathcal{A}^1_t$. Often, Ω is called the Guichardet space, see [G].
An element $\sigma \in \Omega$ looks as

$$\sigma = (\omega_1, k_1, t_1; \ldots; \omega_n, k_n, t_n) \text{ with } n \in \mathbb{N} \text{ and } (t_1 \leq t_2 \leq \ldots \leq t_n) \tag{2.8}$$

For future use, we also define 'number functions' $\overline{n}_t \in \mathbb{N}^\ell$ with components

$$n_{\omega,k,t}(\sigma) \equiv |\sigma \cap \Omega_{\omega,k,t}| \tag{2.9}$$

The notation $\Omega_{\omega,k,t}$ should be understood as Ω^1_t where $\Omega_{\omega,k}$ is an indexed copy of Ω^1.
Consider $\mathcal{W}_t(\sigma) : \mathcal{B}(\mathcal{H}) \to \mathcal{B}(\mathcal{H})$ as the completely positive map depending on $\sigma \in \Omega$

$$\mathcal{W}_t(\sigma) = I_t(\sigma) e^{(t_1)\mathcal{L}_0} \mathcal{J}_{\omega_1,k_1} e^{(t_2-t_1)\mathcal{L}_0} \ldots e^{(t_n-t_{n-1})\mathcal{L}_0} \mathcal{J}_{\omega_n,k_n} e^{(t-t_n)\mathcal{L}_0} \tag{2.10}$$

with $I_t(\cdot)$ the indicator function of $\Omega_t \subset \Omega$ and with the indices $(\omega_i, k_i), i = 1, \ldots, n$ referring to the representation (2.8) of σ. Complete positivity follows from (2.7) and from

$$e^{t\mathcal{L}_0} = e^{-itH_f - \frac{t}{2} V_{\omega,k} V_{\omega,k}} \cdot e^{+itH_f - \frac{t}{2} V_{\omega,k} V_{\omega,k}}$$

We can now write

$$e^{t\mathcal{L}} = \int_\Omega d\sigma \mathcal{W}_t(\sigma) \tag{2.11}$$

which is a Dyson expansion. That expression induces a 'path space measure', or a notion of 'quantum trajectories'.

Lemma 2.1. *Define*

$$\mathbb{P}_{\rho,t}(E) \equiv \int_E d\sigma \quad \rho(\mathcal{W}_t(\sigma)1), \quad E \in \mathcal{A} \tag{2.12}$$

Then $\mathbb{P}_{\rho,t}$ are a consistent family of probability measures on Ω_t and thus we obtain a measure $\mathbb{P}_\rho$ on Ω.

These probability measures are often called 'quantum counting processes' (see [BGM]).

2.2.1 Result

We define the entropy current $\mathbf{w}_t$ as a function on Ω:

$$\mathbf{w}_t(\sigma) = \sum_{\omega,k} \beta_k \omega n_{\omega,k,t}(\sigma) \tag{2.13}$$

with $n_{\omega,k,t}$ as in (2.9). Now we can already formulate the main result of the paper:

Proposition 2.2. *Assume (**A1**). Let $\mathbf{w}_t$ be defined in (2.13). There is a neighborhood $\mathcal{U}$ of 0 in $\mathbb{R}$, such that for all $\kappa \in \mathcal{U}$, the limit*

$$e(\kappa) \equiv \lim_{t\to+\infty} \frac{1}{t} \log \int d\mathbb{P}_\rho e^{-\kappa \mathbf{w}_t} \tag{2.14}$$

exists and is real-analytic.
Write $e(\kappa) \equiv e(\kappa, H_f)$, indicating explicitly the dependence on the effective Hamiltonian H_f. Then

$$e(\kappa, H_f) = e(1-\kappa, -H_f) \tag{2.15}$$

whenever the left-hand side exists.
The mean entropy production is non-negative

$$\int d\mathbb{P}_\rho \mathbf{w}_t \geq 0 \tag{2.16}$$

Let now H_S be non-degenerate, then (2.14) exists and is real-analytic for all κ and $e(\kappa, H_f) = e(\kappa, -H_f)$ and (2.14) verifies

$$e(\kappa) = e(1-\kappa) \tag{2.17}$$

for all $\kappa \in \mathbb{R}$
For non-degenerate H_S, the mean entropy production is strictly positive

$$\int d\mathbb{P}_\rho \mathbf{w}_t > 0 \tag{2.18}$$

whenever there are k, l such that $\beta_k \neq \beta_l$

2.3 QSDE

We will not prove nor even mention all technicalities in this subsection; recommended references are [P] for Quantum Stochastic Calculus and [F] for Canonical Commutation Relations.
Let $\ell \equiv |F| \times m$ (recall that m is the number of reservoirs) and $\mathcal{G} \equiv (L^2(\mathbb{R}))^\ell$. An element $f = (f_1, \dots, f_\ell)$ of $\mathcal{G}$ consists of ℓ components which are labelled by the double index (ω, k). Let $\chi_t \in L^2(\mathbb{R})$ be the indicator function of the interval $[0,t]$ and

$$\chi_{\omega,k,t} = (0, \dots, 0, \underbrace{\chi_t}_{\omega,k}, 0, \dots, 0)$$

Let $\mathbb{1}_{[0,t]} \in \mathcal{B}(L^2(\mathbb{R}))$ be the projector on $L^2([0,t])$ and

$$\mathbb{1}_{\omega,k,t} = (0, \dots, 0, \underbrace{\mathbb{1}_{[0,t]}}_{\omega,k}, 0, \dots, 0) \tag{2.19}$$

We consider the bosonic Fock space (Γ_s denotes symmetrized second quantization)

$$\mathcal{F} = \Gamma_s(\mathcal{G}) = \otimes^\ell \Gamma_s(L^2(\mathbb{R})) \tag{2.20}$$

with the factorization property for $s < t$

$$\mathcal{F} = \mathcal{F}_{s]} \otimes \mathcal{F}_{[s,t]} \otimes \mathcal{F}_{[t} \qquad \mathcal{F}_{[s,t]} \equiv \Gamma_s[(L^2[s,t])^\ell] \tag{2.21}$$

The operators $\mathbf{A}_{k,\omega,t}$ and $\mathbf{A}^*_{k,\omega,t}$ on $\mathcal{F}$ are the usual annihilator and creator of the test function $\chi_{\omega,k,t}$. We omit here the precise definitions which are well-known since we will only need some common wisdom about the solution of (2.22).
Let $\mathcal{B}(\mathcal{H} \otimes \mathcal{F})$ be the algebra of bounded operators on $\mathcal{H} \otimes \mathcal{F}$. That suffices to give meaning to the following Quantum Stochastic Differential Equation (QSDE) on $\mathcal{B}(\mathcal{H} \otimes \mathcal{F})$, see [HP],

$$\begin{aligned} d\mathbf{U}_t &= \sum_{\omega,k} \big(V_{\omega,k} d\mathbf{A}^*_{\omega,k,t} - V^*_{\omega,k} d\mathbf{A}_{\omega,k,t}\big)\mathbf{U}_t \\ &- (iH_f dt + \frac{1}{2}\sum_{\omega,k} V^*_{\omega,k} V_{\omega,k} dt)\mathbf{U}_t, \quad \mathbf{U}_0 = \mathbb{1} \end{aligned} \tag{2.22}$$

The solution to (2.22) is unitary. Let Y_t be the unitary on $\mathcal{G}$ which implements the left shift

$$(Y_t(f))(s) = f(s+t) \tag{2.23}$$

then we obtain a strongly continuous unitary group $\widehat{\mathbf{U}}_t$ by putting

$$\widehat{\mathbf{U}}_t = \begin{cases} d\Gamma_s(Y_t)\mathbf{U}_t & \text{if } t \geq 0 \\ \mathbf{U}^{-1}_{-t} d\Gamma_s(Y_t) & \text{if } t < 0 \end{cases} \tag{2.24}$$

with $\mathbf{U}_t$ the solution of (2.22).
Equation (2.22) is again our nonequilibrium model describing several heat baths at different temperatures connected to the subsystem. In Section 4 we motivate the model physically. The connection with Section 2.1 and in particular with the path-space measure (2.12) is the subject of Lemma 2.30. Note that by taking each $V_{\omega,k} = 0$ or $V_k = 0$ in (2.2) the subsystem decouples from the reservoir and (2.22) is solved by

$$\mathbf{U}_t = e^{-itH_f} \otimes \mathbb{1} \tag{2.25}$$

Remark that in (2.22), the reservoirs are now not only labelled by k, as in the original physical picture, but also by ω; each transition has its own mathematical reservoir.
To formulate our results we also need to specify the state. It is taken of the form

$$\overline{\rho} \equiv \rho \otimes \phi \text{ on } \mathcal{H} \otimes \mathcal{F} \tag{2.26}$$

where ρ is the unique stationary state of the dynamics restricted to the subsystem, see assumption **A1**. The state ϕ denotes the vacuum on all Fock spaces involved: with $\Psi \equiv 1 \oplus 0 \oplus 0 \otimes 0 \oplus \ldots$,

$$\phi(\cdot) = \langle \Psi, \cdot \Psi \rangle \tag{2.27}$$

and, as in (2.21)

$$\Psi = \Psi_{s]} \otimes \Psi_{[s,t]} \otimes \Psi_{[t} \tag{2.28}$$

For $\mathbf{O} \in \mathcal{B}(\mathcal{H} \otimes \mathcal{F})$, write $j_t(\mathbf{O}) \equiv \widehat{\mathbf{U}}_t \mathbf{O} \widehat{\mathbf{U}}_{-t}$ with $\widehat{U}_t$ as in (2.24).
Remark that the state $\overline{\rho}$ is not invariant under the dynamics $\widehat{\mathbf{U}}_{-t} \cdot \widehat{\mathbf{U}}_t$, but it is invariant under the 'free dynamics':
Define $\mathbf{V}_t = e^{itH_S} \otimes d\Gamma_s(Y_t)$, for $\mathbf{O} \in \mathcal{B}(\mathcal{H} \otimes \mathcal{F})$, we have

$$\overline{\rho}(\mathbf{O}) = \overline{\rho}(\mathbf{V}_t \mathbf{O} \mathbf{V}_{-t}) \tag{2.29}$$

and this remains true if we replace H_S by 0. For the reservoir part this is trivial, for the system part it follows from (2.6).
Let $\mathbf{N}_{\omega,k,t}$ be the number operator up to time t on the (ω, k)'th Fock space

$$\mathbf{N}_{\omega,k,t} = d\Gamma_s(\mathbb{1}_{\omega,k,t})$$

defined on a suitable dense domain. The connection of the QSDE with the quantum counting process is provided by the following Lemma, which we will not prove. It can be found for example in [BMK, BGM].

Lemma 2.3. *Let $I_{\omega,k}$ be Borel sets in $\mathbb{R}^+$ and $S_{\omega,k} \subset \mathbb{N}$. Let $E_{\omega,k}$ be the set of ordered subsets $\mathcal{C} \subset I_{\omega,k}$ with $|\mathcal{C}| \in S_{\omega,k}$ and put $E \equiv \prod_{\omega,k} E_{\omega,k} \in \mathcal{A}$. Define the projector $\mathbf{1}_{\omega,k,I_{\omega,k}}$ as in (2.19) and define the spectral projection $\mathbf{P}_{I_{\omega,k}}(S_{\omega,k})$ on $\mathcal{F}$, corresponding to the operator $\Gamma_s[\mathbf{1}_{\omega,k,I_{\omega,k}}]$ and the spectral subset $S_{\omega,k}$. Finally, Define $\mathbf{P}_E \equiv \otimes_{\omega,k}\mathbf{P}_{I_{\omega,k}}(S_{\omega,k})$ and let $A \in \mathcal{B}(\mathcal{H})$. Then*

$$\overline{\rho}(j_t[A \otimes \mathbf{P}_E]) = \int_{\sigma\in E} d\sigma \rho(\mathcal{W}_t(\sigma)(A)) \tag{2.30}$$

In particular, one can take $A = \mathbb{1}$ and $I_{\omega,k} = [0,t]$, then $\Gamma_s[\mathbf{1}_{\omega,k,I_{\omega,k}}] \equiv \mathbf{N}_{\omega,k,t}$ and (2.30) leads to

$$\overline{\rho}(j_t[\mathbf{N}_{\omega,k,t}]) = \sum_{n=0}^{+\infty} n\mathbb{P}_\rho(E_{n,t}) \tag{2.31}$$

with $E_{n,t} = \{\sigma \in \Omega \big| n_{\omega,k,t}(\sigma) = n\}$ and where the left-hand side is to be understood as a limit involving spectral projections (see also the footnote in Proposition 2.5). One can also choose $I_{\omega,k} = [0,+\infty[$ and $N_{\omega,k} = \mathbb{N}$ for all (ω,k) (which leads to $E = \Omega_t$) in (2.30), hence expressing that the QSDE dilates the semigroup

$$\overline{\rho}(j_t[A \otimes \mathbb{1}]) = \rho(e^{t\mathcal{L}}(A)) \tag{2.32}$$

2.4 Results

We define the total energy[6] of the k'th reservoir up to time t

$$\mathbf{N}_{k,t} = \sum_\omega \omega \mathbf{N}_{\omega,k,t} \tag{2.33}$$

and a quantity which we interpret as the total energy of subsystem+reservoirs

$$\mathbf{N}_t \equiv H_S + \sum_{\omega,k} \omega \mathbf{N}_{\omega,k,t} \tag{2.34}$$

We motivate this interpretation by noticing that $\mathbf{N}_t$ is conserved.

Proposition 2.4. *Let $\mathbf{N}_t$ be as in (2.34).For all bounded Borel functions f:*

$$\overline{\rho}(f(j_t[\mathbf{N}_t])) = \overline{\rho}(f(\mathbf{N}_t)) \tag{2.35}$$

[6]We actually mean a renormalized energy: 'The infinite energy of the reservoirs at time 0 has already been subtracted.'

Now we define the change of entropy in the environment up to time t.

$$\mathbf{W}_t \equiv \sum_k \beta_k j_t(\mathbf{N}_{k,t}) \tag{2.36}$$

Its 'steady state expectation' is the entropy production. Our main result is a fluctuation theorem for $\mathbf{W}_t$.

Proposition 2.5. *Assume (**A1**). Let $\mathbf{W}_t$ be defined in (2.36). There is a neighbourhood $\mathcal{U}$ of 0 in $\mathbb{R}$, such that for all $\kappa \in \mathcal{U}$, the limit*

$$\hat{e}(\kappa) \equiv \lim_{t\to+\infty} \frac{1}{t} \log \overline{\rho}[e^{-\kappa \mathbf{W}_t}] \tag{2.37}$$

exists[7] *and is real-analytic.*
We write $\hat{e}(\kappa, H_f)$, indicating explicitly the dependence on the effective Hamiltonian H_f. Similarly, $e(\kappa, H_f)$ is defined by (2.14). Then,

$$\hat{e}(\kappa, H_f) = e(\kappa, H_f) \tag{2.39}$$

and thus all statements in Proposition 2.2 carry over to $\hat{e}(\kappa, H_f)$.

From $\overline{\rho}$ we deduce probability measures $\mathbb{T}_t$ on $\mathbb{R}$. Let $A \subset \mathbb{R}$ be measurable, then

$$\mathbb{T}_t(A) = \overline{\rho}(\mathbf{P}_A) \tag{2.40}$$

where $\mathbf{P}_A$ is the spectral projection on A associated to $\mathbf{W}_t$. Via Legendre-transformation (2.17) implies

$$-\lim_{t\to+\infty} \frac{1}{t} \log \frac{d\mathbb{T}_t(-a)}{d\mathbb{T}_t(a)} = a \tag{2.41}$$

which is (1.1).
Next in Propositions 2.6-2.7 we find that our model satisfies the thermodynamics in the weak-coupling limit.

Proposition 2.6. *Consider $\mathcal{L}_k$ as in (2.5). For all k,*

$$\frac{d}{du}\overline{\rho}(j_u[\mathbf{N}_{k,u}])\big|_{u=0} = \rho(\mathcal{L}_k(H_S)) \tag{2.42}$$

[7] The notation $\overline{\rho}[e^{-\kappa \mathbf{W}_t}]$ is meant as a weak limit of sums of spectral projections $\mathbf{P}_\eta$:

$$\overline{\rho}[e^{-\kappa \mathbf{W}_t}] = \sum_{\eta \in \mathbf{Sp}(\mathbf{W}_0)} e^{-\kappa\eta} \overline{\rho}[j_t(\mathbf{P}_\eta)] \tag{2.38}$$

One can in fact also prove that the vacuum Ψ from (2.27) is in the domain of $j_t(e^{-\kappa \mathbf{W}_t})$ but we do not go into that.

The righthand side of (2.42) are the mean currents as defined in [LS2].
We finally get a Green-Kubo relation between the linear response coefficients $L_{k,l}$ and the current-current correlations

Proposition 2.7. *Assume **(A1)** with $(\beta,\dots,\beta) \in \mathcal{V}$. Define the linear response coefficients for $k,l \in K$*

$$L_{k,l} \equiv \frac{d}{d\beta_l}\frac{d}{dt}\overline{\rho}(j_t[\mathbf{N}_{k,t}])\big|_{t=0,\overline{\beta}=\beta} \tag{2.43}$$

Then,

$$L_{k,l}(H_f) + L_{k,l}(-H_f) \equiv \beta \lim_{t\to+\infty} \frac{1}{t}\overline{\rho}_\beta(j_t[\mathbf{N}_{k,t}\mathbf{N}_{l,t}])(H_f) \tag{2.44}$$

and also

$$L_{k,l}(H_f)+L_{k,l}(-H_f) \equiv \beta \int_{-\infty}^{+\infty} dt \frac{d^2}{dv_1 dv_2}\overline{\rho}_\beta(j_{v_1}[\mathbf{N}_{k,v_1}]j_{t+v_2}[\mathbf{N}_{l,t+v_2}])\big|_{v_1,v_2=0}(H_f) \tag{2.45}$$

Whenever H_S is non-degenerate, we have $L_{k,l}(H_f) = L_{k,l}(-H_f)$ and thus

$$2L_{k,l} \equiv \beta \int_{-\infty}^{+\infty} dt \frac{d^2}{dv_1 dv_2}\overline{\rho}_\beta(j_{v_1}[\mathbf{N}_{k,v_1}]j_{t+v_2}[\mathbf{N}_{l,t+v_2}])\big|_{v_1,v_2=0} \tag{2.46}$$

3 Discussion

3.1 Entropy production for Markov processes

It is well known that the weak coupling generator is 'classical' in the sense that the commutant algebra $\mathcal{A}_{cl} \equiv \{[A,H_S]=0 \big| A \in \mathcal{B}(\mathcal{H})\}$ is invariant. In case the Hamiltonian H_S is non-degenerate, $\mathcal{A}_{cl}$ is a commutative algebra. Then we can construct a Markov process with state space Λ which is the restriction of (the dual of) the semigroup $e^{t\mathcal{L}}$ to $\mathcal{A}_{cl} \cong \mathcal{C}(\Lambda)$. Loosely speaking, let μ be the stationary state, $\Omega^T \equiv \Lambda^{[0,T]}$ the pathspace up to time T, and $\mathcal{P}^T_\mu$ the pathspace measure (starting from μ)of this Markov process. The time reversal operation Θ acts on Ω^T as $(\Omega\xi)(t) = \xi(T-t)$.
For such Markov processes describing a nonequilibrium dynamics, we dispose of a general strategy for identifying the entropy production. It turns out in a lot of interesting cases [M2, M1, MRV] that

$$\ln \frac{d\mathcal{P}^T_\mu}{d\Theta\mathcal{P}^T_\mu}(\xi) = S_t(\xi) + \mathcal{O}(1) \tag{3.1}$$

where $S_t(\xi)$ is the random variable that one physically identifies as the entropy production. The second term in the righthand side is non-extensive in time. The algorithm allows to derive (1.1) from (3.1).
Since we also have a Markov generator, we can apply the same scheme to our setup. To evaluate the result, we however need a physical notion of entropy production in our model. As mentioned earlier, such a notion is rather unambiguous here, see also [LS2, JP3]:

$$\langle \text{current into k'th reservoir} \rangle = \rho(\mathcal{L}_k(H_S)) \tag{3.2}$$

But the mean entropy production based on these currents is **not** equal to the expectation value of (3.1):

$$\sum_k \beta_k \rho(\mathcal{L}_k(H_S)) \neq \mathbb{E}_{\mathcal{P}_\mu}\big(\ln \frac{d\mathcal{P}_\mu}{d\Theta \mathcal{P}_\mu}(\xi)\big) \tag{3.3}$$

For example, take two reservoirs ($k = L, R$), the system Hamiltonian H_S left-right symmetric, and $\psi(\omega, L) = \psi(\omega, R)$ for all ω. One easily checks that the righthand side of (3.3) depends on the temperatures β_L, β_R only through the combination $\frac{\beta_L+\beta_R}{2}$.
This trivial remark shows that it is not enough to look at the semigroup to identify the entropy production.

3.2 Integrated currents within the semigroup approach

Starting from (3.2) one could define the integrated currents operators $\hat{J}_{k,t}$ as

$$\hat{J}_{k,t} = \int_0^t du e^{u\mathcal{L}}(\mathcal{L}_k(H_S)) \tag{3.4}$$

and study their fluctuations.
One can ask whether these fluctuations coincide with these in our model? The answer is partially positive because

Proposition 3.1. *Take $\overline{\beta} = (\beta, \dots, \beta)$. For all $k, l \in K$ and all $u \in \mathbb{R}_0^+$,*

$$\frac{d^2}{dv_1 dv_2} \overline{\rho}_\beta [j_{v_1}(\mathbf{N}_{k,v_1}) j_{u+v_2}(\mathbf{N}_{l,u+v_2})]\big|_{v_1=0, v_2=0} = -\rho_\beta\big(\mathcal{L}_l(H_S) e^{u\mathcal{L}}(\mathcal{L}_k(H_S))\big) \tag{3.5}$$

which gives a relation between Proposition 2.7 and the Green-Kubo relation in [LS2]. Also the averages coincide, leading to (3.2). However it is **not** true that for a reasonable class of functions f

$$\overline{\rho}\big(j_t[f(\mathbf{N}_{k,t})]\big) = \rho\big(f(\hat{J}_{k,t})\big) \tag{3.6}$$

So the mean entropy production and the Green-Kubo-formula can correctly be expressed in terms of the operators $\hat{J}_{k,t}$, but higher order fluctuations of the dissipated heat cannot.

3.3 Connection to microscopic dynamics

We know of two derivations in the literature of this dilation from a microscopic setup:
1 Stochastic Limit
Accardi et al. prove in [AFL] that the weak-coupling limit can be extended to the total evolution of subsystem observables. Let $\mathcal{U}_t^\lambda$ be the evolution (in the interaction picture) on the total system with λ the coupling between subsystem and reservoirs. Then, in a certain sense,

$$\mathcal{U}^\lambda_{-t/\lambda^2}(\mathbb{1}\otimes O)\mathcal{U}^\lambda_{t/\lambda^2} \to_{\lambda\to 0} \mathbf{U}_t^*(\mathbb{1}\otimes O)\mathbf{U}_t \tag{3.7}$$

whereas the traditional weak coupling limit only speaks about convergence in expectation of the left-hand side. The unitary U_t is the solution of (2.22).
2 Repeated Interactions
In [AP], Attal and Pautrat describe a subsystem with Hilbertspace $\mathcal{H}$ interacting repeatedly for a time h with a small reservoir with Hilbertspace $\mathcal{R}$. After each time h, $\mathcal{R}$ is replaced by an identical copy. This procedure ensures that at any time, the subsystem sees a 'fresh' reservoir. In the limit $h \to 0$ the dynamics (in the interaction picture) converges in a certain sense to the solution of a QSDE. One can choose a particular QSDE by tuning the parameters of the interaction. Assume that

$$\mathcal{R} = \bigotimes_{\omega,k} \mathcal{R}_{\omega,k} \tag{3.8}$$

Each $\mathcal{R}_{\omega,k}$ is 2-dimensional with basevectors (θ,ω). Define $a_{\omega,k}$ on $\mathcal{R}_{\omega,k}$ by

$$a_{\omega,k}(\omega) = \theta \qquad a_{\omega,k}\theta = 0 \tag{3.9}$$

Choose the dynamics on $\mathcal{H}\otimes\mathcal{R}$ as $e^{-itH(h)}$ for $0 \le t \le h$ with

$$H(h) = H_f + \frac{1}{\sqrt{h}}\sum_{\omega,k}\left(V_{\omega,k}a^*_{\omega,k} + V^*_{\omega,k}a_{\omega,k}\right) \tag{3.10}$$

Then, through the limiting procedure of [AP], equation (2.22) obtains.

4 Proofs

4.1 Proof of Lemma 2.1

The proof can be found in [BMK]. From

$$\int_\Omega d\sigma \mathcal{W}_t(\sigma)1 = e^{t\mathcal{L}}1 = 1 \tag{4.1}$$

and $\mathcal{W}_t(\sigma) = \mathcal{W}_t(\sigma \cap \Omega_t)$ for all t follows that $\mathbb{P}_{\rho,t}$ is indeed a family of probability measures (for all ρ). Further one checks that for $s < t$ and $\sigma \in \Omega_s$

$$\mathcal{W}_t(\sigma) = \mathcal{W}_s(\sigma)e^{(t-s)\mathcal{L}} \tag{4.2}$$

which yields consistence and hence existence of $\mathbb{P}_\rho$.

4.2 Proof of Proposition 2.4

Fix $t \geq 0$. Remark that $\mathbf{N}_t$ has purely discrete spectrum. Call $\mathbf{P}^t(e)$ a spectral projection of $\mathbf{N}_t$ associated to the eigenvalue e and $\mathbf{P}_{\omega,k,t}(N_{\omega,k})$ a spectral projection of $\mathbf{N}_{\omega,k,t}$ associated to the eigenvalue $N_{\omega,k} \in \mathbb{N}$. Using the notation $\overline{N} \equiv (N_{\omega,k})_{\omega,k} \in \mathbb{N}^\ell$, one has

$$\mathbf{P}^t(e) = \sum_{\substack{x \in \Lambda, \overline{N} \in \mathbb{N}^\ell \\ \epsilon_x + \sum_{\omega,k} \omega N_{\omega,k} = e}} P_x \otimes (\otimes_{\omega,k} \mathbf{P}_{\omega,k,t}(N_{\omega,k})) \tag{4.3}$$

It suffices to prove

$$\overline{\rho}(j_t[\mathbf{P}^t(e)]) = \overline{\rho}(\mathbf{P}^t(e))) \tag{4.4}$$

Remark, using $[V^*_{\omega,k}V_{\omega,k}, H_S] = 0$ and the notation (2.9)

$$P_y\mathcal{W}(\sigma)(P_x)P_y = \begin{cases} \mathcal{W}_t(\sigma)(P_x) & \text{if } \sum_{\omega,k} \omega n_{\omega,k,t} = \epsilon_y - \epsilon_x \\ 0 & \text{otherwise} \end{cases} \tag{4.5}$$

By Lemma 2.30 one has

$$\overline{\rho}\big(j_t[P_x \otimes (\otimes_{\omega,k}\mathbf{P}_{\omega,k,t}(N_{\omega,k}))]\big) = \rho\big(\int_{E(\overline{N},t)} d\sigma \mathcal{W}_t(\sigma)(P_x)\big) \tag{4.6}$$

with $E(\overline{N},t) \equiv \{\sigma \in \Omega \big| n_{\omega,k,t} = N_{\omega,k}\}$. Writing out the left-hand side of (4.4), using (4.5) and (2.6) and adopting a shorthand for the summation of

(4.3)

$$\overline{\rho}\big(j_t(\mathbf{P}^t(e))\big) = \sum_{\epsilon_x+\sum_{\omega,k}\omega N_{\omega,k}=e} \rho\big(\int_{E(\overline{N},t)} \mathcal{W}(\sigma)(P_x)\big) \tag{4.7}$$

$$= \sum_{\epsilon_x+\sum_{\omega,k}\omega N_{\omega,k,t}=e} \rho\big(P_e\big[\int_{E(\overline{N},t)} \mathcal{W}(\sigma)(P_x)\big]P_e\big) \tag{4.8}$$

$$= \sum_x \rho(P_e[e^{t\mathcal{L}}(P_x)]P_e) = \rho(P_e[e^{t\mathcal{L}}(\mathbb{1})]P_e) = \rho(P_e) \tag{4.9}$$

$$\tag{4.10}$$

where we slightly extended the notation by putting $P_e \equiv 0$ whenever $e \notin \mathbf{Sp}(H_s)$. With the same convention, the right-hand side of (4.4) can be expanded as

$$\sum_{\epsilon_x+\sum_{\omega,k}\omega N_{\omega,k,t}=e} (\rho\otimes\phi)(P_x\otimes(\otimes_{\omega,k}\mathbf{P}_{\omega,k,t}(N_{\omega,k})) = \rho(P_e) \tag{4.11}$$

since only for $\overline{N}=0$, the expectation in the vacuum state is non-zero. This proves (4.4) and thus also (2.4).

4.3 Proof of Proposition 2.2 and 2.5

For each κ, define on $\mathcal{B}(\mathcal{H})$

$$\mathcal{L}_\kappa(\cdot) = -i[H_f,\cdot] + \sum_{\omega,k}\big(e^{\kappa\beta_k\omega}V^*_{\omega,k}\cdot V_{\omega,k} - \frac{1}{2}\{V^*_{\omega,k}V_{\omega,k},\cdot\}\big) \tag{4.12}$$

We rely on (2.12) to rewrite (2.14) on the space $\mathcal{B}(\mathcal{H})$

$$e(\kappa) = \lim_{t\to+\infty}\log\rho(e^{t\mathcal{L}_\kappa}(\mathbb{1}))$$

Now the proof is a minor modification of the reasoning in [LS1]. Let $\mathcal{V}\subset\mathcal{B}(\mathcal{H})$ be the (real) subspace of self-adjoint operators and remark that $\mathcal{L}$ leaves $\mathcal{V}$ invariant. Endow $\mathcal{V}$ with the scalar product given by the trace.

$$\langle A,B\rangle = \mathrm{tr}(A^*B)\in\mathbb{R}$$

Remark that $e^{t\mathcal{L}_\kappa}$, viewed as a matrix on $\mathcal{V}$ has real entries. For $\kappa=0$, this matrix has a unique maximal eigenvector (this eigenvector is $\mathbb{1}\in\mathcal{V}$ and the eigenvalue is 1). It is clear that the entries of $e^{\mathcal{L}_\kappa}$ depend on κ through functions like $e^{\kappa\beta_k\omega}$ for some $\beta_k,\omega\in\mathbb{R}$. Hence the entries are real-analytic in κ and we can view $e^{\mathcal{L}_\kappa}$ as a analytic perturbation of $e^{\mathcal{L}_0}$. Since both the multiplicity and the dimension of the Jordan block associated to the eigenvalue 1 is 1, one concludes from basic perturbation theory and from the reality of the matrix entries, that there is a neighbourhood $\mathcal{U}$ of 0 such that for $\kappa \in \mathcal{U}$, $e^{t\mathcal{L}_\kappa}$ has a unique maximal positive eigenvalue E_κ corresponding to a real eigenvector v_κ. Then

$$e(\kappa) = \lim_{t\to+\infty} \frac{1}{t} \log \rho(e^{t\mathcal{L}_\kappa}(\mathbb{1})) = \lim_{t\to+\infty} \frac{1}{t} \log \rho(v_\kappa) e^{tE_\kappa} = E_\kappa$$

since we can choose $\mathcal{U}$ small enough such that $\rho(v_\kappa)$ is bounded away from zero. Because of $V^*_{\omega,k} = e^{\frac{-\beta_k \omega}{2}} V_{-\omega,k}$, one sees that

$$L_\kappa(H_f) = L^*_{1-\kappa}(-H_f) \tag{4.13}$$

Since the highest eigenvalues of L_κ and L^*_κ coincide, and since one can repeat the above argument for L^*_κ, (4.13) yields

$$e_\kappa(H_f) = e_{1-\kappa}(-H_f)$$

If the Hamiltonian H_S is non-degenerate, one can strengthen this result as follows (now completely following [LS1]): Since $\mathcal{L}$ leaves the space of diagonal matrices in the H_S-basis invariant, one can reduce $\mathcal{V}$ to the space of these diagonal matrices. Since we assume that the $V_{\omega,k}$ are non-zero, we can apply the Perron-Frobenius theorem to establish the existence of a unique maximal eigenvalue E_κ and a unique eigenvector v_κ with positive entries. (2.17) follows because of (4.13), since on diagonal matrices there is no difference between $i[H_f, \cdot]$ and $-i[H_f, \cdot]$, both being 0. Finally, (2.39) is a simple consequence of Lemma 2.30.
To prove (2.16) and (2.18), define the time reversal θ on Ω_t as

$$\theta(t_1, \omega_1, k_1; \ldots,) = (t - t_n, -\omega_n, k_n; \ldots,) \tag{4.14}$$

and the time-reversed path space measure $\Theta\mathbb{P}^t(\cdot) = \mathbb{P}^t(\theta\cdot)$. Consider the function $S_t : \Omega^t \to \mathbb{R}$

$$S^t_\mu(\sigma) = \log \frac{d\mathbb{P}^t_\mu(H_f)}{d(\Theta\mathbb{P}^t_\mu)(-H_f)}(\sigma) \tag{4.15}$$

with μ a density matrix on $\mathcal{B}(\mathcal{H})$. Let $\mathrm{Id} \equiv \frac{\mathbb{1}}{d}$ where d is the dimension of the system space $\mathcal{H}$. One checks explicitly

$$S^t_{\mathrm{Id}}(\cdot) = \mathbf{w}_t(\cdot) \tag{4.16}$$

Furthermore, because of Lemma 2.1, one has for all μ on $\mathcal{B}(\mathcal{H})$

$$\int d\mathbb{P}^t_\mu e^{-S^t_\mu} = \int d\Theta\mathbb{P}^t_\mu = 1 \tag{4.17}$$

From Jensen's inequality one gets then for all $t \in \mathbb{R}^+$

$$\int d\mathbb{P}^t_{\mathrm{Id}} \mathbf{w}_t \geq 0 \tag{4.18}$$

Since by **A1** the semigroup relaxes to ρ exponentially fast (see e.g. [LS2]), we also have

$$\lim_{t\to+\infty} \frac{1}{t} \int d\mathbb{P}^t_\rho \mathbf{w}_t = \lim_{t\to+\infty} \frac{1}{t} \int d\mathbb{P}^t_{\mathrm{Id}} \mathbf{w}_t \geq 0 \tag{4.19}$$

In the case of non-degenerate H_S, stationarity of ρ yields

$$\int d\mathbb{P}^t_\rho S^t_\rho = \int d\mathbb{P}^t_\rho \mathbf{w}_t \tag{4.20}$$

Remark that the function $f : \mathbb{R} \to \mathbb{R} : x \to e^{-x} + x - 1$ is positive for all x and monotone for $x \geq 0$. A Chebyshev inequality with $\delta > 0$ yields.

$$\int d\mathbb{P}^t_\rho(S^t_\rho) = \int d\mathbb{P}^t_\rho(e^{-S^t_\rho} + S^t_\rho - 1) \geq (e^{-\delta} + \delta - 1)\mathbb{P}^t_\rho(|S^t_\rho| \geq \delta) \tag{4.21}$$

Whenever not all β_k's are equal, take k, k' such that $\beta_k \neq \beta_{k'}$. Choose $\omega \in F$ and put $\sigma(s,u) = (\omega, k, s; -\omega, k', u)$ and $E = \{\sigma(s,u), s \leq u \leq t\} \in \mathcal{A}_t$. Then,

$$S_t(E) = \mathbf{w}_t(E) = (\beta_k - \beta_{k'})\omega \tag{4.22}$$

and thus $\mathbb{P}^t_\rho\big(|S^t_\rho| \geq |(\beta_k - \beta_{k'})\omega|\big) \geq \mathbb{P}^t_\rho(E) > 0$. Hence, (4.21) implies (2.18).

4.4 Proof of Proposition 2.7

Remark first that a slight generalization of Proposition 2.5 can be proven (in exactly the same way). Write $e(\kappa_1, \ldots, \kappa_m) = e^{-\beta \sum_k \kappa_k \mathbf{N}_k}$. Now (2.15) reads

$$e(\kappa_1, \ldots, \kappa_m, H_f) = e(\frac{\beta_1}{\beta} - \kappa_1, \ldots, \frac{\beta_k}{\beta} - \kappa_m, -H_f) \tag{4.23}$$

By differentiating with respect to κ_k and $\frac{\beta_l}{\beta}$ we obtain

$$L_{k,l}(H_f) + L_{k,l}(-H_f) \equiv \beta \lim_{t\to+\infty} \frac{2}{t} \overline{\rho}_\beta(j_t[\mathbf{N}_{k,t}\mathbf{N}_{l,t}])(-H_f) \tag{4.24}$$

That computation is standard (see again [LS1], the interchange of the derivatives and the limit $t \to +\infty$ is justified by a theorem in [B], stating that analyticity of the large deviation generating function implies the central limit theorem.). One sees that the right-hand side is the same for H_f or $-H_f$. The Green-Kubo relation follows from (4.24) by time-translation invariance of the correlation function, which follows from (2.29), and from the existence of the derivatives, which is proven in Proposition 3.1.

4.5 Proof of Proposition 3.1

Let $\overline{N} = (N_\omega)_\omega, \overline{M} = (M_\omega)_\omega \in \mathbb{N}^{|F|}$. We write the derivative on the right-hand side of (3.5) as a limit $((v_1, v_2) \to (0,0))$[8] of the infinite sum

$$\frac{1}{v_1 v_2}\overline{\rho}\big(j_{v_1}[\mathbf{N}_{k_1,v_1}](j_{u+v_2}[\mathbf{N}_{k_2,u+v_2}]-j_u[\mathbf{N}_{k_2,u}])\big) = \sum_{\overline{N},\overline{M}} \frac{U_{k_1}^{(0,v_1)} U_{k_2}^{(u,v_2)}}{v_1 v_2} \int_{E^{v_1,v_2}_{\overline{N};\overline{M}}} d\sigma \rho(\mathcal{W}(\sigma)(\mathbb{1})) \tag{4.25}$$

with

$$U_{k_1}^{(0,v_1)}(\sigma) = \sum_\omega \omega n_{\omega,k_1,v_1}(\sigma)$$

$$U_{k_2}^{(u,v_2)}(\sigma) = \sum_\omega \omega \big(n_{\omega,k_2,u+v_2}(\sigma) - n_{\omega,k_2,u}(\sigma)\big)$$

and

$$E^{v_1,v_2}_{\overline{M},\overline{M}} = \{\sigma \in \Omega \big| n_{\omega,k_1,v_1}(\sigma) = N_\omega, \big(n_{\omega,k_2,u+v_2}(\sigma) - n_{\omega,k_2,u}(\sigma)\big) = M_\omega\}$$

Put $C \equiv \max \|V_{\omega,k}\|^2$, $D \equiv \max|\omega|$ and $|\overline{N}| = \sum_\omega N_\omega$, $|\overline{M}| = \sum_\omega M_\omega$. By the Dyson series, the terms on the right-hand side of (4.25) satisfy for all $v_1, v_2 \in [0,1]$

$$\Big|\frac{U_{k_1}^{(0,v_1)} U_{k_2}^{(u,v_2)}}{v_1 v_2} \int_{\sigma \in E_{\overline{N},v_1;\overline{M};v_2}} \rho(\mathcal{W}(\sigma)(\mathbb{1}))\Big| \le D^2 |\overline{N}||\overline{M}| \frac{C^{|\overline{N}|+|\overline{M}|}}{|\overline{N}|!|\overline{M}|!} \tag{4.26}$$

and hence (4.25) can be bounded by the absolutely convergent series

$$\sum_{p=1}^{+\infty} C^p (pD)^2 \frac{1}{(\lfloor \frac{p}{2} \rfloor !)^2} \tag{4.27}$$

[8]For notational clarity, we assume v_1, v_2 positive, the full statement follows analogously with the help time-translation invariance (2.29).

such that the p'th term bounds the sum of all terms in (4.25) with $|\overline{N}|+|\overline{M}| = p$. Hence one can perform the limit $(v_1, v_2) \to (0,0)$ term by term to obtain

$$\sum_{\omega_1,\omega_2} \omega_1\omega_2\rho\big(\mathcal{J}_{\omega_2,k_2} e^{u\mathcal{L}} \mathcal{J}_{\omega_1,k_1}(\mathbb{1})\big) \tag{4.28}$$

Putting $\rho \equiv \rho_\beta$, (3.5) follows after some reshuffling, using

$$V^*_{\omega,k} = e^{-\frac{\beta_k\omega}{2}} V_{-\omega,k} \quad \sum_\omega \omega V^*_{\omega,k} V_{\omega,k} = \mathcal{L}_k(H_S) \quad \rho_\beta V_{\omega,k} = V_{\omega,k}\rho_\beta e^{\beta_k\omega} \tag{4.29}$$

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