Entropic Fluctuations of Quantum Dynamical Semigroups

V. Jakšić · C.-A. Pillet · M. Westrich

Abstract We study a class of finite dimensional quantum dynamical semigroups $\{e^{t\mathcal{L}}\}_{t\geq 0}$ whose generators $\mathcal{L}$ are sums of Lindbladians satisfying the detailed balance condition. Such semigroups arise in the weak coupling (van Hove) limit of Hamiltonian dynamical systems describing open quantum systems out of equilibrium. We prove a general entropic fluctuation theorem for this class of semigroups by relating the cumulant generating function of entropy transport to the spectrum of a family of deformations of the generator $\mathcal{L}$. We show that, besides the celebrated Evans-Searles symmetry, this cumulant generating function also satisfies the translation symmetry recently discovered by Andrieux et al., and that in the linear regime near equilibrium these two symmetries yield Kubo's and Onsager's linear response relations.

Keywords Quantum markovian dynamics · Entropy production · Nonequilibrium steady states · Fluctuation theorem · Detailed balance · Weak coupling limit · Davies generator

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1 Introduction

Markov semigroups are widely used to model non-equilibrium phenomena in classical statistical physics. Their non-commutative counterparts—quantum dynamical semigroups—play the same role in quantum statistical physics (see, e.g., [4, 55] for pedagogical introductions to the subject). The development of the mathematical theory of QDS started in 1974 with the seminal works of Brian Davies [10–12] where he showed that QDS emerge as effective dynamics of open systems weakly coupled to extended reservoirs. These groundbreaking works were followed by the celebrated 1976 papers of Lindblad [46, 47] and Gorini, Kossakowski and Sudarshan [35] on the structure of the generator of QDS (see also [7]). Herbert made several fundamental contributions at this early stage of development. In [58, 59] he gave efficient criteria for the existence and uniqueness of a stationary state and approach to equilibrium. Together with Joel Lebowitz, in [48] he developed a comprehensive picture of the nonequilibrium thermodynamics of weakly coupled open systems. This work remains a standard reference and has been a source of inspiration for many later developments on the subject, including the present one. Among other things, in [48] Herbert and Joel introduced the central concept of entropy production, which was further discussed in [60], and developed the linear response theory for thermodynamical forces. The closely related linear response theory for weakly coupled open systems under mechanical drive was developed by Herbert in a joint paper with Brian Davies [23]. In another enlightening work, Herbert and R. Dümcke [24] showed that some of the generators that were (and sometimes still are) used to describe the weak coupling limit lead to negative probabilities. Years later, Herbert came back to the subject and, with Walter Aschbacher, showed that when properly applied to nonequilibrium situations, the algebraic criterion of [59] also ensures the strict positivity of entropy production [5].

In 1993/4, using a new scheme to construct nonequilibrium statistical ensembles of interacting particle systems, Evans, Cohen and Morriss discovered some universal features of the fluctuations of entropy production in transient regimes of deterministic classical systems out of thermal equilibrium [26, 30]. One year later, Gallavotti and Cohen proved that some steady states of highly chaotic dynamical systems (SRB measures of transitive Anosov systems) display the same features [33, 34]. These discoveries, nowadays called fluctuation relations or fluctuation theorems, triggered a large amount of works during the last two decades (see [40, 56] and references therein). In particular, Kurchan showed that the fluctuation relations hold for a Brownian particle in a force field [43]. Subsequently, Herbert and Joel formulated and proved fluctuation relations for general Markov processes [49] while Maes derived a local version of the fluctuation relations from the Gibbsian nature of the path space measure associated to such processes [50] (see also [52]). As shown by Gallavotti [32], the fluctuation relations can be seen as a far from equilibrium generalization of the familiar near equilibrium fluctuation-dissipation relations (Green-Kubo formulae, Onsager reciprocity relations).

The attempts to extend fluctuation relations to quantum domain have led to a number of surprises. The naive quantization of the classical transient fluctuation relations fails and there is no obvious way to implement the steady state fluctuation relations. These problems have attracted a lot of interest and generated a huge literature which we will not try to review here. We shall only mention a few works which, in our opinion, are relevant to the development of a mathematical understanding of the subject. The interested reader can consult [27] for an exhaustive review and an extended list of references to the physics literature and [36, 39] for recent mathematical developments.

To our knowledge, a (transient) quantum fluctuation relation based on operationally defined counting statistics was first derived by Kurchan in 2000 [44]. Shortly afterwards, Matsui and Tasaki obtained an apparently unrelated abstract fluctuation relation for open quantum systems in terms of the spectral measure of a relative modular operator [53]. The connection between their result and the counting statistics of entropic transport was established in [36].

Within the framework of QDS, de Roeck and Maes [22] used the unraveling technique to obtain the first complete transient fluctuation theorem (see Sect. 6). The relation between this Markovian approach to fluctuations and the Hamiltonian description of the dynamics of a small system weakly coupled to an extended environment was discussed by de Roeck in [18] and by Derezinski, de Roeck and Maes [14] (see also Sect. 5). The works [14, 18, 22] complete the program of [48] regarding nonequilibrium thermodynamics of weakly coupled open systems. The first proof of the transient fluctuation theorem for a fully Hamiltonian system (the spin-boson model) was given by de Roeck in the important paper [19] (see also [20, 21, 41]).

Among the non-rigorous works let us mention the important observation of Andrieux, Gaspard, Monnai and Tasaki [2] that global conservation laws (energy and charge) induce translation symmetries in the cumulant generating function of (energy and charge) fluxes. Translation symmetries and entropic fluctuation relation lead to fluctuation relations for individual fluxes and, following the arguments of [32, 49], to Green-Kubo and Onsager relations near thermal equilibrium. We also mention the recent work by Chetrite and Mallik [8] where finite time fluctuation relations (similar to the classical transient Evans-Searles relations) for time-reversible quantum dynamical semigroups are derived.

This work is of a review nature and we do not prove any specific new results. The purpose of the paper is to provide an abstract general setup for the non-equilibrium statistical mechanics of QDS and to generalize and streamline the proof of the full fluctuation theorem of [14, 18, 22] emphasizing (in the spirit of [60]) the minimal mathematical structure behind the result. The fluctuation theorem we discuss includes large deviation bounds and the central limit theorem for individual entropic fluxes, as well as linear response formulae and the fluctuation-dissipation relations near equilibrium, and applies to the weakly coupled quantum systems studied in [48]. Although the paper is mathematically self-contained, it is intended for readers familiar with the works [48, 58–60]. This paper can be also viewed as an introduction to [41] where we discuss fluctuation relations and non-equilibrium statistical mechanics of the fully Hamiltonian Pauli-Fierz systems.

The paper is organized as follows. In Sect. 2 we recall basic definitions and facts about positive maps and QDS. In Sect. 3 we introduce the setup of QDS out of equilibrium, and state our main results. In Sect. 4 we show that open systems weakly coupled to thermal reservoirs fit into our general setup. In Sects. 5 and 6 we relate our results to the full counting statistics of entropic transport and the unraveling of quantum dynamical semigroups. Finally, Sect. 7 is devoted to the proofs.

2 Preliminaries

Let $\mathcal{H}$ be a finite dimensional Hilbert space and $\mathcal{O} = \mathcal{B}(\mathcal{H})$ the C^*-algebra of all linear operators on $\mathcal{H}$ (the identity operator will be always denoted by $\mathbb{1}$). Equipped with the inner product $\langle X|Y \rangle = \mathrm{tr}(X^*Y)$, $\mathcal{O}$ is a Hilbert space. The adjoint and the spectrum of a linear map $\Phi : \mathcal{O} \to \mathcal{O}$ are denoted by Φ^* and $\mathrm{sp}(\Phi)$. Id denotes the identity of $\mathcal{B}(\mathcal{O})$. A subset $\mathcal{A} \subset \mathcal{O}$ is called self-adjoint if $X \in \mathcal{A} \Rightarrow X^* \in \mathcal{A}$. The commutant of a subset $\mathcal{A} \subset \mathcal{O}$ is $\mathcal{A}' = \{B \in \mathcal{O} \,|\, AB = BA \text{ for all } A \in \mathcal{A}\}$.

We denote by $\mathcal{O}_+ = \{X \mid X \geq 0\}$ the cone of positive elements of $\mathcal{O}$. A linear map Φ from $\mathcal{O}$ to another unital C^*-algebra $\mathcal{B}$ is called unital if $\Phi(\mathbb{1}) = \mathbb{1}$, positive if $\Phi(\mathcal{O}_+) \subset \mathcal{B}_+$, and positivity improving if $\Phi(X) > 0$ for all non-zero $X \in \mathcal{O}_+$. A positive linear map is automatically a $*$-map, i.e., it satisfies $\Phi(X^*) = \Phi(X)^*$. A positive linear map $\Phi : \mathcal{O} \to \mathcal{O}$ is called Schwartz if

$$\Phi(X^*)\Phi(X) \leq \|\Phi\|\Phi(X^*X),$$

for all $X \in \mathcal{O}$. Note that if Φ is Schwartz, then $\|\Phi\| = \|\Phi(\mathbb{1})\|$.

A state on $\mathcal{O}$ is a positive and unital linear map $\rho : \mathcal{O} \to \mathbb{C}$. Any state ρ has the form $\rho(X) = \mathrm{tr}(DX)$ for some $D \in \mathcal{O}_+$ satisfying $\mathrm{tr}(D) = 1$. Such an operator D is called a density matrix. In the following, we shall use the same symbol to denote a density matrix and the state it induces on $\mathcal{O}$ (hence, $\rho(X) = \mathrm{tr}(\rho X)$, etc.). With this convention, the set of states on $\mathcal{O}$, which we denote by $\mathfrak{S}$, is a closed convex subset of $\mathcal{O}_+$. A state ρ is called faithful if $\rho > 0$, and we denote by $\mathfrak{S}_f$ the set of faithful states. $\mathfrak{S}_f$ is an open convex and dense subset of $\mathfrak{S}$.

A linear map $\Phi : \mathcal{O} \to \mathcal{O}$ is called completely positive (CP) if $\sum_{i,j} B_i^* \Phi(A_i^* A_j) B_j \geq 0$ for any finite families $\{A_1, \ldots, A_N\}, \{B_1, \ldots, B_N\} \subset \mathcal{O}$. Equivalently, Φ is CP if $\Phi \otimes \mathrm{Id}$ is a positive map on $\mathcal{O} \otimes \mathcal{B}(\mathbb{C}^N)$ for all $N \geq 1$. A CP map is automatically Schwartz. We denote by $\mathrm{CP}(\mathcal{O})$ the monoid of completely positive maps, and by $\mathrm{CP}_1(\mathcal{O})$ the sub-monoid of unital maps. $\mathrm{CP}(\mathcal{O})$ is a convex cone and $\mathrm{CP}_1(\mathcal{O})$ is a convex set. Stinespring's theorem [61] asserts that $\Phi \in \mathrm{CP}(\mathcal{O})$ iff there exists a finite family $\{V_j\}_{j \in J}$ in $\mathcal{O}$ such that

$$\Phi(X) = \sum_{j \in J} V_j^* X V_j, \tag{1}$$

for all $X \in \mathcal{O}$. The formula (1) is called a Kraus representation of Φ. Such representation is in general not unique.

Unital CP maps naturally arise in the quantum mechanics of open systems. Indeed, assume that the quantum system $\mathcal{S}$ with Hilbert space $\mathcal{H}$ interacts with some environment described by the Hilbert space $\mathcal{H}_{\mathrm{env}}$. According to the general structure of quantum mechanics, the evolution of the joint system over some time interval is given by a unitary U on $\mathcal{H} \otimes \mathcal{H}_{\mathrm{env}}$. Thus, if X is an observable of the system $\mathcal{S}$, then its Heisenberg evolution over the considered time interval is given by the map

$$\Phi(X) = \mathrm{tr}_{\mathcal{H}_{\mathrm{env}}}\big((\mathbb{1} \otimes \rho_{\mathrm{env}})U^*(X \otimes \mathbb{1})U\big),$$

where $\mathrm{tr}_{\mathcal{H}_{\mathrm{env}}}(\cdot)$ denotes the partial trace over the environment Hilbert space and ρ_{env} is the initial state of the environment. One easily checks that Φ is a unital CP map such that, for any state ρ of $\mathcal{S}$,

$$\mathrm{tr}\big(\rho\Phi(X)\big) = \mathrm{tr}\big((\rho \otimes \rho_{\mathrm{env}})U^*(X \otimes \mathbb{1})U\big).$$

A positive linear map Φ is called irreducible (in the sense of Davies [9]) if the inequality $\Phi(P) \leq \lambda P$, where P is a projection and $\lambda > 0$, holds only for $P = 0$ or $P = \mathbb{1}$. If Φ is positivity improving, then obviously Φ is irreducible. In terms of a Kraus decomposition, irreducibility can be characterized as follows (see, e.g., [57]):

Theorem 2.1 *Let Φ be a $\mathrm{CP}(\mathcal{O})$ map with a Kraus decomposition (1) and let $\mathcal{A}$ be the subalgebra of $\mathcal{O}$ generated by $\{V_j \mid j \in J\}$ and $\mathbb{1}$. Then Φ is irreducible iff $\mathcal{A}\psi = \mathcal{H}$ for any non-zero vector $\psi \in \mathcal{H}$.*

For reader's convenience, we shall prove Theorem 2.1 in Sect. 7.2.

The adjoint Φ^* of a linear map Φ is positive/positivity improving/CP/irreducible iff Φ is. Φ^* is trace preserving, i.e., $\mathrm{tr}(\Phi^*(X)) = \mathrm{tr}(X)$ for all $X \in \mathcal{O}$, iff Φ is unital. In particular, Φ^* maps $\mathfrak{S}$ into itself iff Φ is positive and unital. A state $\rho \in \mathfrak{S}$ is called Φ-invariant if $\Phi^*(\rho) = \rho$, which is equivalent to $\rho(\Phi(X)) = \rho(X)$ for all $X \in \mathcal{O}$.

Let $\{e^{t\mathcal{L}}\}_{t \geq 0}$ be a continuous semigroup of linear maps on $\mathcal{O}$ generated by a linear map $\mathcal{L}$. This semigroup is called unital/positive/positivity improving/CP($\mathcal{O}$)/CP$_1$($\mathcal{O}$) iff $e^{t\mathcal{L}}$ is for all $t > 0$. A CP$_1$($\mathcal{O}$) semigroup is called *quantum dynamical semigroup* (QDS).[1]

Let $\{e^{t\mathcal{L}}\}_{t \geq 0}$ be a positive unital semigroup. A state ρ is called steady (or stationary) if $\rho(e^{t\mathcal{L}}(X)) = \rho(X)$ for all $t \geq 0$ and $X \in \mathcal{O}$. Clearly, ρ is steady iff $\mathcal{L}^*(\rho) = 0$.

A positive unital semigroup $\{e^{t\mathcal{L}}\}_{t \geq 0}$ is said to be relaxing to a steady state ρ_+ if

$$\lim_{t \to \infty} e^{t\mathcal{L}^*}(\rho) = \rho_+, \tag{2}$$

for all $\rho \in \mathfrak{S}$. The relaxation is exponentially fast if there exists $\gamma > 0$ such that for all states ρ,

$$e^{t\mathcal{L}^*}(\rho) = \rho_+ + O\big(e^{-\gamma t}\big),$$

as $t \to \infty$. The relaxation to a steady state is an ergodic property that plays a fundamental role in the statistical mechanics of QDS.

Our study of the large deviation theory of QDS will be based on the following result.

Theorem 2.2 *Let $\{e^{t\mathcal{L}}\}_{t \geq 0}$ be a positivity improving* CP($\mathcal{O}$) *semigroup and*

$$\ell = \max\big\{\mathrm{Re}\,\lambda \,|\, \lambda \in \mathrm{sp}(\mathcal{L})\big\}.$$

Then ℓ is a simple eigenvalue of $\mathcal{L}$ and is the only eigenvalue of $\mathcal{L}$ on the line $\mathrm{Re}\,z = \ell$. For any state ρ on $\mathcal{O}$, one has

$$\ell = \lim_{t \to \infty} \frac{1}{t} \log \rho\big(e^{t\mathcal{L}}(X)\big), \tag{3}$$

for all non-zero $X \in \mathcal{O}_+$. If in addition the semigroup $\{e^{t\mathcal{L}}\}_{t \geq 0}$ is unital, then $\ell = 0$ and the semigroup is relaxing exponentially fast to a faithful steady state ρ_+.

The proof of this theorem is based on the Perron-Frobenius theory of positive maps developed in [28] and is given in Sect. 7.2.

It is a fundamental result of Lindblad [46, 47], Gorini, Kossakowski and Sudarshan [35], and Christensen and Evans [7], that $\{e^{t\mathcal{L}}\}_{t \geq 0}$ is a CP($\mathcal{O}$) semigroup iff there are $K \in \mathcal{O}$ and $\Phi \in \mathrm{CP}(\mathcal{O})$ such that

$$\mathcal{L}(X) = K^*X + XK + \Phi(X), \tag{4}$$

for all $X \in \mathcal{O}$. For short, we shall call the generator of a CP($\mathcal{O}$) semigroup a *Lindbladian*, and the r.h.s. of Eq. (4) a *Lindblad decomposition* of $\mathcal{L}$. Although the Lindblad decomposition is not unique, it can be effectively used to characterize some important properties of the semigroup. In particular, we have:

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[1]The name *quantum Markov semigroup* is also used in the literature.

Theorem 2.3 *Let* $\{e^{t\mathcal{L}}\}_{t\geq 0}$ *be a* CP($\mathcal{O}$) *semigroup and* $\mathcal{L}(X) = K^*X + XK + \Phi(X)$ *a Lindblad decomposition. If* Φ *is irreducible*, *then the semigroup is positivity improving.*

We shall prove this theorem in Sect. 7.2. Theorems 2.1 and 2.3 provide an effective criterion for verifying the positivity improving assumption of Theorem 2.2 (see Sect. 4).

If $\{e^{t\mathcal{L}}\}_{t\geq 0}$ is a QDS, then $\mathcal{L}(\mathbb{1}) = 0$, and it follows from (4) that

$$\mathcal{L}(X) = \mathrm{i}[T, X] - \frac{1}{2}\{\Phi(\mathbb{1}), X\} + \Phi(X), \qquad (5)$$

where T is a self-adjoint element of $\mathcal{O}$ and $\Phi \in \mathrm{CP}(\mathcal{O})$. We shall also refer to the r.h.s. of Eq. (5) as a *Lindblad decomposition* of $\mathcal{L}$.

The *dissipation function* of a QDS $\{e^{t\mathcal{L}}\}_{t\geq 0}$ is the sesquilinear map $D : \mathcal{O} \times \mathcal{O} \to \mathcal{O}$ defined by

$$D(X, Y) = \mathcal{L}(X^*Y) - \mathcal{L}(X^*)Y - X^*\mathcal{L}(Y).$$

If (5) is the Lindblad decomposition of $\mathcal{L}$ and (1) a Kraus decomposition of Φ, then

$$D(X, X) = \sum_{j\in J}[V_j, X]^*[V_j, X].$$

Hence, $D(X, X) \geq 0$ and $D(X, X) = 0$ iff $X \in \{V_j \mid j \in J\}'$. The dissipation function of a QDS was introduced by Lindblad in [46] and has played an important role in many subsequent works on the subject.

The *detailed balance condition* and *time-reversal invariance* will play an important role in our work. Both properties refer to a pair $(\rho, \mathcal{L})$, where ρ is a faithful state and $\mathcal{L}$ is the generator of a QDS. Note that any faithful state induces an inner product $\langle X|Y\rangle_\rho = \langle X\rho^{1/2}|Y\rho^{1/2}\rangle = \mathrm{tr}(\rho X^*Y)$ on $\mathcal{O}$. We call the ρ-adjoint of a linear map Φ its adjoint Φ^ρ w.r.t. this inner product. In particular, we say that a linear map Φ is ρ-self-adjoint if $\Phi^\rho = \Phi$.

Definition 2.4 Consider a pair $(\rho, \mathcal{L})$, where ρ is a faithful state and $\mathcal{L}$ is a Lindbladian generating a QDS.

(a) The pair $(\rho, \mathcal{L})$ is said satisfy the detailed balance condition if $\mathcal{L}^*(\rho) = 0$ and there exists Lindblad decomposition $\mathcal{L} = \mathrm{i}[T, \cdot] - \frac{1}{2}\{\Phi(\mathbb{1}), \cdot\} + \Phi$ such that Φ is ρ-self-adjoint.
(b) The pair $(\rho, \mathcal{L})$ is said to be time-reversal invariant (TRI) if there exists an involutive anti-linear $*$-automorphism $\Theta : \mathcal{O} \to \mathcal{O}$, called the time-reversal, such that $\mathcal{L}^\rho \circ \Theta = \Theta \circ \mathcal{L}$ and $\Theta(\rho) = \rho$.

Definition 2.4(a) is equivalent to the definition of detailed balance given by Kossakowski, Frigerio, Gorini, and Verri [42] (see Theorem 7.2 below). The above definition, however, is technically and conceptually more suitable for our purposes.[2] The detailed balance condition is characteristic of QDS describing the interaction of a system $\mathcal{S}$ with an environment at equilibrium (see [42, 48]).

For the motivation regarding the definition of time-reversal we refer the reader to Sect. 4 and [31, 51]. We recall that $\Theta : \mathcal{O} \to \mathcal{O}$ is an involutive anti-linear $*$-automorphism iff there exists an anti-unitary involution $\theta : \mathcal{H} \to \mathcal{H}$ such that $\Theta(X) = \theta X\theta$ (see Exercise 4.36 in [36]), and that $\Theta(\rho) = \rho$ iff $\rho(\Theta(X)) = \rho(X^*)$ for all $X \in \mathcal{O}$.

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[2]Alternative definitions of detailed balance can be found in [1, 3].

3 Quantum Dynamical Semigroups Out of Equilibrium

3.1 The Setup

We shall study QDS $\{e^{t\mathcal{L}}\}_{t\geq 0}$ on $\mathcal{O} = \mathcal{B}(\mathcal{H})$, $\dim \mathcal{H} < \infty$, satisfying the following ergodicity condition

(**ER**) The QDS $\{e^{t\mathcal{L}}\}_{t\geq 0}$ is positivity improving.

Furthermore, we shall focus on semigroups whose generator $\mathcal{L}$ has the special structure given by

(**DB**) There are M pairs $(\rho_j, \mathcal{L}_j)$ satisfying the detailed balance condition such that

$$\mathcal{L} = \sum_{j=1}^{M} \mathcal{L}_j.$$

We shall interpret each of the *M sub-Lindbladians* $\mathcal{L}_j$ as describing the interaction of a quantum system $\mathcal{S}$ with some reservoir $\mathcal{R}_j$. A QDS is out of equilibrium if Hypothesis (DB) holds and $\rho_j \neq \rho_k$ for some pair j, k (such a QDS describes the interaction of the system $\mathcal{S}$ with an environment $\sum_j \mathcal{R}_j$ out of equilibrium).

A QDS satisfying (DB) is called time-reversible if

(**TR**) All the pairs $(\rho_j, \mathcal{L}_j)$ are time-reversal invariant with the same time-reversal map Θ.

Finally, we shall investigate more closely the special case where the states ρ_j are thermal equilibrium states at inverse temperature $\beta_j > 0$ for the same Hamiltonian.

(**KMS$_\beta$**) $\beta = (\beta_1, \ldots, \beta_M) \in \mathbb{R}_+^M$ and there exists a self-adjoint element $H_\mathcal{S} \in \mathcal{O}$ such that

$$\rho_j = \frac{e^{-\beta_j H_\mathcal{S}}}{\mathrm{tr}(e^{-\beta_j H_\mathcal{S}})},$$

for $j = 1, \ldots, M$.

As we shall see in Sect. 4, Hypotheses (ER), (DB) and (KMS$_\beta$) are naturally satisfied by the QDS describing the weak coupling (van Hove) limit dynamics of an open quantum system $\mathcal{S}$ with Hilbert space $\mathcal{H}$ interacting with an environment made of M thermal reservoirs. In this case, the Lindbladian $\mathcal{L}_j$ pertains to the interaction of $\mathcal{S}$ with the jth reservoir and the state ρ_j is a steady state of the system coupled only to this reservoir. If the joint dynamics of the system and reservoirs is time-reversal invariant, then Hypothesis (TR) is also satisfied.

3.2 Main Result

Suppose that a QDS $\{e^{t\mathcal{L}}\}_{t\geq 0}$ satisfies Hypothesis (DB). Our main technical result concerns the properties of the deformations of this QDS generated by

$$\mathcal{L}_{(\alpha)}(X) = \sum_{j=1}^{M} \mathcal{L}_j(X\rho_j^{-\alpha_j})\rho_j^{\alpha_j}, \tag{6}$$

where $\alpha = (\alpha_1, \ldots, \alpha_M) \in \mathbb{R}^M$. We will use the notation $1 = (1, \ldots, 1)$ wherever the meaning is clear within the context, e.g., $1 - \alpha = (1 - \alpha_1, \ldots, 1 - \alpha_M)$. Let

$$e(\alpha) = \max\{\operatorname{Re} \lambda \mid \lambda \in \operatorname{sp}(\mathcal{L}_{(\alpha)})\}.$$

Theorem 3.1 *Suppose that Hypothesis* (DB) *holds. Then:*

(1) $\{e^{t\mathcal{L}_{(\alpha)}}\}_{t\geq 0}$ *is a* CP($\mathcal{O}$) *semigroup for all* $\alpha \in \mathbb{R}^M$.
(2) *For any state* ρ *on* $\mathcal{O}$, *there is a Borel probability measure* P_ρ^t *on* $\mathbb{R}^M$ *such that*

$$\operatorname{tr}\big(\rho e^{t\mathcal{L}_{(\alpha)}}(\mathbb{1})\big) = \int_{\mathbb{R}^M} e^{-t\alpha \cdot \varsigma} \, dP_\rho^t(\varsigma).$$

We denote by $\langle \cdot \rangle_{\rho,t}$ *the expectation w.r.t. this measure.*

In the remaining statements we assume that Hypothesis (ER) *is satisfied.*

(3) *For all* $\alpha \in \mathbb{R}^M$ *the* CP($\mathcal{O}$) *semigroup* $\{e^{t\mathcal{L}_{(\alpha)}}\}_{t\geq 0}$ *is positivity improving. In particular, the QDS* $\{e^{t\mathcal{L}}\}_{t\geq 0}$ *is relaxing exponentially fast to a steady state* ρ_+.
(4) *For all* $\alpha \in \mathbb{R}^M$, $e(\alpha)$ *is a simple eigenvalue of* $\mathcal{L}_{(\alpha)}$ *and this operator has no other eigenvalues on the line* $\operatorname{Re} z = e(\alpha)$. *Moreover, for any state* ρ *and all* $\alpha \in \mathbb{R}^M$,

$$\lim_{t\to\infty} \frac{1}{t} \log\langle e^{-t\alpha \cdot \varsigma}\rangle_{\rho,t} = e(\alpha). \tag{7}$$

(5) *The function* $\mathbb{R}^M \ni \alpha \mapsto e(\alpha)$ *is real analytic and convex.*
(6) *Relation* (7) *holds for* α *in an open neighborhood of* $\mathbb{R}^M$ *in* $\mathbb{C}^M$.
(7) *If Hypothesis* (TR) *is satisfied, then*

$$e(1 - \alpha) = e(\alpha), \tag{8}$$

for all $\alpha \in \mathbb{R}^M$.
(8) *If Hypothesis* (KMS$_\beta$) *is satisfied, then*

$$e\big(\alpha + \lambda \beta^{-1}\big) = e(\alpha),$$

for all $\alpha \in \mathbb{R}^M$ *and all* $\lambda \in \mathbb{R}$ *with* $\beta^{-1} = (\beta_1^{-1}, \ldots, \beta_M^{-1})$.

Remark 1 The identity (8) is the QDS analog of the generalized Evans-Searles symmetry of time-reversal invariant classical dynamical systems (see [26, 30, 40]). However, contrary to the classical case, we do not expect that the function

$$\alpha \mapsto e_{\rho,t}(\alpha) = \log \operatorname{tr}\big(\rho e^{t\mathcal{L}_{(\alpha)}}(\mathbb{1})\big),$$

satisfies this symmetry for fixed finite time t. A notable exception is provided by the very special "chaotic state" $\rho = \rho_{\text{ch}} = \mathbb{1}/\dim \mathcal{H}$. Indeed, it follows from the fact that $\Theta \circ \mathcal{L}_{(\alpha)}^* = \mathcal{L}_{(1-\alpha)} \circ \Theta$ (see the proof of Theorem 3.1) that $e_{\rho_{\text{ch}},t}(1 - \alpha) = e_{\rho_{\text{ch}},t}(\alpha)$ for all $\alpha \in \mathbb{R}^M$ and all $t \geq 0$.

Remark 2 Property (8) is a consequence of energy conservation. It was first proposed by Andrieux et al. in the framework of Hamiltonian dynamics on the basis of a formal calculation [2].

We shall call the probability measure P_ρ^t the *entropic full counting statistics* (EFCS) of the QDS generated by $\mathcal{L}$ (w.r.t. the specific decomposition $\mathcal{L} = \sum_j \mathcal{L}_j$). As explained in Sect. 5, in cases where this QDS arises as a weak coupling limit of the dynamics of a system $\mathcal{S}$ coupled to M thermal reservoirs, the EFCS is the scaling limit of a measure $\mathbb{P}_\rho^t$ which describes the mean rate of entropy exchange between the system and the M reservoirs during the time interval $[0, t]$ (see Eq. (38) below).

An alternative interpretation of the measures P_ρ^t is based on the well-known unraveling technique. In other words, these measures can be understood in terms of a classical stochastic process which provides a coarse grained description of the dynamics of the system by so called quantum trajectories. Within this framework, P_ρ^t is the joint distribution of M random variables which describe the exchange of entropy between the system and the M reservoirs (see Sect. 6).

3.3 Entropic Fluctuations

As a direct consequence of Theorem 3.1 and the Gärtner-Ellis theorem (see, e.g., [25, 29]), we have

Corollary 3.2 *Assume that Hypotheses* (DB) *and* (ER) *hold and let*

$$I(\varsigma) = - \inf_{\alpha \in \mathbb{R}^M} \big(\alpha \cdot \varsigma + e(\alpha) \big).$$

$I(\varsigma)$ *is the Fenchel-Legendre transform of* $e(-\alpha)$. *Then*:

(1) $I(\varsigma)$ *takes values in* $[0, \infty]$ *and is a convex lower-semicontinuous function with compact level sets.*[3]
(2) $I(\varsigma) = 0$ *iff* $\varsigma = \overline{\varsigma}$, *where* $\overline{\varsigma} = -\nabla e(0)$. *Moreover, for any* $\epsilon > 0$ *there exists a positive constant* $a(\epsilon)$ *such that*

$$P_\rho^t \big(\{ \varsigma \in \mathbb{R}^M \mid |\varsigma - \overline{\varsigma}| \geq \epsilon \} \big) \leq e^{-ta(\epsilon)},$$

for all $t > 0$.
(3) *The family of measures* $\{P_\rho^t\}_{t \geq 0}$ *satisfies the large deviation principle with rate function* I. *More precisely, for any Borel set* $G \subset \mathbb{R}^M$ *we have*

$$- \inf_{\varsigma \in \mathrm{int}(G)} I(\varsigma) \leq \liminf_{t \to \infty} \frac{1}{t} \log P_\rho^t(G) \leq \limsup_{t \to \infty} \frac{1}{t} \log P_\rho^t(G) \leq - \inf_{\varsigma \in \mathrm{cl}(G)} I(\varsigma), \quad (9)$$

where $\mathrm{int}(G)$ *and* $\mathrm{cl}(G)$ *denote the interior and the closure of the set* G.
(4) *If Hypothesis* (TR) *is satisfied, then the rate function satisfies*

$$I(-\varsigma) = 1 \cdot \varsigma + I(\varsigma). \quad (10)$$

(5) *If Hypothesis* (KMS$_\beta$) *is satisfied, then* $I(\varsigma) = +\infty$ *for any* $\varsigma \in \mathbb{R}^M$ *such that* $\beta^{-1} \cdot \varsigma \neq 0$.

---

[3] The level sets of I are $\{\varsigma \mid I(\varsigma) \leq l\}$ where $l \in [0, \infty[$.

Remark 1 The components of $\overline{\varsigma} = (\overline{\varsigma}_1, \ldots, \overline{\varsigma}_M)$ describe the asymptotic rates of entropy transport between the system $\mathcal{S}$ and the M reservoirs constituting its environment. The nonnegative number

$$\sigma_+ = 1 \cdot \overline{\varsigma} = \sum_j \overline{\varsigma}_j,$$

is the steady state *entropy production rate* of a QDS satisfying Hypotheses (ER) and (DB) (see the next section for additional information about this important concept). If (TR) holds, then Relation (10) implies $I(-\overline{\varsigma}) = \sigma_+$ and $\sigma_+ > 0$ iff $\overline{\varsigma} \neq 0$.

Remark 2 The large deviation principle (9) quantifies the exponential rate of decay of the measures P_ρ^t away from the asymptotic mean value $\overline{\varsigma}$ and describes the statistics of the fluctuations of the rates of entropy transport over large but finite periods of time. In particular, (9) implies that

$$P_\rho^t\big(\big\{\varsigma \in \mathbb{R}^M \mid \varsigma \simeq \varphi\big\}\big) \simeq \mathrm{e}^{-tI(\varphi)},$$

for large t. Combining Parts (2) and (3) we derive that for large t,

$$\frac{P_\rho^t(\{\varsigma \in \mathbb{R}^M \mid \varsigma \simeq -\varphi\})}{P_\rho^t(\{\varsigma \in \mathbb{R}^M \mid \varsigma \simeq \varphi\})} \simeq \mathrm{e}^{-t1\cdot\varphi}, \tag{11}$$

and in particular that

$$\frac{P_\rho^t(\{\varsigma \in \mathbb{R}^M \mid \varsigma \simeq -\overline{\varsigma}\})}{P_\rho^t(\{\varsigma \in \mathbb{R}^M \mid \varsigma \simeq \overline{\varsigma}\})} \simeq \mathrm{e}^{-t\sigma_+}. \tag{12}$$

The identities (8) and (10), together with the resulting asymptotics (11) and (12), constitute *fluctuation relations* for a QDS out of equilibrium. One important feature of the fluctuation relations is universality (independence of the model).

Theorem 3.1 and Bryc's theorem (see Proposition 1 in [6] and Appendix A in [36]) imply the Central Limit Theorem for the family of measures $\{P_\rho^t\}_{t \geq 0}$.

Corollary 3.3 *Assume that Hypotheses (ER) and (DB) hold. Then for any Borel set $G \subset \mathbb{R}^M$,*

$$\lim_{t \to \infty} P_\rho^t\big(\big\{\varsigma \in \mathbb{R}^M \mid \sqrt{t}\big(\varsigma - \langle\varsigma\rangle_{\rho,t}\big) \in G\big\}\big) = \mu_D(G), \tag{13}$$

where μ_D denotes the centered Gaussian measure on $\mathbb{R}^M$ with covariance D given by

$$D_{ij} = \frac{\partial^2 e(\alpha)}{\partial \alpha_i \partial \alpha_j}\bigg|_{\alpha=0}.$$

Note that if Hypothesis (KMS$_\beta$) holds, then Theorem 3.1 (8) implies that the Gaussian measure μ_D has its support on the hyperplane $\beta^{-1} \cdot \varsigma = 0$. This is of course related to Part (5) of Corollary 3.2 and to energy conservation.

3.4 Thermodynamics

The von Neumann entropy of a state ρ is $\text{Ent}(\rho) = -\text{tr}(\rho \log \rho)$ and we shall call $S = -\log \rho$ the entropy observable associated to ρ. The relative entropy of a state ν w.r.t. to another state μ is

$$\text{Ent}(\nu|\mu) = \begin{cases} \text{tr}(\nu(\log \mu - \log \nu)) & \text{if } \text{Ran}(\nu) \subset \text{Ran}(\mu); \\ -\infty & \text{otherwise.} \end{cases}$$

We refer the reader to the monograph of Ohya and Petz [54] for further information on these fundamental concepts. Following Lebowitz and Spohn [48, 60], we define the entropy production in the state ρ of a QDS $\{e^{t\mathcal{L}}\}_{t \geq 0}$ satisfying Hypothesis (DB) by[4]

$$\sigma(\rho) = \frac{d}{dt} \sum_{j=1}^{M} \text{Ent}\big(e^{t\mathcal{L}_j^*}(\rho)\big|\rho_j\big)\big|_{t=0}. \tag{14}$$

We recall basic properties of the entropy production established in [48, 60].

(a) Since ρ_j is a steady state of the QDS generated by $\mathcal{L}_j$, we have

$$\text{Ent}\big(e^{t\mathcal{L}_j^*}(\rho)\big|\rho_j\big) = \text{Ent}\big(e^{t\mathcal{L}_j^*}(\rho)\big|e^{t\mathcal{L}_j^*}(\rho_j)\big),$$

and Uhlman's monotonicity theorem ([62], see also [36, 54]) implies that the r.h.s. of this identity is a non-decreasing function of t. Hence,

$$\sigma(\rho) \geq 0.$$

(b) An application of a theorem of Lieb [45] gives that the map $\mathfrak{S} \ni \rho \mapsto \sigma(\rho)$ is convex (see Theorem 3 in [60]).
(c) Set $S_j = -\log \rho_j$ and $\mathcal{I}_j = \mathcal{L}_j(S_j)$. An immediate consequence of (14) is the *entropy balance equation*:

$$\frac{d}{dt}\text{Ent}\big(e^{t\mathcal{L}^*}(\rho)\big)\big|_{t=0} = \sigma(\rho) + \sum_{j=1}^{M} \rho(\mathcal{I}_j). \tag{15}$$

The second term on the r.h.s. of Eq. (15) describes the flux of entropy entering the system. Thus, we can interpret the observable $\mathcal{I}_j$ as the entropy flux out of the jth reservoir. Note that if ρ is a steady state, then the l.h.s. of (15) vanishes, and the entropy balance equation takes the form

$$\sigma(\rho) = -\sum_{j=1}^{M} \rho(\mathcal{I}_j). \tag{16}$$

Our next result links the function $e(\alpha)$ to the observables S_j and $\mathcal{I}_j$.

Theorem 3.4 *Let $\{e^{t\mathcal{L}}\}_{t \geq 0}$ be a QDS satisfying Hypotheses (ER) and (DB). Set $\mathcal{J}_j = \mathcal{I}_j - \rho_+(\mathcal{I}_j)$. Then the following holds:*

---

[4] The derivative exists for all $\rho \in \mathfrak{S}$, see Theorem 3 in [60].

(1)

$$\left.\frac{\partial e(\alpha)}{\partial \alpha_j}\right|_{\alpha=0} = \rho_+(\mathcal{I}_j).$$

In particular,

$$\rho_+(\mathcal{I}_j) = -\lim_{t\to\infty}\langle \varsigma_j\rangle_{\rho,t} = -\overline{\varsigma}_j,$$

and $\sigma(\rho_+) = \sum_j \overline{\varsigma}_j.$

(2)

$$\left.\frac{\partial^2 e(\alpha)}{\partial \alpha_j \partial \alpha_k}\right|_{\alpha=0} = -\int_0^\infty \rho_+\big(\mathrm{e}^{t\mathcal{L}}(\mathcal{J}_j)\mathcal{J}_k^+ + \mathrm{e}^{t\mathcal{L}}(\mathcal{J}_k)\mathcal{J}_j^+\big)\mathrm{d}t$$

$$+ \int_0^\infty \rho_+\big(\mathcal{L}_k\big(\mathrm{e}^{t\mathcal{L}}(\mathcal{J}_j)S_k\big) + \mathcal{L}_j\big(\mathrm{e}^{t\mathcal{L}}(\mathcal{J}_k)S_j\big)\big)\mathrm{d}t + \delta_{jk}\rho_+\big(D_j(S_j, S_j)\big)$$

$$= \lim_{t\to\infty} t\big\langle \big(\varsigma_j - \langle\varsigma_j\rangle_{\rho,t}\big)\big(\varsigma_k - \langle\varsigma_k\rangle_{\rho,t}\big)\big\rangle_{\rho,t},$$

where $\mathcal{J}_j^+ = \mathcal{L}_j^{\rho_+}(S_j) = \mathcal{L}_j^*(S_j\rho_+)\rho_+^{-1}$ *and* $D_j(A, B) = \mathcal{L}_j(A^*B) - \mathcal{L}_j(A^*)B - A^*\mathcal{L}_j(B)$ *is the dissipation function of the* j*th Lindbladian.*

Remark 1 Under the assumptions of the theorem the semigroup $\{\mathrm{e}^{t\mathcal{L}}\}_{t\geq 0}$ is relaxing exponentially fast to ρ_+. Since $\rho_+(\mathcal{J}_j) = 0$, this implies that the operators $\mathrm{e}^{t\mathcal{L}}(\mathcal{J}_j)$ are exponentially decaying as $t \to \infty$, and so the time integrals in Part (2) are absolutely convergent.

Remark 2 We shall make use of Part (2) in Sect. 3.6 where we discuss linear response theory.

3.5 Energy Fluxes

The Hypothesis (KMS$_\beta$) allows us to relate entropy fluxes to energy fluxes by simple rescaling and to restate our main results in terms of energy transport. As a preparation for the discussion of the linear response theory, in this section we briefly discuss how this restating is carried out. Until the end of this section we shall assume that Hypotheses (ER), (DB), and (KMS$_\beta$) hold.

The observable describing the energy flux out of the jth reservoir is $\mathcal{F}_j = \mathcal{L}_j(H_\mathcal{S})$ (see [48]). Note that $\mathcal{I}_j = \beta_j \mathcal{F}_j$. If in addition (TR) holds, then $\Theta(H_\mathcal{S}) = H_\mathcal{S}$ and it follows from Parts (1) and (2) of Theorem 7.1 that $\mathcal{L}_j^{\rho_j}(H_\mathcal{S}) = \mathcal{L}_j(H_\mathcal{S})$. Hence,

$$\Theta(\mathcal{F}_j) = \mathcal{L}_j^{\rho_j}\big(\Theta(H_\mathcal{S})\big) = \mathcal{F}_j.$$

The steady state energy fluxes are

$$\overline{\phi}_j = \rho_+(\mathcal{F}_j).$$

Obviously, $\overline{\varsigma}_j = -\beta_j\overline{\phi}_j$, and Eq. (16) takes the form

$$\sigma(\rho_+) = -\sum_{j=1}^M \beta_j \overline{\phi}_j \geq 0. \tag{17}$$

This relation expresses the second law of thermodynamics for QDS satisfying our assumptions. The relation $\mathcal{L}^*(\rho_+) = 0$ yields the first law (conservation of energy):

$$\sum_{j=1}^{M} \overline{\phi}_j = 0. \tag{18}$$

The energetic full counting statistics of the system is the probability measure Q_ρ^t on $\mathbb{R}^M$ given by

$$Q_\rho^t(\phi) = P_\rho^t(-\beta\phi),$$

where $\beta\phi = (\beta_1\phi_1, \ldots, \beta_M\phi_M)$. In particular,

$$\mathrm{tr}\big(\rho e^{t\mathcal{L}(\alpha/\beta)}(\mathbb{1})\big) = \int_{\mathbb{R}^M} e^{t\alpha\cdot\phi} \mathrm{d}Q_\rho^t(\phi),$$

where $\alpha/\beta = (\alpha_1/\beta_1, \ldots, \alpha_M/\beta_M)$. Hence, for $\alpha \in \mathbb{R}^M$,

$$\chi(\alpha) = \lim_{t\to\infty} \frac{1}{t} \log \int_{\mathbb{R}^M} e^{t\alpha\cdot\phi} \mathrm{d}Q_\rho^t(\phi) = e(-\alpha/\beta),$$

and in particular,

$$\left.\frac{\partial\chi(\alpha)}{\partial\alpha_j}\right|_{\alpha=0} = \overline{\phi}_j, \qquad \left.\frac{\partial^2\chi(\alpha)}{\partial\alpha_j\partial\alpha_k}\right|_{\alpha=0} = \frac{1}{\beta_j\beta_k} \left.\frac{\partial^2 e(\alpha)}{\partial\alpha_j\partial\alpha_k}\right|_{\alpha=0}. \tag{19}$$

Note that the translation symmetry of $e(\alpha)$ (described in Part (8) of Theorem 3.1) implies that

$$\chi(\alpha) = \chi(\alpha + \lambda\mathbb{1}), \tag{20}$$

for all $\alpha \in \mathbb{R}^M$, $\lambda \in \mathbb{R}$. If (TRI) holds, then the Evans-Searles symmetry takes the form

$$\chi(\alpha) = \chi(-\beta - \alpha). \tag{21}$$

The large t fluctuations of Q_ρ^t are described by obvious reformulations of Corollaries 3.2 and 3.3.

Finally, we discuss briefly the equilibrium case where $\beta_j = \beta_0$ for $j = 1, \ldots, M$. In this case

$$\rho_j = \rho_0 = \frac{e^{-\beta_0 H_\mathcal{S}}}{\mathrm{tr}(e^{-\beta_0 H_\mathcal{S}})},$$

and $\mathcal{L}_j^*(\rho_0) = 0$ for all j. It follows that $\mathcal{L}^*(\rho_0) = 0$ and hence that $\rho_+ = \rho_0$ and $\overline{\phi}_j = 0$ for all j. Combining Parts (1) and (2) of Theorem 7.1 with Theorem 3.4 (2) one easily derives that $\mathcal{J}_j^+ = \mathcal{J}_j = \mathcal{I}_j = \beta_0\mathcal{F}_j$, and that

$$\left.\frac{\partial^2\chi(\alpha)}{\partial\alpha_j\partial\alpha_k}\right|_{\alpha=0} = -\int_0^\infty \rho_0\big(e^{t\mathcal{L}}(\mathcal{F}_j)\mathcal{F}_k + e^{t\mathcal{L}}(\mathcal{F}_k)\mathcal{F}_j\big)\mathrm{d}t + \delta_{jk}\rho_0\big(D_j(H_\mathcal{S}, H_\mathcal{S})\big). \tag{22}$$

If the pair $(\rho_0, \mathcal{L})$ is TRI, then

$$\rho_0\big(e^{t\mathcal{L}}(\mathcal{F}_j)\mathcal{F}_k\big) = \rho_0\big(\Theta\big(\mathcal{F}_k e^{t\mathcal{L}}(\mathcal{F}_j)\big)\big) = \rho_0\big(\mathcal{F}_k e^{t\mathcal{L}^{\rho_0}}(\mathcal{F}_j)\big) = \rho_0\big(e^{t\mathcal{L}}(\mathcal{F}_k)\mathcal{F}_j\big). \tag{23}$$

3.6 Linear Response Theory

Our last result concerns linear response to thermodynamical forces. We consider a small system $\mathcal{S}$ coupled to M thermal reservoirs $\mathcal{R}_j$ in equilibrium at inverse temperatures β_j where each β_j is close to some common equilibrium value $\beta_0 > 0$. The purpose of linear response theory is to study the behavior of various physical quantities to first order in the thermodynamical forces $\zeta_j = \beta_0 - \beta_j$. It is therefore natural to parametrize $\beta = (\beta_1, \ldots, \beta_M)$ by $\zeta = (\zeta_1, \ldots, \zeta_M)$ so that $\zeta = 0$ corresponds to the equilibrium situation $\beta = \beta_{eq} = (\beta_0, \ldots, \beta_0)$. The precise setup is as follows.

Let $(\mathcal{L}_\zeta)_{\zeta \in \mathcal{U}}$ be a family of Lindbladians indexed by an open neighborhood $\mathcal{U}$ of 0 in $\mathbb{R}^M$ and such that each $\mathcal{L}_\zeta$ satisfies Hypotheses (ER) and (TR). Moreover, we assume Hypotheses (DB) and (KMS$_\beta$) in the following form: for each $\zeta \in \mathcal{U}$,

$$\mathcal{L}_\zeta = \sum_{j=1}^M \mathcal{L}_{\zeta,j},$$

where $\mathcal{L}_{\zeta,j}$ depends only on ζ_j and satisfies the detailed balance condition w.r.t. the state

$$\rho_{\zeta_j} = \frac{e^{-(\beta_0 - \zeta_j)H_\mathcal{S}}}{\mathrm{tr}(e^{-(\beta_0 - \zeta_j)H_\mathcal{S}})},$$

for some ζ-independent self-adjoint $H_\mathcal{S} \in \mathcal{O}$. We shall also assume the following regularity in ζ:

(**RE**) The map $\zeta \mapsto \mathcal{L}_\zeta$ is continuously differentiable at $\zeta = 0$.

In what follows we shall indicate explicitly the dependence on ζ by writing $\mathcal{L}_{\zeta,(\alpha)}$, $e(\zeta, \alpha)$, $\chi(\zeta, \alpha)$, $\mathcal{F}_{\zeta,j}$, $\overline{\phi}_{\zeta,j}$, etc. Our assumptions imply that all partial derivatives of $\mathcal{L}_{\zeta,(\alpha)}$ w.r.t. α are continuously differentiable w.r.t. ζ at $\zeta = 0$.

For all $\alpha \in \mathbb{R}^M$ and $\zeta \in \mathcal{U}$, $e(\zeta, \alpha)$ is a simple eigenvalue of $\mathcal{L}_{\zeta,(\alpha)}$. The perturbation theory of isolated eigenvalues (see the proof of Theorem 3.4) implies that all partial derivatives of $e(\zeta, \alpha)$ w.r.t. α are also continuously differentiable w.r.t. ζ at $\zeta = 0$ and the same holds for the function $\chi(\zeta, \alpha)$. In particular, the maps $\zeta \mapsto \overline{\phi}_{\zeta,j}$ are continuously differentiable at $\zeta = 0$.

Combining (17) and (18) yields the following expressions of the first and second laws of thermodynamics

$$\sum_{j=1}^M \overline{\phi}_{\zeta,j} = 0, \qquad \sum_{j=1}^M \zeta_j \overline{\phi}_{\zeta,j} \geq 0.$$

The kinetic transport coefficients are defined by

$$L_{jk} = \left. \frac{\partial \overline{\phi}_{\zeta,j}}{\partial \zeta_k} \right|_{\zeta=0}.$$

It follows from the first law that

$$\sum_{j=1}^M L_{jk} = 0, \tag{24}$$

while the second law implies that the *real* quadratic form determined by the matrix $[L_{jk}]$ is positive definite.[5] It further follows from the first relation in (19) that

$$L_{jk} = \frac{\partial^2 \chi(\zeta, \alpha)}{\partial \zeta_k \partial \alpha_j}\bigg|_{\zeta=\alpha=0}.$$

In terms of the variable ζ, the Evans-Searles symmetry (21) takes the form $\chi(\zeta, \alpha) = \chi(\zeta, -\beta_{\text{eq}} + \zeta - \alpha)$, while the translation symmetry (20) reads $\chi(\zeta, \alpha) = \chi(\zeta, \alpha + \lambda 1)$. Since $\beta_{\text{eq}} = \beta_0 1$, combining these two symmetries we derive

$$\chi(\zeta, \alpha) = \chi(\zeta, \zeta - \alpha). \tag{25}$$

This relation and the chain rule (see Lemma 4.4 in [40]) yield

$$L_{jk} = \frac{\partial^2 \chi(\zeta, \alpha)}{\partial \zeta_k \partial \alpha_j}\bigg|_{\zeta=\alpha=0} = -\frac{1}{2}\frac{\partial^2 \chi(\zeta, \alpha)}{\partial \alpha_k \partial \alpha_j}\bigg|_{\zeta=\alpha=0}. \tag{26}$$

The equality of mixed partial derivatives $\partial_{\alpha_k}\partial_{\alpha_j}\chi = \partial_{\alpha_j}\partial_{\alpha_k}\chi$ implies the Onsager reciprocity relations $L_{jk} = L_{kj}$. Relations (22), (23), and Corollary 3.3 complete the linear response theory. We summarize:

Theorem 3.5 *Under the Hypotheses formulated at the beginning of this section the following statements hold.*

(1) *The Green-Kubo formulae*:

$$L_{jk} = \int_0^\infty \rho_0\big(\mathrm{e}^{t\mathcal{L}_0}(\mathcal{F}_{0,j})\mathcal{F}_{0,k}\big)\mathrm{d}t - \frac{1}{2}\delta_{jk}\rho_0\big(D_{0,j}(H_\mathcal{S}, H_\mathcal{S})\big),$$

   where $D_{0,j}$ denotes the dissipation function of $\mathcal{L}_{0,j}$.
(2) *The Onsager reciprocity relations*:

$$L_{jk} = L_{kj}.$$

(3) *The Fluctuation-Dissipation Theorem*: *for a state ρ on $\mathcal{O}$ let $Q^t_{\text{eq},\rho}$ be the energetic full counting statistics of the equilibrium system, i.e.,*

$$\text{tr}\big(\rho\mathrm{e}^{t\mathcal{L}_{0,(\alpha/\beta_0)}}(\mathbb{1})\big) = \int_{\mathbb{R}^M} \mathrm{e}^{t\alpha\cdot\phi}\mathrm{d}Q^t_{\text{eq},\rho}(\phi)$$

   and let $\langle\cdot\rangle_{\text{eq},\rho,t}$ denote the expectation w.r.t. the measure $Q^t_{\text{eq},\rho}$. For any Borel set $G \subset \mathbb{R}^M$,

$$\lim_{t\to\infty} Q^t_{\text{eq},\rho}\big(\{\phi \in \mathbb{R}^M \mid \sqrt{t}\big(\phi - \langle\phi\rangle_{\text{eq},\rho,t}\big) \in G\}\big) = \mu_D(G),$$

   where μ_D is the centered Gaussian measure on $\mathbb{R}^M$ with covariance D given by

$$D_{jk} = 2L_{jk}.$$

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[5]This does not imply that $L_{jk} = L_{kj}$.

Remark 1 Concerning the diagonal transport coefficients L_{jj}, the terms $\rho_0(D_{0,j}(H_S, H_S))$ are non-negative, and are strictly positive if S is effectively coupled to the jth-reservoir (see Sect. 4). Parts (1)–(2) of Theorem 7.1 imply that $\rho_0(D_{0,j}(H_S, H_S)) = -2\rho_0(H_S \mathcal{F}_{0,j})$.

Remark 2 In the absence of time-reversal, Part (3) holds with

$$D_{jk} = \int_0^\infty \rho_0\big(e^{t\mathcal{L}_0}(\mathcal{F}_{0,j})\mathcal{F}_{0,k} + e^{t\mathcal{L}_0}(\mathcal{F}_{0,k})\mathcal{F}_{0,j}\big)dt - \delta_{jk}\rho_0\big(D_{0,j}(H_S, H_S)\big).$$

Remark 3 Parts (1) and (2) of Theorem 3.5 were first proven in [48] by a different method. For comparison purposes we sketch the proof of [48]. Since $\mathcal{L}_{\zeta,k}^*(\rho_{\zeta k}) = 0$,

$$\frac{d\mathcal{L}_{\zeta,k}^*}{d\zeta_k}(\rho_{\zeta k}) = -\mathcal{L}_{\zeta,k}^*\left(\frac{d\rho_{\zeta k}}{d\zeta_k}\right) = \mathcal{L}_{\zeta,k}^*(H_S \rho_{\zeta k}) = \mathcal{F}_{\zeta,k}\rho_{\zeta k},$$

where the last equality follows from Parts (1) and (2) of Theorem 7.1. Hypotheses (ER) and (RE) imply that the map $\zeta \mapsto \rho_{\zeta,+}$ is continuously differentiable at $\zeta = 0$. Differentiating $\mathcal{L}_\zeta^*(\rho_{\zeta,+}) = 0$ w.r.t. ζ_k at $\zeta = 0$, we get

$$\frac{d\mathcal{L}_{\zeta,k}^*}{d\zeta_k}\bigg|_{\zeta=0}(\rho_0) = -\mathcal{L}_0^*\left(\frac{\partial\rho_{\zeta,+}}{\partial\zeta_k}\bigg|_{\zeta=0}\right).$$

The last two relations give

$$\mathcal{L}_0^*\left(\frac{\partial\rho_{\zeta,+}}{\partial\zeta_k}\bigg|_{\zeta=0}\right) = -\mathcal{F}_{0,k}\rho_0. \tag{27}$$

Since

$$\lim_{t\to\infty} e^{t\mathcal{L}_0^*}(\mathcal{F}_{0,k}\rho_0) = \rho_0(\mathcal{F}_{0,k})\rho_0 = 0, \tag{28}$$

the operators $e^{t\mathcal{L}_0^*}(\mathcal{F}_{0,k}\rho_0)$ are exponentially decaying as $t \to \infty$, and we deduce from (27) that there is a constant c such that

$$\frac{\partial\rho_{\zeta,+}}{\partial\zeta_k}\bigg|_{\zeta=0} = c\rho_0 + \int_0^\infty e^{t\mathcal{L}_0^*}(\mathcal{F}_{0,k}\rho_0)dt.$$

If $j \neq k$ then $\mathcal{F}_{\zeta,j} = \mathcal{L}_{\zeta,j}(H_S)$ does not depend on ζ_k and it follows that

$$L_{jk} = \text{tr}\left(\mathcal{F}_{0,j}\frac{\partial\rho_{\zeta,+}}{\partial\zeta_k}\bigg|_{\zeta=0}\right) = \int_0^\infty \rho_0\big(e^{t\mathcal{L}_0}(\mathcal{F}_{0,j})\mathcal{F}_{0,k}\big)dt.$$

The conservation law (24), the limit (28) and the last formula in Remark 1 yield

$$L_{kk} = -\sum_{j:j\neq k} L_{jk} = \int_0^\infty \rho_0\big(e^{t\mathcal{L}_0}(\mathcal{F}_{0,k})\mathcal{F}_{0,k} - e^{t\mathcal{L}_0}\big(\mathcal{L}_0(H_S)\big)\mathcal{F}_{0,k}\big)dt$$

$$= \int_0^\infty \rho_0\big(e^{t\mathcal{L}_0}(\mathcal{F}_{0,k})\mathcal{F}_{0,k}\big)dt - \int_0^\infty \frac{d}{dt}\text{tr}\big(e^{t\mathcal{L}_0^*}(\mathcal{F}_{0,k}\rho_0)H_S\big)dt$$

$$= \int_0^\infty \rho_0\big(e^{t\mathcal{L}_0}(\mathcal{F}_{0,k})\mathcal{F}_{0,k}\big)dt + \rho_0(H_{\mathcal{S}}\mathcal{F}_{0,k})$$

$$= \int_0^\infty \rho_0\big(e^{t\mathcal{L}_0}(\mathcal{F}_{0,k})\mathcal{F}_{0,k}\big)dt - \frac{1}{2}\rho_0\big(D_{0,k}(H_{\mathcal{S}}, H_{\mathcal{S}})\big).$$

Note that the above argument did not make use of Hypothesis (TR) and so Part (1) of Theorem 3.5 holds without time-reversal assumption (in fact, Lebowitz and Spohn do not discuss time-reversal at all in [48]). However, if the pair $(\rho_0, \mathcal{L}_0)$ is time-reversal invariant, then Part (1) and Relation (23) yield the Onsager reciprocity relations.

In contrast to the direct argument of [48], the proof described in this section exploits fundamentally the symmetry (25). The advantage of this derivation in context of a QDS out of equilibrium is conceptual. The fluctuation relations are structural model independent features of non-equilibrium statistical mechanics. As observed by Gallavotti [32], in the linear regime near equilibrium the fluctuation relations reduce to familiar fluctuation-dissipation formulae, and this structural model independent view of linear response theory is of fundamental conceptual importance (see [36, 40, 49] for a pedagogical discussion of this point). Our proof shows how a QDS out of equilibrium fit into this general picture and complements the derivation of [48] from the conceptual point of view.

4 Weakly Coupled Open Quantum Systems

We consider a small quantum system $\mathcal{S}$, described by the Hamiltonian $H_{\mathcal{S}}$ acting on the finite dimensional Hilbert space $\mathcal{H}_{\mathcal{S}}$. To induce a dissipative dynamics on $\mathcal{S}$, we couple this system to several infinitely extended thermal reservoirs $\mathcal{R}_1, \dots, \mathcal{R}_M$. Each reservoir $\mathcal{R}_j$ is initially in a thermal equilibrium state at inverse temperature $\beta_j > 0$.[6] By passing to the GNS representations induced by these states, each $\mathcal{R}_j$ is described by a Hilbert space $\mathcal{H}_j$, a W^*-algebra $\mathcal{O}_j \subset \mathcal{B}(\mathcal{H}_j)$ of observables, and a self-adjoint operator L_j (the Liouvillean) acting on $\mathcal{H}_j$, such that the Heisenberg dynamics $\tau_j^t(A) = e^{itL_j} A e^{-itL_j}$ leaves $\mathcal{O}_j$ invariant. The initial state of $\mathcal{R}_j$ is given by $\mathcal{O}_j \ni A \mapsto \omega_j(A) = \langle \xi_j | A \xi_j \rangle$, where $\xi_j \in \mathcal{H}_j$ is a unit vector such that $L_j \xi_j = 0$. Moreover, the state ω_j satisfies the KMS boundary condition: for all $A, B \in \mathcal{O}_j$,

$$\omega_j\big(A\tau_j^t(B)\big) = \omega_j\big(\tau_j^{t-i\beta_j}(B)A\big). \tag{29}$$

The Hilbert space of the joint system $\mathcal{S} + \mathcal{R}_1 + \dots + \mathcal{R}_M$ is $\mathcal{H} = \mathcal{H}_{\mathcal{S}} \otimes \mathcal{H}_1 \otimes \dots \otimes \mathcal{H}_M$ and we shall denote $H_{\mathcal{S}} \otimes \mathbb{1} \otimes \dots \otimes \mathbb{1}, \mathbb{1} \otimes H_1 \otimes \dots \otimes \mathbb{1}, \dots$ simply by $H_{\mathcal{S}}, H_1, \dots$.

The interaction between the system $\mathcal{S}$ and the reservoir $\mathcal{R}_j$ is described by the Hamiltonian

$$H_{\mathcal{S}\mathcal{R}_j} = \sum_{k=1}^{n_j} Q_j^{(k)} \otimes R_j^{(k)},$$

---

[6]Here, we could also consider conserved charges and introduce associated chemical potentials. We refrain to do so in order to keep notation as simple as possible.

where each $Q_j^{(k)}$ is a self-adjoint operator on $\mathcal{H}_{\mathcal{S}}$ and each $R_j^{(k)}$ is a self-adjoint element of $\mathcal{O}_j$ such that $\omega_j(R_j^{(k)}) = 0$.[7] The full Hamiltonian (more precisely the semi-standard Liouvillean in the terminology of [17]) of the coupled system is

$$L_\lambda = H_{\mathcal{S}} + \sum_{j=1}^{M} (L_j + \lambda H_{\mathcal{S}\mathcal{R}_j}),$$

where λ is a coupling constant. The effective dynamics of the system $\mathcal{S}$ is then defined by the family of linear map $\{\mathcal{T}_\lambda^t\}_{t \in \mathbb{R}}$ on $\mathcal{B}(\mathcal{H}_{\mathcal{S}})$ determined by

$$\langle \psi | \mathcal{T}_\lambda^t(X)\psi \rangle = \langle \psi \otimes \xi | e^{itL_\lambda}(X \otimes \mathbb{1})e^{-itL_\lambda}\psi \otimes \xi \rangle,$$

where $X \in \mathcal{B}(\mathcal{H}_{\mathcal{S}})$, $\psi \in \mathcal{H}_{\mathcal{S}}$, and $\xi = \xi_1 \otimes \cdots \otimes \xi_M$.

Except in trivial cases, $\{\mathcal{T}_\lambda^t\}_{t \geq 0}$ is not a semigroup. However, under appropriate conditions on the decay of the multi-time correlation functions $\omega_j(\tau_j^{t_1}(R_j^{(k_1)}) \cdots \tau_j^{t_n}(R_j^{(k_n)}))$, Davies has shown (see Theorem 2.3 in [10]) that there exists a Lindbladian $\mathcal{L}$ generating a QDS such that $\mathcal{L}$ commutes with $\mathcal{L}_{\mathcal{S}}(X) = i[H_{\mathcal{S}}, X]$, and

$$\lim_{\lambda \to 0} \sup_{\lambda^2 t \in I} \| \mathcal{T}_\lambda^t - e^{t(\mathcal{L}_{\mathcal{S}} + \lambda^2 \mathcal{L})} \| = 0,$$

holds for any compact interval $I = [0, \tau] \subset \mathbb{R}$. In other words, in the limit of small coupling $\lambda \to 0$ and for times of the order λ^{-2} the effective dynamics of $\mathcal{S}$ is well approximated by the quantum dynamical semigroup generated by $\mathcal{L}_{\mathcal{S}} + \lambda^2 \mathcal{L}$. This theory is well-known and we refer the reader to the in depth exposition of [15, 48] for further details. To write down the explicit form of the generator $\mathcal{L}$, we introduce the functions

$$h_j^{(kl)}(\omega) = \int_{-\infty}^{\infty} e^{-i\omega t} \langle \xi_j | R_j^{(k)} \tau_j^t(R_j^{(l)})\xi_j \rangle dt = 2\pi \langle R_j^{(k)}\xi_j | \delta(L_j - \omega)R_j^{(l)}\xi_j \rangle,$$

and their Hilbert transforms

$$s_j^{(kl)}(\omega) = \mathcal{P}\int_{-\infty}^{\infty} \frac{h_j^{(kl)}(\nu)}{\nu - \omega} \frac{d\nu}{2\pi} = \langle R_j^{(k)}\xi_j | \mathcal{P}(L_j - \omega)^{-1} R_j^{(l)}\xi_j \rangle,$$

where $\mathcal{P}$ denotes Cauchy's principal value (the hypotheses of the above mentioned theorem of Davies ensure the existence of these integrals). Note that the $n_j \times n_j$-matrices

$$h_j(\omega) = [h_j^{(kl)}(\omega)], \qquad s_j(\omega) = [s_j^{(kl)}(\omega)],$$

are respectively positive and self-adjoint and that the KMS condition (29) implies the relation

$$h_j^{(kl)}(-\omega) = e^{-\beta_j \omega} h_j^{(lk)}(\omega). \tag{30}$$

---

[7] In some models (like the spin-boson system) the operators $R_j^{(k)}$ are unbounded and only affiliated to the W^*-algebra $\mathcal{O}_j$. With some additional technicalities the discussions of this and the next three section easily extend to such cases, see any of the references [15, 17, 19, 41, 48].

We denote by P_μ the spectral projection of H_S associated to the eigenvalue $\mu \in \mathrm{sp}(H_S)$, and for

$$\omega \in \Omega = \big\{ \mu - \nu \,\big|\, \mu, \nu \in \mathrm{sp}(H_S) \big\},$$

we define

$$V_j^{(k)}(\omega) = \sum_{\mu - \nu = \omega} P_\nu Q_j^{(k)} P_\mu = V_j^{(k)*}(-\omega). \tag{31}$$

Obviously,

$$\mathrm{e}^{\alpha H_S} V_j^{(k)}(\omega) \mathrm{e}^{-\alpha H_S} = \mathrm{e}^{-\alpha \omega} V_j^{(k)}(\omega), \tag{32}$$

for all $\alpha \in \mathbb{C}$.

The generator $\mathcal{L}$ has the Lindblad form (5), with the self-adjoint operator T given by

$$T = \sum_{j=1}^{M} T_j, \quad T_j = \sum_{k,l=1}^{n_j} \sum_{\omega \in \Omega} s_j^{(kl)}(\omega) V_j^{(k)*}(\omega) V_j^{(l)}(\omega),$$

and the CP map Φ given by

$$\Phi(X) = \sum_{j=1}^{M} \Phi_j(X), \quad \Phi_j(X) = \sum_{k,l=1}^{n_j} \sum_{\omega \in \Omega} h_j^{(kl)}(\omega) V_j^{(k)*}(\omega) X V_j^{(l)}(\omega).$$

A Kraus decomposition of Φ_j is constructed as follows. Denote by $u_j(\omega) = [u_j^{(kl)}(\omega)]$ a unitary matrix which diagonalize the positive matrix $h_j(\omega)$,

$$u_j(\omega)^* h_j(\omega) u_j(\omega) = \big[\delta_{kl} g_j^{(k)}(\omega) \big].$$

Setting $W_j^{(k)}(\omega) = \sqrt{g_j^{(k)}(\omega)} \sum_l u_j^{(kl)}(\omega) V_j^{(l)}(\omega)$, we obtain

$$\Phi_j(X) = \sum_{k=1}^{n_j} \sum_{\omega \in \Omega} W_j^{(k)*}(\omega) X W_j^{(k)}(\omega).$$

Note that $\mathcal{L}$ can be written as the sum of the Lindbladians

$$\mathcal{L}_j(X) = \mathrm{i}[T_j, X] - \frac{1}{2} \big\{ \Phi_j(\mathbb{1}), X \big\} + \Phi_j(X),$$

where $\mathcal{L}_j$ describes the interaction of the small system $\mathcal{S}$ with a single reservoir $\mathcal{R}_j$. Using (30) and (32) one easily verifies that $\mathcal{L}_j$ satisfies the detailed balance condition w.r.t. the faithful state

$$\rho_j = \frac{\mathrm{e}^{-\beta_j H_S}}{\mathrm{tr}(\mathrm{e}^{-\beta_j H_S})}. \tag{33}$$

Thus, Hypotheses (DB) and (KMS$_\beta$) are automatically satisfied by the weak coupling Lindbladian $\mathcal{L}$.

Regarding time-reversibility, assuming that

(a) each reservoir is time-reversal invariant, i.e., there exists antiunitary involution θ_j acting on $\mathcal{H}_j$ such that $L_j \theta_j = \theta_j L_j$ and $\theta_j \xi_j = \xi_j$;

(b) the small system $\mathcal{S}$ is time-reversal invariant, i.e., there is an antiunitary involution $\theta_\mathcal{S}$ on $\mathcal{H}_\mathcal{S}$ such that $\theta_\mathcal{S} H_\mathcal{S} = H_\mathcal{S} \theta_\mathcal{S}$;

(c) $\theta_j R_j^{(k)} = R_j^{(k)} \theta_j$ and $\theta_\mathcal{S} Q_j^{(k)} = Q_j^{(k)} \theta_\mathcal{S}$ for all j, k,

we easily conclude that $h_j^{(kl)}(\omega) = \overline{h_j^{(lk)}(\omega)}$, $s_j^{(kl)}(\omega) = \overline{s_j^{(lk)}(\omega)}$, and $\theta_\mathcal{S} V_j^{(k)}(\omega) = V_j^{(k)}(\omega)\theta_\mathcal{S}$. It immediately follows that $\theta_\mathcal{S} T_j = T_j \theta_\mathcal{S}$ and $\Phi_j(\theta_\mathcal{S} X \theta_\mathcal{S}) = \theta_\mathcal{S} \Phi_j(X)\theta_\mathcal{S}$. Hence, Hypothesis (TR) is satisfied with $\Theta(X) = \theta_\mathcal{S} X \theta_\mathcal{S}$.

We now turn to the ergodicity Hypothesis (ER). Clearly, $\{Q_j^{(k)}\}'_{j,k} \cap \{H_\mathcal{S}\}' \subset \operatorname{Ker}\mathcal{L}$ and the condition

$$\left\{Q_j^{(k)}\right\}'_{j,k} \cap \{H_\mathcal{S}\}' = \mathbb{C}\mathbb{1}, \tag{34}$$

is obviously necessary for (ER) to hold. On the other hand, assuming that the matrices $h_j(\omega)$ are strictly positive for all $1 \leq j \leq M$ and $\omega \in \Omega$, the construction of the Kraus family $\{W_j^{(k)}(\omega)\}_{j,k,\omega}$ shows that its linear span coincides with the linear span of the family $\mathcal{V} = \{V_j^{(k)}(\omega)\}_{j,k,\omega}$. By Eq. (31), the family $\mathcal{V}$ is self-adjoint, and von Neumann's bicommutant theorem implies that the smallest subalgebra of $\mathcal{O}$ containing $\mathcal{V}$ is the bicommutant $\mathcal{V}''$. As shown by Spohn (see Theorem 3 in [59]), the condition $\mathcal{V}'' = \mathcal{O}$ is equivalent to (34). Hence, assuming strict positivity of the matrices $h_j(\omega)$ for all j and ω, Theorems 2.1 and 2.3 imply that the Spohn condition (34) is also sufficient for Hypothesis (ER) to hold.

Note that

$$\sigma(\rho) = \sum_{j=1}^{M} \sigma_j(\rho),$$

where $\sigma_j(\rho)$ is the entropy production of the system $\mathcal{S}$ interacting only with the reservoir $\mathcal{R}_j$ via the Lindbladian $\mathcal{L}_j$. If the matrix $h_j(\omega)$ is strictly positive and

$$\left\{Q_j^{(k)}\right\}'_k \cap \{H_\mathcal{S}\}' = \mathbb{C}\mathbb{1}, \tag{35}$$

then, as discussed above, the QDS $\{e^{t\mathcal{L}_j}\}_{t\geq 0}$ is positivity improving. Moreover, $\mathcal{L}_j^*(\rho) = 0$ iff $\rho = e^{-\beta_j H_\mathcal{S}}/\operatorname{tr}(e^{-\beta_j H_\mathcal{S}})$.[8] Hence, we arrive at the following elegant condition (see [5, 48]) which ensures that $\sigma(\rho) > 0$ for all states ρ: there exists a pair j_1, j_2 such that $\beta_{j_1} \neq \beta_{j_2}$, the relation (35) holds for $j = j_1, j_2$, and the matrix $h_j(\omega)$ is strictly positive for all ω and $j = j_1, j_2$.

In conclusion, under very general and natural conditions the class of weak coupling limit QDS introduced in [48] satisfies Hypotheses (ER), (DB), (TR), (KMS$_\beta$), and has strictly positive entropy production.[9] Starting with the seminal paper [48], such semigroups have been one of the basic paradigms of non-equilibrium quantum statistical mechanics.

5 Full Counting Statistics

In this section, we elucidate the physical meaning of the measure P_ρ^t introduced in Theorem 3.1 in cases where the Lindbladian $\mathcal{L}$ describes a weakly coupled open quantum system

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[8]The same conditions ensure that the terms $\rho_{\beta_0}(D_j(H_\mathcal{S}, H_\mathcal{S}))$ in Theorem 3.5(1) are strictly positive, providing of course that $H_\mathcal{S} \notin \mathbb{C}\mathbb{1}$.

[9]At the current level of generality, the verification of Hypothesis (RE) requires supplementing Davies' conditions with additional regularity assumptions which we shall not discuss for reasons of space. In practice, i.e. in the context of concrete models, the verification of (RE) is typically an easy exercise.

as discussed in the preceding section. We shall keep our presentation at a formal level; the interested reader should consult Sect. 5 of [36] for a more detailed discussion as well as [14, 20, 21, 41] for a mathematically rigorous treatment of some specific models.

We start with the open system described in Sect. 4, but we assume now that the reservoirs $\mathcal{R}_j$ are confined to finite boxes. More precisely, working in the Schrödinger representation, we assume that the reservoir Hamiltonians H_j have purely discrete spectrum and that the operators $\mathrm{e}^{-\beta H_j}$ are trace class for all $\beta > 0$. The initial state of the combined system is $\rho = \rho_S \otimes \rho_\mathcal{R}$, where

$$\rho_\mathcal{R} = \rho_{\mathcal{R}_1} \otimes \cdots \otimes \rho_{\mathcal{R}_M}, \quad \rho_{\mathcal{R}_j} = \frac{\mathrm{e}^{-\beta_j H_j}}{\mathrm{tr}(\mathrm{e}^{-\beta_j H_j})},$$

and ρ_S is the initial state of the small system S.

The full counting statistics of the entropy fluxes across the system S is defined as follows. Set $S = (S_1, \ldots, S_M)$ with $S_j = \beta_j H_j$. The observables S_j commute and hence can be simultaneously measured. Let Π_s denote the joint spectral projection of S associated to the eigenvalue $s \in \mathrm{sp}(S)$. Two successive measurements of S at time t_0 and at time $t_0 + t$ are described by the positive map valued measure (PMVM) (see, e.g., [13]) which, to any subset $A \in \mathrm{sp}(S) \times \mathrm{sp}(S)$, associate the CP map

$$\mathcal{E}_A(X) = \sum_{(s,s') \in A} \Pi_{s'} \mathrm{e}^{-\mathrm{i}t H_\lambda} \Pi_s X \Pi_s \mathrm{e}^{\mathrm{i}t H_\lambda} \Pi_{s'}.$$

Indeed, if ρ_{t_0} denotes the state of the system at time t_0, one easily checks that, according to the usual rules of projective measurements,

$$\mathrm{tr}\, \mathcal{E}_{\{(s,s')\}}(\rho_{t_0}),$$

is the joint probability for the first measurement to yield the result s and for the second one to yield the result s'. Hence, the probability distribution of $\varsigma = (s' - s)/t$, the mean rate of entropy transport from the system S to the M reservoirs over the time interval $[0, t]$, is given in terms of the initial state ρ_S by the formula

$$\mathbb{P}^t_{\rho_S}(\varsigma) = \mathrm{tr}\, \mathcal{E}_{\{s' - s = t\varsigma\}}(\rho_S \otimes \rho_\mathcal{R}).$$

The atomic probability measure $\mathbb{P}^t_{\rho_S}$ on $\mathbb{R}^M$ is the *full counting statistics* of the energy/entropy flow. An elementary calculation shows that the Laplace transform of this measure is given by

$$\ell^t_{\rho_S}(\alpha) = \int_{\mathbb{R}^M} \mathrm{e}^{-t\alpha \cdot \varsigma} \mathrm{d}\mathbb{P}^t_{\rho_S}(\varsigma) = \mathrm{tr}\left((\rho_S \otimes \rho_\mathcal{R}) \rho_\mathcal{R}^{-\alpha} \mathrm{e}^{\mathrm{i}t H_\lambda} \rho_\mathcal{R}^\alpha \mathrm{e}^{-\mathrm{i}t H_\lambda}\right),$$

where, for $\alpha = (\alpha_1, \ldots, \alpha_M) \in \mathbb{R}^M$, we have set

$$\rho_\mathcal{R}^\alpha = \mathbb{1} \otimes \rho_{\mathcal{R}_1}^{\alpha_1} \otimes \cdots \otimes \rho_{\mathcal{R}_M}^{\alpha_M}.$$

Assuming that the operators

$$\tau_j^{\mathrm{i}s\beta_j/2}\left(R_j^{(k)}\right) = \mathrm{e}^{-s\beta_j H_j/2} R_j^{(k)} \mathrm{e}^{s\beta_j H_j/2},$$

are entire analytic functions of s, we can define the deformed Hamiltonian

$$H_{\lambda,\alpha} = \rho_{\mathcal{R}}^{\alpha/2} H_\lambda \rho_{\mathcal{R}}^{-\alpha/2} = H_{\mathcal{S}} + \sum_{j=1}^{M} \left(H_j + \lambda^2 \sum_{k=1}^{n_j} Q_j^{(k)} \otimes \tau_j^{\mathrm{i}\alpha_j\beta_j/2}\big(R_j^{(k)}\big) \right),$$

and write

$$\ell_{\rho_{\mathcal{S}}}^t(\alpha) = \mathrm{tr}\big((\rho_{\mathcal{S}} \otimes \rho_{\mathcal{R}})\mathrm{e}^{\mathrm{i}t H_{\lambda,\alpha}^*} \mathbb{1} \mathrm{e}^{-\mathrm{i}t H_{\lambda,\alpha}}\big). \tag{36}$$

At this point, one can pass to the GNS representation of the reservoirs and perform a thermodynamic limit, letting the size of the confining boxes become infinite. If the deformed operators $\tau_j^{\mathrm{i}\alpha_j\beta_j/2}(R_j^{(k)})$ remain well defined elements of the W^*-algebras $\mathcal{O}_j$ in this limit, then we can define the effective deformed dynamics of the open system with infinitely extended reservoirs

$$\langle\psi|\mathcal{T}_{\lambda,\alpha}^t(X)\psi\rangle = \langle\psi \otimes \xi|\mathrm{e}^{\mathrm{i}t L_{\lambda,\alpha}^*}(X \otimes \mathbb{1})\mathrm{e}^{-\mathrm{i}t L_{\lambda,\alpha}}\psi \otimes \xi\rangle, \tag{37}$$

with the deformed semi-standard Liouvillean

$$L_{\lambda,\alpha} = H_{\mathcal{S}} + \sum_{j=1}^{M} \left(L_j + \sum_{k=1}^{n_j} Q_j^{(k)} \otimes \tau_j^{\mathrm{i}\alpha_j\beta_j/2}\big(R_j^{(k)}\big) \right).$$

Assuming that the thermodynamic limit

$$\mathrm{TD}-\lim \mathrm{tr}\big((|\psi\rangle\langle\psi| \otimes \rho_{\mathcal{R}})\,\mathrm{e}^{\mathrm{i}t H_{\lambda,\alpha}^*}(X \otimes \mathbb{1})\mathrm{e}^{-\mathrm{i}t H_{\lambda,\alpha}}\big) = \langle\psi|\mathcal{T}_{\lambda,\alpha}^t(X)\psi\rangle,$$

exists for any $\psi \in \mathcal{H}_{\mathcal{S}}$, $X \in \mathcal{B}(\mathcal{H}_{\mathcal{S}})$, and $\alpha \in \mathbb{R}^M$, we conclude that the Laplace transform $\ell_{\rho_{\mathcal{S}}}^t(\alpha)$ of the full counting statistics $\mathbb{P}_{\rho_{\mathcal{S}}}^t$ has a well defined thermodynamic limit

$$\mathrm{TD}-\lim \ell_{\rho_{\mathcal{S}}}^t(\alpha) = \mathrm{tr}\big(\rho_{\mathcal{S}}\mathcal{T}_{\lambda,\alpha}^t(\mathbb{1})\big),$$

for all $\alpha \in \mathbb{R}^M$. Then one can show that, as the size of the reservoir increases, the full counting statistics $\mathbb{P}_{\rho_{\mathcal{S}}}^t$ converges weakly to a Borel probability measure which we again denote by $\mathbb{P}_{\rho_{\mathcal{S}}}^t$ which satisfies

$$\int_{\mathbb{R}^M} \mathrm{e}^{-t\alpha\cdot\varsigma}\mathrm{d}\mathbb{P}_{\rho_{\mathcal{S}}}^t(\varsigma) = \mathrm{tr}\big(\rho_{\mathcal{S}}\mathcal{T}_{\lambda,\alpha}^t(\mathbb{1})\big),$$

(see Proposition 4.1 in [37]). We call the limiting measure $\mathbb{P}_{\rho_{\mathcal{S}}}^t$ the *full counting statistics* of the open system $\mathcal{S}$ coupled to the infinitely extended reservoirs $\mathcal{R}_1, \ldots, \mathcal{R}_M$. Note that since infinitely extended reservoirs have an infinite energy, it is not possible to implement directly the successive measurement procedure we have described to this model, and that one is forced to invoke the thermodynamic limit to construct its full counting statistics.

Applying the Davies procedure to extract the weak coupling limit of the deformed effective dynamics leads to

$$\lim_{\lambda\to 0} \sup_{\lambda^2 t \in I} \big\| \mathcal{T}_{\lambda,\alpha}^t - \mathrm{e}^{t(\mathcal{L}_{\mathcal{S}}+\lambda^2\mathcal{K}_{(\alpha)})} \big\| = 0,$$

where $\mathcal{K}_{(\alpha)}$ is a deformed generator commuting with $\mathcal{L}_{\mathcal{S}}$. An explicit calculation shows that the only difference between $\mathcal{K}_{(\alpha)}$ and the undeformed Lindbladian $\mathcal{L} = \mathcal{K}_0$ is that the

functions $h_j^{(kl)}$ are replaced with (recall that $L_j \xi_j = 0$),

$$
\begin{aligned}
h_{j,\alpha_j}^{(kl)}(\omega) &= 2\pi \langle \tau_j^{i\alpha_j\beta_j/2} (R_j^{(k)}) \xi_j | \delta(L_j - \omega) \tau_j^{i\alpha_j\beta_j/2} (R_j^{(l)}) \xi_j \rangle \\
&= 2\pi \langle e^{-\alpha_j\beta_j L_j/2} R_j^{(k)} \xi_j | \delta(L_j - \omega) e^{-\alpha_j\beta_j L_j/2} R_j^{(l)} \xi_j \rangle \\
&= e^{-\alpha_j\beta_j\omega} h_j^{(kl)}(\omega).
\end{aligned}
$$

Using Eq. (32), one finally concludes that, with the ρ_j defined in Eq. (33),

$$
\mathcal{K}_{(\alpha)}(X) = \sum_{j=1}^{M} \mathcal{L}_j \big(X\rho_j^{-\alpha_j} \big) \rho_j^{\alpha_j},
$$

and so $\mathcal{K}_{(\alpha)}$ coincides with the deformed Lindbladian $\mathcal{L}_{(\alpha)}$. We conclude that if $[H_{\mathcal{S}}, \rho_{\mathcal{S}}] = 0$, then the measure P_ρ^t introduced in Theorem 3.1 is related to the full counting statistics $\mathbb{P}_{\rho_{\mathcal{S}}}^t$ through the scaling limit

$$
\lim_{\lambda \to 0} \int f\big(\lambda^{-2}\varsigma\big) d\mathbb{P}_{\rho_{\mathcal{S}}}^{t/\lambda^2}(\varsigma) = \int f(\varsigma) dP_{\rho_{\mathcal{S}}}^t(\varsigma). \tag{38}
$$

We note that in the weak coupling regime, the energy/entropy fluxes are of order λ^2 so the scaling $\lambda^{-2}\varsigma$ which appears on the left hand side of the last identity is natural. The measure $P_{\rho_{\mathcal{S}}}^t$ thus describes the rescaled mean energy/entropy fluxes at the Van Hove time scale t/λ^2. To the best of our knowledge, this observation is due to de Roeck [18].

For some specific models it is possible to show that

$$
\lim_{\lambda \to 0} \sup_{t>0} \big\| \mathcal{T}_{\lambda,\alpha}^t - e^{t(\mathcal{L}_{\mathcal{S}} + \lambda^2 \mathcal{K}_\alpha)} \big\| = 0,
$$

(see [19–21, 41]), and in such cases one can relate the large deviation principle of Corollary 3.2 to the large deviation principle of the full counting statistics $\mathbb{P}_{\rho_{\mathcal{S}}}^t$.

The link between full counting statistics and deformations of the semi-standard Liouvillean (relations (36) and (37)) goes back to [18]. The link between full counting statistics and deformations of the standard Liouvillean can be traced back to [16, 38, 53], was fully elaborated in [36], and plays the key role in the work [41]. The second link relates full counting statistics to modular theory of operator algebras and deformed Lindbladians $\mathcal{L}_{(\alpha)}$ to Fermi Golden Rule for spectral resonances of the deformed standard Liouvilleans. This point is discussed in detail in [41] and we refer the reader to this work for additional information.

6 Unraveling of the Deformed Semigroup $e^{t\mathcal{L}_{(\alpha)}}$

In this section we follow [22] and present an alternative and more general interpretation of the measure P_ρ^t based on the standard unraveling technique. As a byproduct of this construction, we shall get a proof of Parts (1) and (2) of Theorem 3.1. We shall assume that Hypothesis (DB) holds throughout the section and use the elementary properties of Lindbladians summarized in Theorem 7.1.

Let $\mathcal{L}_j(X) = i[T_j, X] - \frac{1}{2}\{\Phi_j(\mathbb{1}), X\} + \Phi_j(X)$ denote a Lindblad decomposition of $\mathcal{L}_j$ and set

$$
\mathcal{K}(X) = -K^*X - XK, \quad K = \sum_{j=1}^{M} K_j, \quad K_j = \frac{1}{2}\Phi_j(\mathbb{1}) + iT_j.
$$

By Theorem 7.1, K_j commutes with ρ_j and Φ_j admits a decomposition

$$\Phi_j = \sum_{\omega \in \Omega_j} \Phi_{j,\omega},$$

where $\Omega_j = \{\mu - \nu \mid \mu, \nu \in \mathrm{sp}(\log \rho_j)\}$ and $\Phi_{j,\omega} \in \mathrm{CP}(\mathcal{O})$ satisfies $\Phi_{j,\omega}(X\rho_j^{-\alpha_j})\rho_j^{\alpha_j} = \mathrm{e}^{-\alpha_j \omega}\Phi_{j,\omega}(X)$. It follows that

$$\mathcal{L}_{(\alpha)} = \mathcal{K} + \sum_{j=1}^{M} \sum_{\omega \in \Omega_j} \mathrm{e}^{-\alpha_j \omega}\Phi_{j,\omega}, \tag{39}$$

is of the Lindblad form (4) for $\alpha \in \mathbb{R}^M$, which proves Part (1) of Theorem 3.1. Using the Dyson expansion of the cocycle $\Gamma_\alpha^t = \mathrm{e}^{-t\mathcal{K}} \circ \mathrm{e}^{t\mathcal{L}_{(\alpha)}}$, we obtain the representation

$$
\begin{aligned}
\langle \rho | \mathrm{e}^{t\mathcal{L}_{(\alpha)}}(\mathbb{1}) \rangle &= \langle \mathrm{e}^{t\mathcal{K}^*}(\rho) | \Gamma_\alpha^t(\mathbb{1}) \rangle = \langle \rho_t | \mathbb{1} \rangle \\
&+ \sum_{N \geq 1} \sum_{\substack{(j_1,\ldots,j_N) \in \{1,\ldots,M\}^N \\ (\omega_1,\ldots,\omega_N) \in \Omega_{j_1} \times \cdots \times \Omega_{j_N}}} \mathrm{e}^{-\sum_{k=1}^{N} \alpha_{j_k}\omega_k} \\
&\times \int_{0 \leq s_1 \leq \cdots \leq s_N \leq t} \langle \rho_t | \Phi_{j_N,\omega_N,s_N} \circ \cdots \circ \Phi_{j_1,\omega_1,s_1}(\mathbb{1}) \rangle \mathrm{d}s_1 \cdots \mathrm{d}s_N, \tag{40}
\end{aligned}
$$

where $\rho_t = \mathrm{e}^{t\mathcal{K}^*}(\rho)$ and $\Phi_{j,\omega,s} = \mathrm{e}^{-s\mathcal{K}} \circ \Phi_{j,\omega} \circ \mathrm{e}^{s\mathcal{K}}$.

Unraveling consists of rewriting this expression in terms of a probability measure μ_ρ^t on a set Ξ^t of quantum trajectories defined as follows. For $N \geq 1$, let

$$
\begin{aligned}
\Xi_N^t = \big\{ \xi = [\xi_1, \ldots, \xi_N] \,\big|\, \xi_k = (j_k, \omega_k, s_k), \; j_k \in \{1, \ldots, M\}, \\
\omega_k \in \Omega_{j_k}, 0 \leq s_1 \leq \cdots \leq s_N \leq t \big\},
\end{aligned}
$$

and set $\Xi_0^t = \{\emptyset\}$. On the disjoint union

$$\Xi^t = \bigsqcup_{N \geq 0} \Xi_N^t,$$

one defines the positive measure μ_ρ^t by

$$
\begin{aligned}
\int_{\Xi^t} f(\xi) \, \mathrm{d}\mu_\rho^t(\xi) = f(\{\emptyset\}) \langle \rho_t | \mathbb{1} \rangle + \sum_{N \geq 1} \sum_{\substack{(j_1,\ldots,j_N) \in \{1,\ldots,M\}^N \\ (\omega_1,\ldots,\omega_N) \in \Omega_{j_1} \times \cdots \times \Omega_{j_N}}} \\
\int_{0 \leq s_1 \leq \cdots \leq s_N \leq t} f\big([(j_1,\omega_1,s_1), \ldots, (j_N,\omega_N,s_N)]\big) \\
\times \langle \rho_t | \Phi_{j_N,\omega_N,s_N} \circ \cdots \circ \Phi_{j_1,\omega_1,s_1}(\mathbb{1}) \rangle \mathrm{d}s_1 \cdots \mathrm{d}s_N.
\end{aligned}
$$

Comparison with Eq. (40) shows that

$$\int_{\Xi^t} \mathrm{d}\mu_\rho^t(\xi) = \langle \rho_t | \Gamma_0^t(\mathbb{1}) \rangle = \langle \rho | \mathrm{e}^{t\mathcal{L}}(\mathbb{1}) \rangle = 1,$$

and hence μ_ρ^t is a probability measure. An element $\xi \in \Xi^t$ is a quantum trajectory which represent the history of the system during the time interval $[0, t]$. Observe that the system can exchange entropy with the reservoir $\mathcal{R}_j$ only in quanta of the form $\omega \in \mathrm{sp}(S_j) - \mathrm{sp}(S_j) = \Omega_j$ where $S_j = -\log \rho_j$. An element $\xi = [\xi_1, \ldots, \xi_N]$ of Ξ^t is a chronologically ordered list of elementary events $\xi_k = (j_k, \omega_k, s_k)$ which we interpret in the following way: at time s_k the system has exchanged an entropy quantum ω_k with reservoir $\mathcal{R}_{j_k}$. According to this interpretation, the random variable

$$\varsigma_j(\xi) = \frac{1}{t} \sum_{k: j_k = j} \omega_k,$$

represents the mean rate of entropy exchange of the system with reservoir $\mathcal{R}_j$ during the time interval $[0, t]$. It follows that one can rewrite the expansion (40) as

$$\langle \rho | e^{t \mathcal{L}_{(\alpha)}}(\mathbb{1}) \rangle = \int_{\Xi^t} e^{-t \sum_{j=1}^M \alpha_j \varsigma_j(\xi)} \, d\mu_\rho^t(\xi).$$

This proves Part (2) of Theorem 3.1 and identifies the measure P_ρ^t as the law of the random variable $\varsigma(\xi) = (\varsigma_1(\xi), \ldots, \varsigma_M(\xi))$ induced by the measure μ_ρ^t.

7 Proofs

7.1 Detailed Balance

To a faithful state ρ, we associate two groups of transformations of $\mathcal{O}$, the modular group $\Delta_\rho^z(X) = \rho^z X \rho^{-z}$, and the group $\mathcal{R}_\rho^z(X) = \rho^z X \rho^z$, $z \in \mathbb{C}$. $\Delta_\rho^1 = \Delta_\rho$ is the modular operator of the state ρ. Note that $\Delta_\rho^{i\alpha} \in \mathrm{CP}_1(\mathcal{O})$ and $\mathcal{R}_\rho^\alpha \in \mathrm{CP}(\mathcal{O})$ for $\alpha \in \mathbb{R}$.

Theorem 7.1 *Let ρ be a faithful state on $\mathcal{O}$ and $\mathcal{L} = \mathrm{i}[T, \cdot] - \frac{1}{2}\{\Phi(\mathbb{1}), \cdot\} + \Phi$ a Lindbladian generating a QDS. Suppose that $\mathcal{L}^*(\rho) = 0$ and $\Phi^\rho = \Phi$. Then:*

(1) *The Hermitian and anti-Hermitian parts of $\mathcal{L}$ w.r.t. the inner product induced by ρ are given by*

$$\mathcal{L}^{(d)}(X) = \frac{1}{2}(\mathcal{L} + \mathcal{L}^\rho)(X) = -\frac{1}{2}\{\Phi(\mathbb{1}), X\} + \Phi(X),$$

$$\mathcal{L}^{(h)}(X) = \frac{1}{2}(\mathcal{L} - \mathcal{L}^\rho)(X) = \mathrm{i}[T, X].$$

They are also called dissipative and Hamiltonian parts of $\mathcal{L}$.
(2) *$\mathcal{L}, \mathcal{L}^{(h)}, \mathcal{L}^{(d)}$ and Φ commute with the modular operator Δ_ρ. In particular, T and $\Phi(\mathbb{1})$ commute with ρ and $\mathcal{L}^{(d)*}(\rho) = \mathcal{L}^{(h)*}(\rho) = 0$.*
(3) *The CP map Φ admits a decomposition*

$$\Phi = \sum_{\omega \in \mathrm{sp}(\log \Delta_\rho)} \Phi_\omega,$$

where $\Phi_\omega \in \mathrm{CP}(\mathcal{O})$ satisfies $\Phi_\omega(X\rho^{-\alpha})\rho^\alpha = e^{-\alpha\omega}\Phi_\omega(X)$, $\Phi_\omega^\rho = \Phi_{-\omega}$ and $\Phi_\omega^* = e^\omega \Phi_{-\omega}$.

(4) *For $\alpha \in \mathbb{C}$ define $\mathcal{L}_\alpha = \mathcal{R}_\rho^{\alpha/2} \circ \mathcal{L} \circ \mathcal{R}_\rho^{-\alpha/2}$. Then*

$$\mathcal{L}_\alpha(X) = \mathcal{L}(X\rho^{-\alpha})\rho^\alpha = \mathrm{i}[T, X] - \frac{1}{2}\{\Phi(\mathbb{1}), X\} + \sum_{\omega \in \mathrm{sp}(\log \Delta_\rho)} \mathrm{e}^{-\alpha\omega}\Phi_\omega(X), \qquad (41)$$

holds for all $X \in \mathcal{O}$. Moreover, $\{\mathrm{e}^{t\mathcal{L}_\alpha}\}_{t \geq 0}$ is a CP($\mathcal{O}$) semigroup for $\alpha \in \mathbb{R}$.

(5) *If the pair $(\rho, \mathcal{L})$ is time-reversible with time-reversal Θ, then for all $\alpha \in \mathbb{R}$*

$$\Theta \circ \mathcal{L}_\alpha^* = \mathcal{L}_{1-\alpha} \circ \Theta.$$

Remark The proofs of Parts (1)–(3) can be found in [3, 42]. For the readers convenience we provide a complete proof below.

Proof We start with the simple remarks that $\mathcal{L}^*(X) = -\mathrm{i}[T, X] - \frac{1}{2}\{\Phi(\mathbb{1}), X\} + \Phi^*(X)$ and that $\mathcal{M}^\rho(X) = \mathcal{M}^*(X\rho)\rho^{-1}$ for any linear map $\mathcal{M}$ on $\mathcal{O}$. We recall that $\mathcal{M}$ is a $*$-map if $\mathcal{M}(X^*) = \mathcal{M}(X)^*$ for all $X \in \mathcal{O}$. The maps $\mathcal{L}$ and $\mathcal{L}^*$, as generators of positive semigroups, and Φ as a positive map, are all $*$-maps.

The fact that Φ is ρ-self-adjoint implies $\Phi^*(X\rho) = \Phi(X)\rho$ for all $X \in \mathcal{O}$ and in particular that $\Phi^*(\rho) = \Phi(\mathbb{1})\rho$. Thus, since T, $\Phi(\mathbb{1})$ and ρ are self-adjoint, it follows from

$$0 = \mathcal{L}^*(\rho) = -\mathrm{i}[T, \rho] + \Phi^*(\rho) - \frac{1}{2}\{\Phi(\mathbb{1}), \rho\} = \frac{1}{2}[\Phi(\mathbb{1}) - 2\mathrm{i}T, \rho],$$

that ρ commutes with T and $\Phi(\mathbb{1})$. A simple calculation yields

$$\mathcal{L}^\rho(X) = -\mathrm{i}[T, X] + \Phi(X) - \frac{1}{2}\{\Phi(\mathbb{1}), X\}, \qquad (42)$$

and Part (1) follows.

The formula (42) implies that $\mathcal{L}^\rho$ is a $*$-map. Thus, one can write

$$\mathcal{L}^\rho(\Delta_\rho(X)) = \mathcal{L}^*(\rho X)\rho^{-1} = \mathcal{L}^*((X^*\rho)^*)\rho^{-1} = \mathcal{L}^*(X^*\rho)^*\rho^{-1}$$

$$= (\mathcal{L}^\rho(X^*)\rho)^*\rho^{-1} = \Delta_\rho(\mathcal{L}^\rho(X)). \qquad (43)$$

It follows that $[\mathcal{L}^\rho, \Delta_\rho] = 0$ and, since Δ_ρ is ρ-self-adjoint, that $[\mathcal{L}, \Delta_\rho] = 0$. Clearly, $[T, \rho] = 0$ implies that $[\mathcal{L}^{(h)}, \Delta_\rho] = 0$ and $\mathcal{L}^{(h)*}(\rho) = 0$. Thus, one also has $[\mathcal{L}^{(d)}, \Delta_\rho] = 0$ and

$$\mathcal{L}^{(d)*}(\rho) = \mathcal{L}^*(\rho) - \mathcal{L}^{(h)*}(\rho) = 0.$$

Finally, $[\Phi(\mathbb{1}), \rho] = 0$ implies $[\Phi, \Delta_\rho] = 0$, which concludes the proof of Part (2).

Denote by $\log \rho = \sum_\lambda \lambda P_\lambda$ the spectral representation of $\log \rho$. The operator $\log \Delta_\rho = [\log \rho, \cdot]$ is self-adjoint on $\mathcal{O}$, with spectrum $\mathrm{sp}(\log \Delta_\rho) = \mathrm{sp}(\log \rho) - \mathrm{sp}(\log \rho)$. Its spectral representation is given by

$$\log \Delta_\rho = \sum_{\omega \in \mathrm{sp}(\log \Delta_\rho)} \omega \mathcal{P}_\omega, \qquad \mathcal{P}_\omega(X) = \sum_{\lambda - \mu = \omega} P_\lambda X P_\mu.$$

Since Φ commutes with Δ_ρ, it commutes with each of the spectral projection $\mathcal{P}_\omega$, and in particular one has $\mathcal{P}_\omega \circ \Phi \circ \mathcal{P}_\omega = \mathcal{P}_\omega \circ \Phi$. Thus, setting

$$\Phi_\omega(X) = \sum_{\substack{\lambda-\mu=\omega \\ \lambda'-\mu'=\omega}} P_\mu \Phi(P_\lambda X P_{\lambda'}) P_{\mu'},$$

we have

$$\sum_{\omega \in \mathrm{sp}(\log \Delta_\rho)} \Phi_\omega(X) = \sum_{\lambda-\mu=\lambda'-\mu'} P_\mu \Phi(P_\lambda X P_{\lambda'}) P_{\mu'} = \sum_{\lambda-\lambda'=\mu-\mu'} P_\mu \Phi(P_\lambda X P_{\lambda'}) P_{\mu'}$$

$$= \sum_{\omega \in \mathrm{sp}(\log \Delta_\rho)} \mathcal{P}_\omega\big(\Phi\big(\mathcal{P}_\omega(X)\big)\big) = \sum_{\omega \in \mathrm{sp}(\log \Delta_\rho)} \mathcal{P}_\omega\big(\Phi(X)\big) = \Phi(X).$$

Moreover, since Φ is completely positive, it follows from the identity

$$\sum_{i,j} B_i^* \Phi_\omega\big(A_i^* A_j\big) B_j = \sum_{(i,\lambda),(j,\mu)} B_{i,\lambda}^* \Phi\big(A_{i,\lambda}^* A_{j,\mu}\big) B_{j,\mu},$$

where $A_{i,\lambda} = A_i P_\lambda$ and $B_{i,\lambda} = P_{\lambda-\omega} B_i$, that Φ_ω is completely positive. Next, note that the identity

$$P_\mu \Phi\big(P_\lambda X \rho^{-\alpha} P_{\lambda'}\big) P_{\mu'} \rho^\alpha = P_\mu \Phi(P_\lambda X P_{\lambda'}) P_{\mu'} \mathrm{e}^{-\alpha(\lambda'-\mu')},$$

implies

$$\Phi_\omega\big(X\rho^{-\alpha}\big)\rho^\alpha = \mathrm{e}^{-\alpha\omega}\Phi_\omega(X). \tag{44}$$

The identity $\Phi^*(X) = \Phi(X\rho^{-1})\rho$ and a simple calculation yield

$$\Phi_\omega^*(X) = \sum_{\substack{\lambda-\mu=\omega \\ \lambda'-\mu'=\omega}} P_\lambda \Phi^*(P_\mu X P_{\mu'}) P_{\lambda'} = \sum_{\substack{\lambda-\mu=\omega \\ \lambda'-\mu'=\omega}} P_\lambda \Phi\big(P_\mu X P_{\mu'} \rho^{-1}\big) \rho P_{\lambda'}$$

$$= \sum_{\substack{\lambda-\mu=\omega \\ \lambda'-\mu'=\omega}} P_\lambda \Phi(P_\mu X P_{\mu'}) P_{\lambda'} \mathrm{e}^{\lambda'-\mu'} = \mathrm{e}^\omega \Phi_{-\omega}(X).$$

The last identity combined with Eq. (44) gives

$$\Phi_\omega^\rho(X) = \Phi_\omega^*(X\rho)\rho^{-1} = \mathrm{e}^\omega \Phi_{-\omega}(X\rho)\rho^{-1} = \Phi_{-\omega}(X),$$

and Part (3) follows.

To prove Part (4), note that since $\mathcal{L}$ commutes with Δ_ρ, one has

$$\mathcal{L}_\alpha(X) = \mathcal{R}_\rho^{\alpha/2} \circ \mathcal{L} \circ \mathcal{R}_\rho^{-\alpha/2}(X) = \mathcal{R}_\rho^{\alpha/2} \circ \Delta_\rho^{-\alpha/2} \circ \mathcal{L} \circ \Delta_\rho^{\alpha/2} \circ \mathcal{R}_\rho^{-\alpha/2}(X)$$

$$= \rho^{\alpha/2} \rho^{-\alpha/2} \mathcal{L}\big(\rho^{\alpha/2} \rho^{-\alpha/2} X \rho^{-\alpha/2} \rho^{-\alpha/2}\big) \rho^{\alpha/2} \rho^{\alpha/2} = \mathcal{L}\big(X\rho^{-\alpha}\big)\rho^\alpha.$$

The formula (41) follows from the relation $\mathcal{L}_\alpha(X) = \mathcal{L}(X\rho^{-\alpha})\rho^\alpha$, the fact that ρ commutes with T and $\Phi(\mathbb{1})$ and Eq. (44). Since $\mathrm{e}^{t\mathcal{L}_\alpha} = \mathcal{R}_\rho^{\alpha/2} \circ \mathrm{e}^{t\mathcal{L}} \circ \mathcal{R}_\rho^{-\alpha/2}$, $\{\mathrm{e}^{t\mathcal{L}_\alpha}\}_{t\geq 0}$ is a CP($\mathcal{O}$) semigroup for all $\alpha \in \mathbb{R}$, and Part (4) follows.

It remains to prove Part (5). Define $\mathcal{L}_\alpha^{(d)}(X) = \mathcal{L}^{(d)}(X\rho^{-\alpha})\rho^\alpha$. A simple calculation gives

$$\mathcal{L}_\alpha^{(d)*}(X) = \mathcal{L}^{(d)*}(X\rho^\alpha)\rho^{-\alpha},$$

and Part (1) implies

$$\mathcal{L}^{(d)*}(X) = \mathcal{L}^{(d)\rho}(X\rho^{-1})\rho = \mathcal{L}^{(d)}(X\rho^{-1})\rho.$$

Hence,

$$\mathcal{L}_\alpha^{(d)*} = \mathcal{L}_{1-\alpha}^{(d)}.$$

Since Θ is involutive, the relation $\mathcal{L}^\rho \circ \Theta = \Theta \circ \mathcal{L}$ implies $\mathcal{L} \circ \Theta = \Theta \circ \mathcal{L}^\rho$. It follows from Part (1) that $\mathcal{L}^{(h)} \circ \Theta = -\Theta \circ \mathcal{L}^{(h)}$ and $\mathcal{L}^{(d)} \circ \Theta = \Theta \circ \mathcal{L}^{(d)}$. Moreover, $\Theta(\rho^\alpha) = \rho^\alpha$ implies $\mathcal{L}_\alpha^{(d)} \circ \Theta = \Theta \circ \mathcal{L}_\alpha^{(d)}$. Thus, one has

$$\mathcal{L}_\alpha^* = \mathcal{L}^{(h)*} + \mathcal{L}_\alpha^{(d)*} = -\mathcal{L}^{(h)} + \mathcal{L}_{1-\alpha}^{(d)},$$

and

$$\Theta \circ \mathcal{L}_\alpha^* = \left(\mathcal{L}^{(h)} + \mathcal{L}_{1-\alpha}^{(d)}\right) \circ \Theta = \mathcal{L}_{1-\alpha} \circ \Theta.$$

$\square$

We finish this section with:

Theorem 7.2 *Let ρ be a faithful state and $\mathcal{L}$ a Lindbladian on $\mathcal{O}$ generating a QDS. Suppose that $\mathcal{L}^*(\rho) = 0$. Then the following statements are equivalent*:

(1) *There exist a self-adjoint $T \in \mathcal{O}$ such that the Hermitian part of $\mathcal{L}$ w.r.t. the inner product induced by ρ has the form*

$$\mathcal{L}^{(h)}(X) = \frac{1}{2}\left(\mathcal{L} - \mathcal{L}^\rho\right)(X) = \mathrm{i}[T, X].$$

(2) *There exists a Lindblad decomposition $\mathcal{L} = \mathrm{i}[T, \cdot] - \frac{1}{2}\{\Phi(\mathbb{1}), \cdot\} + \Phi$ such that $\Phi^\rho = \Phi$.*

Remark This theorem establishes that Definition 2.4 (a) is equivalent to the definition of detailed balance given in [42] (see also Sect. IV in [48]). Although we shall not make use of this result in the sequel, we include the proof for reader's convenience.

Proof The implication (2) $\Rightarrow$ (1) follows from Part (1) of Theorem 7.1. To prove the implication (1) $\Rightarrow$ (2), note first that (1) implies that

$$\mathcal{L}^{(d)} = \frac{1}{2}\left(\mathcal{L} + \mathcal{L}^\rho\right),$$

is a Lindbladian generating a QDS. Since $\mathcal{L}^{(d)}$ is ρ-self-adjoint, arguing as in (43) one deduces that $[\Delta_\rho, \mathcal{L}^{(d)}] = 0$. Let now $\mathcal{L} = \mathrm{i}[S, \cdot] - \frac{1}{2}\{\Psi(\mathbb{1}), \cdot\} + \Psi$ be a Lindblad decomposition. Since

$$\mathcal{L}^{(d)} = \lim_{T\to\infty} \frac{1}{T} \int_0^T \Delta_\rho^{\mathrm{i}t} \circ \mathcal{L}^{(d)} \circ \Delta_\rho^{-\mathrm{i}t} dt,$$

setting

$$M = \lim_{T \to \infty} \frac{1}{T} \int_0^T \rho^{\mathrm{i}t} S \rho^{-\mathrm{i}t} \mathrm{d}t, \qquad \varXi = \lim_{T \to \infty} \frac{1}{T} \int_0^T \Delta_\rho^{\mathrm{i}t} \circ \Psi \circ \Delta_\rho^{-\mathrm{i}t} \mathrm{d}t,$$

we deduce that

$$\mathcal{L}^{(d)} = \mathrm{i}[M, \cdot] - \frac{1}{2}\big\{\varXi(\mathbb{1}), \cdot\big\} + \varXi,$$

is also a Lindblad decomposition. Clearly, $[\Delta_\rho, \varXi] = 0$, $[M, \rho] = 0$, $[\varXi(\mathbb{1}), \rho] = 0$. Hence,

$$\mathcal{L}^{(d)\rho} = -\mathrm{i}[M, \cdot] - \frac{1}{2}\big\{\varXi(\mathbb{1}), \cdot\big\} + \varXi^\rho,$$

$$\varXi^\rho(X) = \rho^{-1/2} \varXi^*\big(\rho^{1/2} X \rho^{1/2}\big)\rho^{-1/2},$$

and we derive that $\varXi^\rho(\mathbb{1}) = \varXi(\mathbb{1}) + \mathcal{L}^{(d)\rho}(\mathbb{1}) = \varXi(\mathbb{1}) + \mathcal{L}^{(d)}(\mathbb{1}) = \varXi(\mathbb{1})$. Setting

$$\varPhi = \frac{1}{2}\big(\varXi + \varXi^\rho\big),$$

we get

$$\mathcal{L}^{(d)} = \frac{1}{2}\big(\mathcal{L}^{(d)} + \mathcal{L}^{(d)\rho}\big) = -\frac{1}{2}\big\{\varPhi(\mathbb{1}), \cdot\big\} + \varPhi,$$

where $\varPhi$ is CP and $\varPhi^\rho = \varPhi$. Hence, $\mathcal{L} = \mathrm{i}[T, \cdot] - \frac{1}{2}\{\varPhi(\mathbb{1}), \cdot\} + \varPhi$ is a Lindblad decomposition of $\mathcal{L}$ with $\varPhi^\rho = \varPhi$. $\qquad\square$

7.2 Irreducibility and Positivity Improving

We start with the following observation of [57]:

Proposition 7.3 *A positive linear map* $\varPhi : \mathcal{O} \to \mathcal{O}$ *is irreducible iff* $\mathrm{e}^{t\varPhi}$ *is positivity improving for some* (*and hence all*) $t > 0$.

Proof If $\varPhi$ is irreducible, then it follows from Lemma 2.1 in [28] that

$$(\mathrm{Id} + \varPhi)^{\dim \mathcal{H}-1},$$

is positivity improving, and so $\mathrm{e}^{t\varPhi}$ is positivity improving for all $t > 0$. To prove the converse, suppose that $\mathrm{e}^{t\varPhi}$ is positivity improving and that $\varPhi(P) \leq \lambda P$, where $\lambda > 0$ and $P \neq 0$ is a projection. Then $\varPhi^n(P) \leq \lambda^n P$ for all n, and so $0 < \mathrm{e}^{t\varPhi}(P) \leq \mathrm{e}^{\lambda t} P$. The last relation implies that $P = \mathbb{1}$. $\qquad\square$

Proof of Theorem 2.1 We follow [57]. Let $\varphi, \psi \in \mathcal{H}$ be non-zero vectors and $t > 0$. Expanding $\mathrm{e}^{t\varPhi^*}$ into a power series, we get

$$\big\langle \varphi \big| \mathrm{e}^{t\varPhi^*}\big(|\psi\rangle\langle\psi|\big)\varphi \big\rangle = \big|\langle\varphi|\psi\rangle\big|^2 + \sum_{n=1}^{\infty} \frac{t^n}{n!} \sum_{j_1,\dots,j_n} \big|\langle\varphi|V_{j_1} \cdots V_{j_n}\psi\rangle\big|^2.$$

Hence, $\langle\varphi|\mathrm{e}^{t\varPhi^*}(|\psi\rangle\langle\psi|)\varphi\rangle = 0$ iff $\varphi \perp \mathcal{A}\psi$, and we deduce that $\mathrm{e}^{t\varPhi^*}$ is positivity improving iff $\mathcal{A}\psi = \mathcal{H}$ for all non-zero vectors $\psi \in \mathcal{H}$. Since $\mathrm{e}^{t\varPhi^*}$ is positivity improving iff $\mathrm{e}^{t\varPhi}$ is, the result follows from Proposition 7.3. $\qquad\square$

Proof of Theorem 2.2 The proof of based on Perron-Frobenius theory of positive maps developed in [28]. Let $t > 0$ be given. The map $e^{t\mathcal{L}}$ is positive and its spectral radius is $e^{t\ell}$. It follows from Theorem 2.5 in [28] that $e^{t\ell}$ is an eigenvalue of $e^{t\mathcal{L}}$, and that there exists a non-zero $M \in \mathcal{O}_+$ such that

$$e^{t\mathcal{L}}(M) = e^{t\ell}M.$$

Since the map $e^{t\mathcal{L}}$ is positivity improving, $M > 0$. Define

$$\Psi(X) = M^{-1/2}e^{t(\mathcal{L}-\ell)}\big(M^{1/2}XM^{1/2}\big)M^{-1/2}.$$

The map Ψ is unital, completely positive (hence Schwartz), and positivity improving (hence irreducible). The same holds for Ψ^n, $n \geq 1$, and it follows from Theorem 4.2 in [28] that 1 is a simple eigenvalue of Ψ and that Ψ has no other eigenvalues on the unit circle $|z| = 1$. Hence, $\mathcal{L}$ has a simple eigenvalue at ℓ and no other eigenvalues on the line $\mathrm{Re}\,z = \ell$.

Denote by μ the eigenvector of $\mathcal{L}^*$ associated to the eigenvalue ℓ. Since $e^{t\mathcal{L}^*}$ is positivity improving by duality, one can chose $\mu > 0$ and normalize it by $\langle\mu|M\rangle = 1$. Let $\delta > 0$ be the distance from $\mathrm{sp}(\mathcal{L}) \setminus \{\ell\}$ to the line $\mathrm{Re}\,z = \ell$. Then, for any $\epsilon > 0$,

$$\big\langle\rho\big|e^{t\mathcal{L}}(X)\big\rangle = e^{t\ell}\big(\langle\rho|M\rangle\langle\mu|X\rangle + O\big(e^{-t(\delta-\epsilon)}\big)\big), \tag{45}$$

holds for all states ρ and all $X \in \mathcal{O}$. Since $\langle\rho|M\rangle > 0$ and $\langle\mu|X\rangle > 0$ for non-zero $X \in \mathcal{O}_+$, Eq. (3) follows.

If $\mathcal{L}(\mathbb{1}) = 0$, then $e^{t\mathcal{L}}(\mathbb{1}) = \mathbb{1}$ and since $\|e^{t\mathcal{L}}\| = \|e^{t\mathcal{L}}(\mathbb{1})\| = 1$, it follows that $\ell = 0$ and $M = \mathbb{1}$. By duality, (45) yields

$$e^{t\mathcal{L}^*}(\rho) = \mu + O\big(e^{-t(\delta-\epsilon)}\big),$$

and the semigroup $\{e^{t\mathcal{L}}\}_{t \geq 0}$ is relaxing exponentially fast to the faithful state $\rho_+ = \mu$. $\quad\square$

Proof of Theorem 2.3 Note that $\mathcal{K}_0 : X \mapsto K^*X + XK$ generates a continuous group of completely positive maps on $\mathcal{O}$, namely $e^{t\mathcal{K}_0}(X) = e^{tK^*}Xe^{tK}$. Denoting $\Gamma^t = e^{-t\mathcal{K}_0} \circ e^{t\mathcal{K}}$, it is sufficient to show that $\langle\varphi|\Gamma^t(|\psi\rangle\langle\psi|)\varphi\rangle > 0$ for any non-zero vectors $\varphi, \psi \in \mathcal{H}$ and all $t > 0$. To prove this claim, let us assume that $\langle\varphi|\Gamma^{t_0}(|\psi\rangle\langle\psi|)\varphi\rangle = 0$ for some $t_0 > 0$. The Dyson expansion for Γ^{t_0} gives

$$\Gamma^{t_0} = \mathrm{Id} + \sum_{n=1}^{\infty}\int_{\Delta_n}\Phi_s^n\,\mathrm{d}s,$$

where

$$\Delta_n = \big\{s = (s_1, \ldots, s_n) \in \mathbb{R}^n \,\big|\, 0 \leq s_1 \leq \cdots \leq s_n \leq t_0\big\},$$

$\Phi_s = e^{-s\mathcal{K}_0} \circ \Phi \circ e^{s\mathcal{K}_0}$ for $s \in \mathbb{R}$, and $\Phi_s^n = \Phi_{s_1} \circ \Phi_{s_2} \circ \cdots \circ \Phi_{s_n}$ for $s \in \Delta_n$. It follows that

$$\big\langle\varphi\big|\Gamma^{t_0}\big(|\psi\rangle\langle\psi|\big)\varphi\big\rangle = \big|\langle\varphi|\psi\rangle\big|^2 + \sum_{n=1}^{\infty}\int_{\Delta_n}\big\langle\varphi\big|\Phi_s^n\big(|\psi\rangle\langle\psi|\big)\varphi\big\rangle\,\mathrm{d}s = 0.$$

Since the functions $s \mapsto \langle\varphi|\Phi_s^n(|\psi\rangle\langle\psi|)\varphi\rangle$ are continuous and non-negative, we infer that $\langle\varphi|\Phi_s^n(|\psi\rangle\langle\psi|)\varphi\rangle = 0$ for all n and all $s \in \Delta_n$, and in particular that $\langle\varphi|\Phi^n(|\psi\rangle\langle\psi|)\varphi\rangle = 0$ for all n. Hence, $\langle\varphi|e^{t\Phi}(|\psi\rangle\langle\psi|)\varphi\rangle = 0$ for all $t \geq 0$, and Proposition 7.3 implies that $\varphi = 0$ or $\psi = 0$. $\quad\square$

For later reference, we mention the following simple fact:

Proposition 7.4 *Let Φ_j, $j = 1, \ldots, n$, be positive linear maps such that $\sum_j \Phi_j$ is irreducible. If $\lambda_1, \ldots, \lambda_n$ are strictly positive then $\sum_j \lambda_j \Phi_j$ is irreducible.*

Proof The result follows from the obvious inequality

$$\min_i \lambda_i \sum_j \Phi_j \leq \sum_j \lambda_j \Phi_j \leq \max_i \lambda_i \sum_j \Phi_j.$$

$\square$

7.3 Proof of Theorem 3.1

(1)–(2) were already proven in Sect. 6.

(3)–(4) By Eq. (39), Proposition 7.4, and Theorem 2.3, the CP semigroup $\{e^{t\mathcal{L}_{(\alpha)}}\}_{t \geq 0}$ is positivity improving for all $\alpha \in \mathbb{R}^M$, and the statement follows from Theorem 2.2.

(5) Note that the map $\mathbb{C}^M \ni \alpha \mapsto \mathcal{L}_{(\alpha)}$ is entire analytic. Since $e(\alpha)$ is a simple eigenvalue of $\mathcal{L}_{(\alpha)}$ for all $\alpha \in \mathbb{R}^M$, the regular perturbation theory implies that $e(\alpha)$ is a real analytic function of α. Property (2) and Hölder's inequality yield that $e(\alpha)$ is a convex function of α.

(6) This part also follows from regular perturbation theory. Fix $\alpha_0 \in \mathbb{R}^M$ and set

$$\delta = \frac{1}{2} \min\{e(\alpha_0) - \operatorname{Re} z \,|\, z \in \operatorname{sp}(\mathcal{L}_{(\alpha_0)}) \setminus \{e(\alpha_0)\}\} > 0.$$

If ϵ is small enough and $\alpha \in D_\epsilon = \{z \in \mathbb{C}^M \,|\, |\alpha - \alpha_0| < \epsilon\}$, one has

$$\langle \rho | e^{t\mathcal{L}_{(\alpha)}}(\mathbb{1}) \rangle = e^{te(\alpha)}\big(\langle \rho | M_\alpha \rangle \langle \mu_\alpha | \mathbb{1} \rangle + O\big(e^{t(-\delta + O(\epsilon))}\big)\big),$$

where $e(\alpha)$, M_α and μ_α are analytic functions of α such that $\langle \rho | M_\alpha \rangle \langle \mu_\alpha | \mathbb{1} \rangle - \langle \rho | M_{\alpha_0} \rangle \langle \mu_{\alpha_0} | \mathbb{1} \rangle = O(\epsilon)$ and $\langle \rho | M_{\alpha_0} \rangle \langle \mu_{\alpha_0} | \mathbb{1} \rangle > 0$. It follows that there exists $\epsilon > 0$ such that for $\alpha \in D_\epsilon$,

$$\lim_{t \to \infty} \frac{1}{t} \log \langle \rho | e^{t\mathcal{L}_{(\alpha)}}(\mathbb{1}) \rangle = e(\alpha).$$

(7) Let Θ be the time-reversal map. By Property (5) of Theorem 7.1 one has

$$\Theta \circ \mathcal{L}^*_{(\alpha)} = \mathcal{L}_{(1-\alpha)} \circ \Theta,$$

for all $\alpha \in \mathbb{R}^M$. It follows that $\operatorname{sp}(\mathcal{L}_{(\alpha)}) = \operatorname{sp}(\mathcal{L}_{(1-\alpha)})$ and hence $e(\alpha) = e(1 - \alpha)$.

(8) If Hypothesis (KMS$_\beta$) is satisfied, then $\rho_j = Z_j^{-1} \nu^{\beta_j}$ with $\nu = e^{-H_S}$ and $Z_j = \operatorname{tr}(\nu^{\beta_j})$. Hence, $\mathcal{R}_{\rho_j}^{\alpha_j} = Z_j^{-2\alpha_j} \mathcal{R}_\nu^{\alpha_j \beta_j}$ and Part (4) of Theorem 7.1 yields

$$\mathcal{L}_{(\alpha)} = \sum_{j=1}^M \mathcal{R}_\nu^{\alpha_j \beta_j / 2} \circ \mathcal{L}_j \circ \mathcal{R}_\nu^{-\alpha_j \beta_j / 2}.$$

It follows that

$$\mathcal{L}_{(\alpha + \lambda \beta^{-1})} = \mathcal{R}_\nu^{\lambda/2} \circ \mathcal{L}_{(\alpha)} \circ \mathcal{R}_\nu^{-\lambda/2},$$

and so $\operatorname{sp}(\mathcal{L}_{(\alpha + \lambda \beta^{-1})}) = \operatorname{sp}(\mathcal{L}_{(\alpha)})$. In particular, $e(\alpha + \lambda \beta^{-1}) = e(\alpha)$.

7.4 Proof of Theorem 3.4

(1) At $\alpha = 0$, the spectral projection of $\mathcal{L}_{(\alpha)} = \mathcal{L}$ associated to its dominant eigenvalue 1 is $|\mathbb{1}\rangle\langle\rho_+|$. Thus, for α sufficiently close to $0 \in \mathbb{R}^M$, $e(\alpha) = E_1(\alpha)/E_0(\alpha)$ where

$$E_n(\alpha) = \oint_\Gamma z^n \langle \rho_+ | (z - \mathcal{L}_{(\alpha)})^{-1}(\mathbb{1})\rangle \frac{\mathrm{d}z}{2\pi\mathrm{i}},$$

and Γ is a small circle centred at 1 such that no other point of $\mathrm{sp}(\mathcal{L})$ is on or inside Γ. Since $(z - \mathcal{L})^{-1}(\mathbb{1}) = z^{-1}$, one has $E_1(0) = 0$ and $E_0(0) = 1$ and hence $(\partial_{\alpha_j} e)(0) = (\partial_{\alpha_j} E_1)(0)$. An elementary calculation yields

$$(\partial_{\alpha_j} E_n)(0) = \oint_\Gamma z^n \langle \rho_+ | (z - \mathcal{L})^{-1} \circ \mathcal{L}_{;\alpha_j} \circ (z - \mathcal{L})^{-1}(\mathbb{1})\rangle \frac{\mathrm{d}z}{2\pi\mathrm{i}},$$

where

$$\mathcal{L}_{;\alpha_j}(X) = \partial_{\alpha_j} \mathcal{L}_{(\alpha)}(X)|_{\alpha=0} = \mathcal{L}_j(X S_j) - \mathcal{L}_j(X) S_j. \tag{46}$$

The identities

$$\mathcal{L}_{;\alpha_j} \circ (z - \mathcal{L})^{-1}(\mathbb{1}) = z^{-1} \mathcal{L}_{;\alpha_j}(\mathbb{1}) = z^{-1} \mathcal{I}_j,$$

$$\langle \rho_+ | (z - \mathcal{L})^{-1} = z^{-1} \langle \rho_+ |,$$

yield

$$(\partial_{\alpha_j} E_0)(0) = 0, \qquad (\partial_{\alpha_j} E_1)(0) = \rho_+(\mathcal{I}_j),$$

and the statement follows.

(2) From the previous calculation, we easily infer

$$(\partial_{\alpha_k} \partial_{\alpha_j} e)(0) = (\partial_{\alpha_k} \partial_{\alpha_j} E_1)(0)$$

$$= \oint_\Gamma \frac{1}{z} \langle \rho_+ | \mathcal{L}_{;\alpha_k} \circ (z - \mathcal{L})^{-1}(\mathcal{I}_j) + \mathcal{L}_{;\alpha_j} \circ (z - \mathcal{L})^{-1}(\mathcal{I}_k) + \mathcal{L}_{;\alpha_k \alpha_j}(\mathbb{1})\rangle \frac{\mathrm{d}z}{2\pi\mathrm{i}}, \tag{47}$$

where

$$\mathcal{L}_{;\alpha_k \alpha_j}(\mathbb{1}) = \partial_{\alpha_k} \partial_{\alpha_j} \mathcal{L}_{(\alpha)}(\mathbb{1})|_{\alpha=0} = \delta_{kj} (\mathcal{L}_j(S_j^2) - 2\mathcal{L}_j(S_j) S_j).$$

Theorem 7.1 (2) implies

$$\mathcal{L}_j(\mathrm{e}^{t S_j} X \mathrm{e}^{-t S_j}) = \mathrm{e}^{t S_j} \mathcal{L}_j(X) \mathrm{e}^{-t S_j},$$

and hence $\mathcal{L}_j([S_j, X]) = [S_j, \mathcal{L}_j(X)]$. It follows that $[S_j, \mathcal{L}_j(S_j)] = 0$ and $\mathcal{L}_{;\alpha_k \alpha_j}(\mathbb{1}) = \delta_{kj} D_j(S_j, S_j)$. Using the fact that

$$\oint_\Gamma \frac{1}{z} \langle \rho_+ | \mathcal{L}_{;\alpha_k} \circ (z - \mathcal{L})^{-1}(\rho_+(\mathcal{I}_j))\rangle \frac{\mathrm{d}z}{2\pi\mathrm{i}} = \oint_\Gamma \frac{1}{z^2} \rho_+(\mathcal{I}_k) \rho_+(\mathcal{I}_j) \frac{\mathrm{d}z}{2\pi\mathrm{i}} = 0,$$

we can replace $\mathcal{I}_{j/k}$ with $\mathcal{J}_{j/k} = \mathcal{I}_{j/k} - \rho_+(\mathcal{I}_{j/k})$ in Eq. (47). Since $\rho_+(\mathcal{J}_j) = 0$, the meromorphic function $(z - \mathcal{L})^{-1}(\mathcal{J}_j)$ is regular at $z = 0$ and one has

$$\lim_{z \to 0}(z - \mathcal{L})^{-1}(\mathcal{J}_j) = \int_0^\infty \mathrm{e}^{t\mathcal{L}}(\mathcal{J}_j)\,\mathrm{d}t,$$

the integral on the r.h.s. being absolutely convergent. We therefore have

$$(\partial_{\alpha_k}\partial_{\alpha_j}e)(0) = \int_0^\infty \rho_+\big(\mathcal{L}_{;\alpha_k}\big(\mathrm{e}^{t\mathcal{L}}(\mathcal{J}_j)\big) + \mathcal{L}_{;\alpha_j}\big(\mathrm{e}^{t\mathcal{L}}(\mathcal{J}_k)\big)\big)\,\mathrm{d}t + \delta_{kj}\rho_+\big(D_j(S_j, S_j)\big).$$

The relation

$$\frac{\partial^2 e(\alpha)}{\partial \alpha_j \partial \alpha_k}\bigg|_{\alpha=0} = -\int_0^\infty \rho_+\big(\mathrm{e}^{t\mathcal{L}}(\mathcal{J}_j)\mathcal{J}_k^+ + \mathrm{e}^{t\mathcal{L}}(\mathcal{J}_k)\mathcal{J}_j^+\big)\mathrm{d}t$$

$$+ \int_0^\infty \rho_+\big(\mathcal{L}_k\big(\mathrm{e}^{t\mathcal{L}}(\mathcal{J}_j)S_k\big) + \mathcal{L}_j\big(\mathrm{e}^{t\mathcal{L}}(\mathcal{J}_k)S_j\big)\big)\mathrm{d}t + \delta_{jk}\rho_+\big(D_j(S_j, S_j)\big),$$

now follows from Eq. (46) and the identity

$$\rho_+\big(\mathcal{L}_k\big(\mathrm{e}^{t\mathcal{L}}(\mathcal{J}_j)\big)S_k\big) = \big\langle \mathcal{L}_k\big(\mathrm{e}^{t\mathcal{L}}(\mathcal{J}_j)\big)\big|S_k\big\rangle_{\rho_+} = \big\langle \mathrm{e}^{t\mathcal{L}}(\mathcal{J}_j)\big|\mathcal{L}_k^{\rho_+}(S_k)\big\rangle_{\rho_+} = \rho_+\big(\mathrm{e}^{t\mathcal{L}}(\mathcal{J}_j)\mathcal{J}_k^+\big).$$

Finally, an application of Vitali's convergence theorem (see Appendix B in [36]) gives

$$\frac{\partial^2 e(\alpha)}{\partial \alpha_j \partial \alpha_k}\bigg|_{\alpha=0} = \lim_{t \to \infty} t\big\langle\big(\varsigma_j - \langle\varsigma_j\rangle_{\rho,t}\big)\big(\varsigma_k - \langle\varsigma_k\rangle_{\rho,t}\big)\big\rangle_{\rho,t}.$$

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