Fluctuation Relations for Quantum Markovian Dynamical Systems

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We derive a general set of fluctuation relations for a nonequilibrium open quantum system described by a Lindblad master equation. In the special case of conservative Hamiltonian dynamics, these identities allow us to retrieve quantum versions of Jarzynski and Crooks relations. In the linear response regime, these fluctuation relations yield a fluctuation-dissipation theorem (FDT) valid for a stationary state arbitrarily far from equilibrium. For a closed system, this FDT reduces to the celebrated Callen-Welton-Kubo formula.

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Fluctuations in nonequilibrium systems have been shown to satisfy various remarquable relations [1, 2] discovered during the last fifteen years. These results have lead to fierce discussions concerning the nature of heat, work and entropy, raising the fundamental issue of understanding the interactions between a given system and its environment (e.g., a thermal bath). In the classical realm, these problems have been progressively clarified while they are still under investigation in the quantum world. Many works on quantum fluctuation relations, including the pioneering ones, consider only closed systems prepared in a Gibbs state and isolated from their environment during their evolution which is thus unitary [3–7]. The general case of an open system continuously interacting with its surroundings can be formally reduced to the previous situation by considering the system together with its environment to be a closed global system: this is the approach adopted in [3–7] (see [7] for a review). However, one must project out the degrees of freedom of the bath to derive an effective dynamics for the system of interest. It is known in classical mechanics, that after such an elimination procedure the Hamiltonian dynamics for the global system becomes effectively a stochastic dynamics for the initial system. Similarly, in the quantum case, after integrating out the degrees of freedom of the environment and using a Markovian approximation, a quantum master equation for the density matrix of the system is obtained. Under some further assumptions [11–12], this master equation can be brought into a form known as the Lindblad equation. The Lindbladian evolution is a non-unitary dynamics for the density matrix \( \rho_t \) of the open system, described by a differential equation with generator \( L_t \). This effective Markovian description is widely used in Quantum Optics [12].

In the present work, we study the time-reversal properties of an open quantum system modeled by a Lindblad equation. A similar point of view was adopted in previous works; in [13–15], the time evolution was discretized in an ad-hoc manner; in [16–18] the quantum master equation was treated as an effective classical master equation and the concept of trajectory and the fluctuation relations for classical system were used. Our approach, here, is to work directly with the continuous time Lindblad equation without referring to any classical effective system, to define an associated time-reversed dynamics and to derive fluctuation relations with quantum observables.

The key results of this work are given in Eqs. (9,12,13,15) and represent an original contribution to quantum non-equilibrium statistical mechanics. Thanks to a suitable deformation of the master equation, we prove a generic relation amongst correlation functions, a kind of book-keeping formula which yields the quantum analog of Jarzynski and Crooks relations. Furthermore, by a lowest order expansion, we derive a generalized fluctuation-dissipation theorem valid in the vicinity of a quantum non-equilibrium steady state. For the special case of a closed system, our approach retrieves previously known work identities [3–6, 13–15] as well as the quantum equilibrium fluctuation-dissipation theorem [12–20].

The density matrix \( \rho_t \) of the open quantum system prepared initially with \( \rho_0 \), evolves according to the master equation

\[
\frac{\partial_t \rho_t}{\rho_t} = L_t^\dagger \rho_t \tag{1}
\]

where \( L_t \) is the Lindbladian superoperator (i.e. a linear map in the space of operators) and \( L_t^\dagger \) its adjoint with respect to the operator scalar product \( (X,Y) = Tr(X^\dagger Y) \), \( X \) and \( Y \) being arbitrary operators and \( X^\dagger \) the hermitian conjugate of \( X \). Under suitable hypotheses (such as trace preserving and complete positivity [11–12]) the Lindbladian \( L_t \) takes the generic form:

\[
L_t X = i[H_t, X] - \frac{1}{2} \sum_{i=1}^I \left( A_i^{\dagger} A_i X + X A_i^{\dagger} A_i - 2 A_i^{\dagger} X A_i \right) . \tag{2}
\]
The first term $i[H_t, X]$ is the conservative part, $H_t$ being the Hamiltonian of the system that may depend on time. The second term models the interactions with the environment (dissipation and coherence effects): the $A^j_\tau$’s are, in general, non-hermitian operators that may depend explicitly on time. In a closed system, $A^j_\tau \equiv 0$. To the time-dependent Lindbladian $L_t$ we associate the accompanying [21] density-matrix $\pi_t$ such that $L^\dagger_t \pi_t = 0$. Physically, $\pi_t$ represents the stationary density-matrix in a system where time is frozen at its instantaneous value $t$. However, because $\pi_t$ depends on time, it does not satisfy Eq. (1). For a closed system, we have $\pi_t = Z_t^{-1} \exp(-\beta H_t)$.

The formal solution of Eq. (1) can be written as $\rho_t = (P^0_t)^\dagger \rho_0$ where the evolution superoperator is given by

$$P^t_s = \exp\left(\int_s^t L_u \, du\right) \equiv \sum_n \int_{s \leq s_1 \leq s_2 \leq \ldots \leq s_n \leq t} L_{s_1} L_{s_2} \ldots L_{s_n} \prod_{i=1}^n ds_i. \tag{3}$$

For $0 \leq t_1 \leq t_2 \leq \ldots \leq t_N \leq T$, the time-ordered correlation of observables $O_0, O_1, O_2 \ldots O_N$ is defined as [12]

$$(O_1(t_1)O_2(t_2)\ldots O_N(t_N)) = \text{Tr} \left( \pi_0 P^t_{t_1} \pi_1 P^t_{t_2} \pi_2 \ldots P^t_{t_{N-1}} \pi_N O_N \right). \tag{4}$$

Note that $P^t_{t_i+1}$ operates on all the terms to its right and that the initial density matrix is given by $\rho_0 = \pi_0$.

A crucial element in our approach is the time-reversed system, characterized by the following Lindbladian:

$$L^R_t = K\pi_t^{-1} L^1_t \pi_t K \quad \text{with} \quad t^* = T - t. \tag{5}$$

The superoperator $K$ acts on an operator $X$ as $K X = \theta X \theta^{-1}$ [22], $\theta$ being the time inversion anti-unitary operator, with $\theta^2 = 1$, that implements time-reversal on the states $\psi$ of the Hilbert space. The superoperator $K$ is antiunitary, with $K^2 = 1$, $K = K^{-1} = K^\dagger$, and is multiplicative i.e. $K(XY) = K(X)K(Y)$. Note that in Eq. (5), $\pi_t$ and $\pi_t^{-1}$ denote left-multiplication superoperators. It is a non-trivial fact that the r.h.s of Eq. (5) defines a bona fide Lindbladian. This is ensured, for example, by imposing at each time $t$ the quantum microreversibility condition (or detailed balance) [22], which implies $L^R_t = L_t$. Here, we do not assume detailed balance and we only require the weaker condition that $L^R_t$ is a well-defined Lindbladian. Using Eq. (5) and the relation $L_t 1 = 0$, we find $(L^R_t)^\dagger K \pi_t = 0$ and hence $P^t_{t_i} = K \pi_t$, thus relating the accompanying distribution of the time-reversed system with that of the original system.

From Eqs. (3) and (4), the evolution superoperator of the time-reversed system is $P^R_s = \exp\left(\int_s^t L^R_u \, du\right)$ and the time-ordered correlations are:

$$(O_1(t_1)O_2(t_2)\ldots O_N(t_N))^R = \text{Tr} \left( \pi_0 P^R_{t_1} \pi_1 P^R_{t_2} \pi_2 \ldots P^R_{t_{N-1}} \pi_N O_N \right). \tag{6}$$

Given a scalar $\alpha$, we deform the Lindbladian $L_t$ and $L^R_t$ to superoperators $L_t(\alpha)$ and $L^R_t(\alpha)$, that act on an observable $X$ as follows:

$$L_t(\alpha) X = (L_t + \alpha \pi_t^{-1} \partial_t \pi_t) X \quad \text{and} \quad L^R_t(\alpha) X = (L^R_t + \alpha (\pi_t)^{-1} \partial_t \pi_t^R) X. \tag{7}$$

The corresponding evolution superoperators are $P^t_s(\alpha) = \exp\left(\int_s^t L_u(\alpha) \, du\right)$ and $P^R_s(\alpha) = \exp\left(\int_s^t L^R_u(\alpha) \, du\right)$. Furthermore, the following conjugation identity between superoperators is satisfied:

$$\pi_0 P^R_s(\alpha) = \left[\pi_T K P^R_{t_0} (1 - \alpha) K\right]_s. \tag{8}$$

This relation stems from the fact that the operator $U_t = \pi_0 P^0_t(\alpha) \pi^{-1}_t$ satisfies the evolution equation $\partial_t U_t = U_t \left(K L^R_t (1 - \alpha) K\right)^\dagger$. Equation (8) is the key duality relation, similar to the identity that lies at the heart of the proof of the classical Jarzynski identity [2] and of the Gallavotti-Cohen theorem in Langevin and Markovian systems [22]. Besides, all these derivations rely on a suitably modified dynamics with respect to a continuous parameter.

Applying the fundamental identity (5) to two arbitrary observables $A$ and $B$ leads to (using the fact that $K$ is multiplicative and anti-unitary)

$$\text{Tr} \left( B^\dagger \pi_0 P^T_t(\alpha) A \right) = \text{Tr} \left( (K A^\dagger) \pi^R_0 P^T_{t_0} (1 - \alpha) (K B) \right). \tag{9}$$

This equation, which is the essence of the quantum fluctuation theorem, expresses a generalized detailed balance condition. For a stationary state $\pi_t = \pi$ (so that the $\alpha$-dependence drops out) that is also reversible (i.e. $P^T_{t_0} = P^T_{t_0}$), Eq. (8) becomes equivalent to the detailed balance condition of [22].
Equation (9) can be brought into a more familiar form by introducing
\[ W_t = -\langle \pi_t^{-1} \partial_t \pi_t \rangle \quad \text{and} \quad W_t^R = -\langle \pi_t^R \rangle^{-1} \partial_t \pi_t^R, \] (10)
(in the classical limit, these operators reduce to the injected power), and by using the following relation, valid for two operators \( X \) and \( Y \)
\[ \text{Tr} \left( \pi_0 Y P_T^R(\alpha) X \right) = \left\langle Y(0) \exp \left\{ \alpha \int_0^T W_u du \right\} X(T) \right\rangle. \] (11)
This formula is proved by expanding \( P_T^R(\alpha) \) w.r.t. the deformation parameter \( \alpha \) (Dyson expansion), rewriting the trace as a correlation function via Eq. (4) and finally identifying the result with the r.h.s. [29]. Inserting the symmetry condition into equation (11), the Fluctuation Relation for an open quantum Markovian system is obtained:
\[ \left\langle \left\langle \pi_0 B \pi_0^{-1} \right\rangle \left( \pi_0^R \right)^{\dagger} (0) \exp \left\{ -\alpha \int_0^T W_u du \right\} A(T) \right\rangle = \] (12)
\[ \left\langle \left\langle \pi_0^R (KA) \left( \pi_0^R \right)^{-1} \right\rangle \left( 0 \right) \exp \left\{ -\left( 1 - \alpha \right) \int_0^T W_u^R du \right\} (KB)(T) \right\rangle^R. \]
This identity is original, it encodes the main results of our work and will allow us to derive various relations for quantum systems far from equilibrium. If we interpret the mean values as classical averages and the operators as commuting c-numbers, then Eq. (12) becomes Crooks' relation. In the quantum case, if we take \( A = B = \mathbb{1} \) and \( \alpha = 1 \), then Eq. (12) yields a quantum analog of the Jarzynski identity:
\[ \left\langle \exp \left\{ -\alpha \int_0^T W_u du \right\} \right\rangle = 1. \] (13)
This quantum Jarzynski relation was first derived in [13], but the operator-ordering issue was not accurately taken into account [30].

We now derive a Quantum Fluctuation-Dissipation Theorem for a system in the vicinity of an equilibrium steady state. Suppose that the Lindbladian is given by \( L_t = L_0 + h^a(t) M_a \) where \( L_0 \) is time-independent with invariant density-matrix given by \( \pi_0 \). The time-dependent perturbations \( h^a(t) \) are supposed to be small and a summation over the repeated index \( a \) is understood. At first order, the accompanying density-matrix \( \pi_t \), with \( \dot{L}^t \pi_t = 0 \), is given by \( \pi_t = \pi_0 + h^a(t) \epsilon_a \) where \( \epsilon_a \) satisfies \( \dot{L}^t \epsilon_a = M_a \pi_0 \) and \( W_t \) defined in (10) reads \( W_t \equiv -h^a(t) D_a \) with \( D_a = \pi_0^{-1} \epsilon_a \). Using Eq. (12) with an arbitrary operator \( A \), with \( \alpha = 1 \) and \( B = \mathbb{1} \), and taking its functional derivative w.r.t. \( h^a(u) \) with \( u < T \), we obtain
\[ \left. \frac{\delta}{\delta h^a(u)} \left( A(T) \right) \right|_{h=0} + \left. \frac{\delta}{\delta h^a(u)} \left( -\alpha \int_0^T W_u du \right) \right|_{h=0} A(T) \right\rangle_0 = 0. \] (14)
Using the first order expansions derived above, we obtain
\[ \left. \frac{\delta}{\delta h^a(u)} \left( A(T) \right) \right|_{h=0} = \frac{d}{du} \left( D_a(u) A(T) \right)_0. \] (15)
We emphasize that the expectation value on the r.h.s. is taken with respect to the unperturbed density matrix \( \pi_0 \). By choosing \( A_T = D_0(T) \), Eq. (15) becomes structurally similar to the usual equilibrium fluctuation dissipation theorem. This generalizes to the quantum case, a recently obtained result for classical systems [24] (see [22] for an alternative approach).

In the last part of this work, we consider the special case of an isolated system. Here, the Lindbladian reduces to the Liouville operator: \( L_t X = i[H_t, X] \). The Hamiltonian is time-dependent but the evolution of the system is unitary and in this framework the general relation (12) reduces to the recently obtained Quantum work relations [3–6]. For a closed system, the evolution superoperator on an observable \( X \) reduces to the unitary action \( P_T^R X = (U_0^T)^\dagger X U_0^T \), where the evolution operator \( U_0^T = \exp \left\{ \int_0^T (\dot{H}_a) du \right\} \). It can be shown then that the time-reversed system (15) is also valid, with Hamiltonian \( H_t^R = K H_t \) and evolution operator \( U_0^{R,T} = K (U_T^T)^\dagger \) [31].
Using identity (8) for \( \alpha = 1 \) and the fact that \( K \) is multiplicative, we find that \( P_0^T (\alpha = 1) A \) is equal to

\[
\pi_0^{-1} K \left( (P_0^{T,R})^\dagger K (\pi_T A) \right) = \pi_0^{-1} K \left( U_0^{T,R} K (\pi_T A) \left( U_0^{T,R} \right)^\dagger \right) = \pi_0^{-1} K \left( U_0^R \right)^\dagger \pi_T A U_0^T.
\]

(16)

Substituting the last expression in Eq. (9) leads to

\[
Tr \left( B^\dagger \pi_0 \pi_0^{-1} U_0^{T,R} \pi_T A U_0^T \right) = Tr \left( K (A^\dagger) \pi_0^R \left( U_0^{T,R} \right)^\dagger K (B) U_0^{T,R} \right).
\]

(17)

Recalling that \( \pi_0^{-1} \) and \( \pi_T \) are given by the Boltzmann law, the above equation becomes in the Heisenberg representation denoted by the superscript \( H \),

\[
Tr \left( B^\dagger \pi_0 \exp(\beta H_0^H(0)) \exp(-\beta H_T^H(T)) A^H(T) \right) = \frac{Z_T}{Z_0} Tr \left( K (A^\dagger) \pi_0^R \left( U_0^{T,R} \right)^\dagger K (B) U_0^{T,R} \right).
\]

(18)

We emphasize that for \( B = \mathbb{1} \), Eq. (17) is a tautology (because \( K \) is anti-unitary), however it implies the non-trivial result (15): this feature is characteristic of most of the derivations of the work identities. If we now take \( A = B = \mathbb{1} \), we end up with the quantum Jarzynski relation for closed systems as first found by Kurchan and Tasaki [3, 4]:

\[
Tr \left( \pi_0 \exp(\beta H_0^H(0)) \exp(-\beta H_T^H(T)) \right) = \frac{Z_T}{Z_0}.
\]

(19)

Rewriting the l.h.s. as a time-ordered exponential and using \( \frac{d}{dt} H_T^H = (\partial_t H_t)^H \) we conclude as in [2] that

\[
Tr \left( \pi_0 \bar{\exp} \left( -\beta \int_0^T (\partial_s H_s)^H (s) ds \right) \right) = \frac{Z_T}{Z_0}.
\]

(20)

Considering now a system perturbed near equilibrium with \( H_a = H - h_a^\alpha (t) O_a \), we can calculate explicitly the first order perturbation to the canonical density-matrix, \( \exp(-\beta H_t) = \exp(-\beta H) + h_a^\alpha \int_0^\beta \exp(-\alpha H) O_a \exp(\alpha H) \exp(-\beta H) d\alpha \) and find that \( D_a = -\beta \langle O_a \rangle_0 + f_0^\beta \exp(\alpha H) O_a \exp(-\alpha H) d\alpha \). Finally, the Fluctuation-Dissipation Theorem (15) becomes

\[
\frac{\delta \langle A(T) \rangle}{\delta h^\alpha (u)} \bigg|_{h=0} = i \langle O_a (A(T-u + i\beta) - A(T-u)) \rangle_0.
\]

(21)

To obtain this result, we performed the derivation w.r.t. time \( u \) on the r.h.s. of Eq. (15) and then used an analytic continuation to write the inverse temperature \( \beta \) as the imaginary part of the time, as allowed by the KMS condition [20]. This is the real space version of the celebrated result derived amongst others by Callen and Welton, and by Kubo [18, 20]. For a closed system, an alternative proof that relation (19) implies Eq. (21) is given in [3].

In this work, we have derived fluctuation relations for an open quantum system described by a Lindblad dynamics that takes into account the interactions with the environment as well as measurement processes. We prove the fluctuations relations thanks to a suitable deformation of the system’s dynamics: this key technical idea provides a truly unified picture of the fluctuations relations, whether classical or quantum, and does not require to define the concept of work at the quantum level. A possible extension of our work would be to study particles with non-zero spins such as Dirac spinors. Besides, choosing a time inversion different from that of Eq. (5) may lead to various families of fluctuation relations, as happened in the classical case [27]. Other extensions would be to derive exact solutions for specific models (such as quantum Brownian motions) thus providing experimentally testable predictions. Finally, the investigation of time-reversal properties of general non-Markovian quantum systems, in which the characteristic time scale of the environment cannot be neglected w.r.t. that of the system, should also yield interesting fluctuation relations.

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[29] In full rigor, the integrand is \( \int_{0}^{T} du W_{1}(u) \) because a supplementary time dependence is introduced through the use of Eq. \( \text{(4)} \), that should appear as the argument of the time-dependent \( W_{1} \) operator.

[30] Note that the Jarzynski relation does not involve the time-reversed dynamics. Thus, the proof of Eq. \( \text{(13)} \) does not require to assume that \( L_{\text{R}}^{\text{R}} \) is a Lindbladian. In particular, no quantum microreversibility condition is needed.

[31] For a spin-0 particle in a magnetic field, the time-inversion operation \( \text{(5)} \) is supplemented by the requirement that the reversed system evolves with vector potential \( A_{\text{R}}^{\text{R}} = -A_{\text{t}} \).