Quantum Bochkov-Kuzovlev Work
Fluctuation Theorems

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The quantum version of the Bochkov-Kuzovlev identity is derived on the basis of the appropriate definition of work as the difference of the measured internal energies of a quantum system at the beginning and at the end of an external action on the system given by a prescribed protocol. According to the spirit of the original Bochkov-Kuzovlev approach, we adopt the “exclusive” viewpoint, meaning that the coupling to the external work-source is not counted as part of the internal energy. The corresponding canonical and microcanonical quantum fluctuation theorems are derived as well, and are compared to the respective theorems obtained within the “inclusive” approach. The relations between the quantum inclusive-work \( w \), the exclusive-work \( w_0 \) and the dissipated-work \( w_{\text{dis}} \), are discussed and clarified. We show by an explicit example that \( w_0 \) and \( w_{\text{dis}} \) are distinct stochastic quantities obeying different statistics.

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1. Introduction

One of the main objectives of nonequilibrium thermodynamics is the study of the response of physical systems to applied external perturbations. Around the mid of the last century major advancements were obtained in this field with the development of linear response theory by several authors, among which we mention Cullen & Welton (1951), Green (1952), Kubo (1957). This theory, inspired by the works of Einstein (1926) on the Brownian movement and of Johnson (1928) and Nyquist (1928) on noise in electrical circuits, established that, under certain circumstances, the linear response to small perturbations is determined by the equilibrium fluctuations of the system. In particular, the linear response coefficients are proportional to two-point correlation functions for Hamiltonian systems (Kubo, 1957) as well as for stochastic, generally non-equilibrium systems (Hänggi & Thomas, 1982). In principle, an infinite hierarchy of higher order fluctuation-dissipation relations connects the \( n \)-th order response coefficients to \((n+1)\)-point correlation functions.

In contrast, fluctuation theorems are compact relations that provide information about the fully nonlinear response. Accordingly, fluctuation-dissipation relations of all orders can be derived therefrom. Bochkov & Kuzovlev (1977, 1981) were the first to put forward one such fully nonlinear fluctuation theorem. These authors noticed that, for a classical system, their general fluctuation theorem implies the following, extremely simple nonequilibrium identity:

\[ \langle e^{-\beta W_0} \rangle = 1 \]

(1.1)
where $W_0$ is the work done on the system by the external perturbation during one specific realization thereof, $\langle \cdot \rangle$ denotes the average over many realizations of the same perturbation, and $\beta = (k_B T)^{-1}$, with $T$ the initial temperature of the system and $k_B$ Boltzmann’s constant. Due to the properties of convexity of the exponential function, an almost immediate consequence of (1.1), is the second law of thermodynamics in the form, $\langle W_0 \rangle \geq 0$; i.e. when a system is perturbed from an initial thermal equilibrium, on average, it can only absorb energy.

The works of Bochkov & Kuzovlev (1977, 1981) has recently re-gained a great deal of attention, after Jarzynski (1997) derived, within the framework of classical mechanics, a salient result similar to Eq. (1.1):

$$\langle e^{-\beta W} \rangle = e^{-\beta \Delta F} \quad (1.2)$$

which, in contrast to Eq. (1.1), allows to extract an equilibrium property of the system, i.e., its free energy (difference) $F$, from nonequilibrium fluctuations of work $W$. Evidently, the definitions of work adopted by Jarzynski (1997) and Bochkov & Kuzovlev (1977, 1981) (denoted here respectively as $W$ and $W_0$) do not coincide. The relationships between these two work definitions and the corresponding nonequilibrium identities, Eqs. (1.1,1.2), were discussed in a very clear and elucidating manner in Jarzynski (2007), which, for the sake of clarity, we summarize below.

Let us express the time dependent Hamiltonian of the driven classical system as the sum of the unperturbed system Hamiltonian $H_0$ and the interaction energy stemming from the coupling of the external time dependent perturbation $X(t)$ to a certain system observable $Q$:

$$H(q,p;t) = H_0(q,p;t) - X(t)Q(q,p) \quad (1.3)$$

We restrict ourselves to the simplest case of a protocol governed by a single “field” $X(t)$ coupling to the conjugated generalized coordinate $Q$. Generalization to several fields $X_i(t)$ coupling to different generalized coordinates $Q_i$ is straightforward.

The definition of work $W$ according to Jarzynski (1997) stems from an inclusive viewpoint, where one counts the term $X(t)Q$ as being part of the system internal energy. In contrast the definition of work $W_0$ according to Bochkov & Kuzovlev (1977, 1981) belongs to an exclusive viewpoint where instead, such term is not counted as part of the energy of the system. More explicitly, if $q_0, p_0$ is a certain initial condition that evolves to $q_f, p_f$ in a time $t_f - t_0$, according to the Hamiltonian evolution generated by $H$, then the two different definitions of work become:

$$W \doteq H(q_f, p_f; t_f) - H(q_0, p_0; t_0) \quad (1.4)$$

$$W_0 \doteq H_0(q_f, p_f) - H_0(q_0, p_0) \quad (1.5)$$

It is important to stress that Bochkov & Kuzovlev (1977, 1981) only obtained Eq. (1.1) in the classical case, notwithstanding the fact that they developed a quantum version of their theory, as well. This difficulty is related to the fact that work was identified by Bochkov & Kuzovlev (1977, 1981) with the quantum expectation of a pretended work operator, given by the difference of final and initial Hamiltonian in the Heisenberg representation. To be more clear, if the quantum Hamiltonian reads:

$$H(t) = H_0 - X(t)Q$$

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where now $H, H_0$ and $Q$ are hermitian operators, the work operator was defined by Bochkov & Kuzovlev (1977, 1981) as:

$$W_0 \equiv H_0^H(t_f) - H_0$$

(1.7)

where the superscript $H$ denotes Heisenberg picture. A similar approach was employed also within the inclusive viewpoint, with work defined as (Allahverdyan & Nieuwenhuizen, 2005)

$$W = H^H(t_f) - H_0$$

(1.8)

As pointed out clearly by some of us before with the work in (Talkner et al., 2007) the Jarzynski Equality (1.2) cannot be obtained on the basis of the work operator (1.8). Likewise the Bochkov-Kuzovlev identity (1.1) cannot be obtained on the basis of Eq. (1.7). It is by now clear that the impossibility of extending the classical results (1.1,1.2) on the basis of quantum work operators (1.7,1.8), respectively, is related to the fact that work characterizes a process, rather than a state of the system, and consequently cannot be represented by an observable that would yield work as the result of a single projective measurement. In contrast, energy must be measured twice in order to determine work, once before the protocol starts and a second time immediately after it has ended. The difference of the outcomes of these two measurements then yields the work performed on the system in a particular realization (Talkner et al., 2007).

In this paper we will adopt the exclusive viewpoint of Bochkov & Kuzovlev (1977, 1981), but use the proper definition of work as the difference between the outcomes of two projective measurements of $H_0$, to obtain the quantum version of Eq. (1.1). Indeed we will develop the theory of quantum work fluctuations within the exclusive two-point measurements viewpoint in great generality. In Sec. 2 we study the characteristic function of work. In Sec. 3 and Sec. 4 we derive the exclusive versions of the Tasaki-Crooks quantum fluctuation theorem (Talkner & Hänggi, 2007; Talkner, Campisi & Hänggi, 2008; Campisi et al., 2009), and of the microcanonical quantum fluctuation theorem (Talkner, Hänggi & Morillo, 2008), respectively. Sec. 5 closes the paper with some remarks concerning the relationships between the inclusive-work, the exclusive-work, and the dissipated-work.

### 2. Characteristic function of work

As mentioned in the introduction, work is properly defined in quantum mechanics as the difference of the energies measured at the beginning and end of the protocol, i.e., at times $t_0$ and $t_f > t_0$, respectively. According to the exclusive viewpoint that we adopt here, this energy is given by the unperturbed Hamiltonian $H_0$. Let $e_n$ and $|n, \lambda\rangle$, denote the eigenvalues and eigenvectors of $H_0$:

$$H_0 |n, \lambda\rangle = e_n |n, \lambda\rangle$$

(2.1)

Here $n$ is the quantum number labeling the eigenvalues of $H_0$ and $\lambda$ denotes further quantum numbers needed to specify uniquely the state of the system, in case of

Bochkov & Kuzovlev (1977) defined the “operator of energy absorbed by the system” $E = \int_{t_0}^{t_f} X(\tau)Q^H(\tau)d\tau$, where $Q^H(\tau)$ is the operator $Q$ in the Heisenberg representation. It is not difficult to prove that $E$ coincides with $W_0$ in Eq. (1.7).
degenerate energies. A measurement of $H_0$ at time $t_0$ gives a certain eigenvalue $e_n$ while a subsequent measurement of $H_0$ at time $t_f$ gives another eigenvalue $e_m$, so that

$$w_0 = e_m - e_n \quad (2.2)$$

Evidently $w_0$ is a stochastic variable due to the intrinsic randomness entailed in the quantum measurement processes and possibly in the statistical mixture nature of the initial preparation. In the following we derive the statistical properties of $w_0$, in terms of its probability density function (pdf), and the associated characteristic function of work.

Let the system be prepared at time $t < t_0$ in a certain state described by the density matrix $\rho(t_0)$. We further assume that the perturbation $X(t)$ is switched on at time $t_0$. At the same time the first measurement of $H_0$ is performed, with outcome $e_n$. This occurs with probability:

$$p_n = \sum_\lambda \langle n, \lambda | \rho(t_0) | n, \lambda \rangle = \text{Tr} P_n \rho(t_0) \quad (2.3)$$

where $P_n$ is the projector onto the eigenspace spanned by the eigenvectors belonging to the eigenvalue $e_n$:

$$P_n = \sum_\lambda | n, \lambda \rangle \langle n, \lambda | \quad (2.4)$$

and $\text{Tr}$ denotes trace over the Hilbert space. According to the postulates of quantum mechanics, immediately after the measurement the system is found in the state:

$$\rho_n = P_n \rho(t_0) P_n / p_n \,. \quad (2.5)$$

For times $t > t_0$ the system evolves according to

$$\rho(t) = U_{t,t_0} \rho_n U_{t,t_0}^\dagger \quad (2.6)$$

with $U_{t,t_f}$ denoting the unitary time evolution operator obeying the Schrödinger equation governed by the full time dependent Hamiltonian (Eq. 1.6):

$$i \hbar \frac{\partial}{\partial t} U_{t,t_0} = H(t) U_{t,t_0} \,, \quad U_{t_0,t_0} = 1 \,.$$

At time $t_f$ the second measurement of $H_0$ is performed, and the eigenvalue $e_m$ is obtained with probability:

$$p(m|n) = \text{Tr} P_m \rho_n(t_f) \,. \quad (2.8)$$

Therefore the probability density to observe a certain value of work $w_0$ is given by:

$$p_{0}^{t_f,t_0}(w_0) = \sum_{m,n} \delta(w_0 - [e_m - e_n]) p(m|n) p_n \,. \quad (2.9)$$

We use the superscript 0 throughout this paper to indicate the exclusive viewpoint. The same symbols, without the superscript 0 denote the respective quantities within the inclusive viewpoint.

The Fourier transform of the probability density of work gives the characteristic function of work

$$G_{t_f,t_0}^0(u) = \int dw_0 p_{0}^{t_f,t_0}(w_0) e^{i u w_0} \quad (2.10)$$
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which allows quick derivations of fluctuation theorems and nonequilibrium equalities. Performing calculations analogous to those reported by Talkner, Hänggi & Morillo (2008) we find the characteristic function of work, in the form of a two point quantum correlation function:

$$G^0_{t_f, t_0}(u) = \text{Tr} e^{iuH^H(t_f)} e^{-iuH_0} \bar{\rho}(t_0) \equiv \langle e^{iuH^H(t_f)} e^{-iuH_0} \rangle$$ (2.11)

where $\bar{\rho}(t_0)$ is defined as:

$$\bar{\rho}(t_0) = \sum_n p_n \rho_n = \sum_n P_n \rho(t_0) P_n$$ (2.12)

and the superscript $H$ stands for Heisenberg representation, i.e.:

$$H^H_{t_f, t_0} = U^\dagger_{t_f, t_0} H_0 U_{t_f, t_0}$$ (2.13)

This exclusive-work characteristic function $G^0_{t_f, t_0}$ should be compared to the inclusive-work characteristic function $G_{t_f, t_0}$ that is obtained when looking at the difference $w$ of the outcomes $E_n(t_0)$ and $E_m(t_f)$ of measurements of the total time dependent Hamiltonian $H(t)$. In this case one obtains (Talkner et al., 2007; Talkner, Hänggi & Morillo, 2008):

$$G_{t_f, t_0}(u) = \text{Tr} e^{iuH(t_f)} e^{-iuH_0} \bar{\rho}(t_0) \equiv \langle e^{iuH(t_f)} e^{-iuH_0} \rangle$$ (2.14)

The difference lies in the distinct fact that $H^H_{t_f, t_0}$ appears in the exclusive approach in place of the full $H^H(t_f)$.

(a) Reversed protocol

Consider next the reversed protocol

$$\tilde{X}(t) = X(t_f + t_0 - t)$$ (2.15)

which consecutively assumes values as if time was reversed. Let $\tilde{H}(t)$ be the resulting Hamiltonian:

$$\tilde{H}(t) = H_0 - \tilde{X}(t)Q$$ (2.16)

The characteristic function of work now reads:

$$\tilde{G}_{t_f, t_0}(u) = \text{Tr} e^{iu\tilde{H}(t_f)} e^{-iuH_0} \tilde{\rho}(t_0) \equiv \langle e^{iu\tilde{H}(t_f)} e^{-iuH_0} \rangle$$ (2.17)

where

$$\tilde{H}^H_{t_f, t_0} = \tilde{U}^\dagger_{t_f, t_0} H_0 \tilde{U}_{t_f, t_0}$$ (2.18)

and $\tilde{U}_{t_f, t_0}$ is the time evolution operator generated by $\tilde{H}(t)$:

$$i\hbar \partial_t \tilde{U}_{t_f, t_0} = \tilde{H}(t) \tilde{U}_{t_f, t_0} \quad \tilde{U}_{t_0, t_0} = 1$$ (2.19)

Assuming that the Hamiltonian $H(t)$ is invariant under time reversal i.e.:

$$\Theta H(t) \Theta^{-1} = H(t)$$ (2.20)

Here we assume that the Hamiltonian does not depend on any odd parameter, e.g., a magnetic field. Treating that case is straightforward and amounts to reverse the sign of the odd parameter in the r.h.s. of Eq. (2.20), see Andrieux et al., 2009.

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where Θ is the antiunitary time reversal operator (Messiah, 1962), the time evolution operators associated to the forward and backward protocols are related by the following important relation, see Appendix A:

\[ U_{t_f,t_0} = U_{t_f,t_0}^\dagger = \Theta \tilde{U}_{t_f,t_0} \Theta^{-1}. \]  

(2.21)

In the following section we will derive the quantum version of Eq. (1.1) and its associated work fluctuation theorem. This will be accomplished by choosing the initial density matrix to be a Gibbs canonical state. In Sec. 4 we will, instead, assume an initial microcanonical state.

3. Canonical initial state

For a system staying at time \( t_0 \) in a canonical Gibbs state:

\[ \rho(t_0) = \bar{\rho}(t_0) = e^{-\beta H_0}/Z_0 \]  

(3.1)

where \( Z_0 = Tr e^{-\beta H_0}, \bar{\rho}(t_0) \) coincides with \( \rho(t_0) \) because the latter is diagonal with respect to the eigenbasis of \( H_0 \) (see Eq. 2.12). Plugging Eq. (3.1) into (2.11), we obtain:

\[ G_{t_f,t_0}^0(\beta; u) = \text{Tr} e^{iuH_0(t_f)} e^{-iuH_0} e^{-\beta H_0}/Z_0 \]  

(3.2)

where for completeness we have listed the dependence upon \( \beta \) among the arguments of \( G_{t_f,t_0}^0 \). The quantum version of Eq. (1.1) immediately follows by setting \( u = i\beta \):

\[ \langle e^{-\beta w_0} \rangle = G_{t_f,t_0}^0(\beta; i\beta) = \text{Tr} e^{-\beta H_0(t_f)}/Z_0 = \text{Tr} e^{-\beta H_0}/Z_0 = 1 \]  

(3.3)

where in the third equation we have used Eq. (2.13), the cyclic property of the trace and the unitarity of the time evolution operator: \( U_{t_f,t_0}^\dagger U_{t_f,t_0} = 1 \).

Moreover we find the following important relation between \( G_{t_f,t_0}^0 \) and \( \tilde{G}_{t_f,t_0}^0 \), see Appendix B:

\[ G_{t_f,t_0}^0(\beta; u) = \tilde{G}_{t_f,t_0}^0(\beta; -u + i\beta). \]  

(3.4)

By means of inverse Fourier transform, the following quantum Bochkov-Kuzovlev fluctuation relation between the forward and backward work probability density functions is obtained:

\[ \frac{p_{t_f,t_0}^0(\beta; w_0)}{\tilde{p}_{t_f,t_0}(\beta; -w_0)} = e^{\beta w_0}. \]  

(3.5)

This must be compared to the quantum Tasaki-Crooks relation that is obtained within the inclusive viewpoint (Talkner & Hänggi, 2007):

\[ \frac{p_{t_f,t_0}(\beta; w)}{\tilde{p}_{t_f,t_0}(\beta; -w)} = e^{\beta(w - \Delta F)} \]  

(3.6)

where, in contrast to Eq. (3.5) the term \( \Delta F = -\beta^{-1} \ln \text{Tr} e^{-\beta H(t)} - \ln \text{Tr} e^{-\beta H_0} \), appears.
(a) Remarks

Eqs. (3.3, 3.5) constitute original quantum results that do not appear in the works of Bochkov & Kuzovlev [1977, 1981]. In the classical case they found a fluctuation theorem similar to Eq. (3.7), reading:

\[
P[Q(\tau); X(\tau)] = \exp \left[ \beta \int_{t_0}^{t_f} X(\tau) \delta(\tau) \right]
\]  

where \( P[Q(\tau); X(\tau)] \) is the probability density functional to observe a certain trajectory \( Q(\tau) \) given a certain protocol \( X(\tau) \). Here \( Q(\tau) \) is a short hand notation for \( Q(q(q_0, p_0, \tau), p(q_0, p_0, \tau)) \), see Eq. (3.3), where \( (q(q_0, p_0, \tau), p(q_0, p_0, \tau)) \) is the evolved initial condition \( q_0, p_0 \) at some time \( \tau \in [t_0, t_f] \), for a certain protocol \( X(\tau) \). The symbol \( \varepsilon \) denotes the parity of the observable \( Q \) under time reversal (assumed to be equal to 1 in this paper). The symbol \( \sim \) denotes quantities referring to the reversed protocol. The classical probability of work \( p^{cl,0}_{t_f, t_0}(W_0) \) is obtained from the classical trajectory probability density functional \( P[Q(\tau); X(\tau)] \) via the formula:

\[
p^{cl,0}_{t_f, t_0}(W_0) = \int DQ(\tau) P[Q(\tau); X(\tau)] \delta \left[ W_0 - \int_{t_0}^{t_f} X(\tau) \delta(\tau) \right]
\]  

where the integration is a functional integration over all possible trajectories such that \( \int_{t_0}^{t_f} X(\tau) \delta(\tau) = W_0 \). With this formula one finds from Eq. (3.7) the exclusive version of the classical Crooks fluctuation theorem for the work probability densities (Jarzynski & Horowitz, 2007)

\[
p^{cl,0}_{t_f, t_0}(\beta; W_0) = p^{cl,0}_{t_f, t_0}(\beta; -W_0) e^{\beta W_0}.
\]  

Notably, a quantum version of Eq. (3.7) does not exists because: “in the quantum case it is impossible to introduce unambiguously a […] probability functional” (Bochkov & Kuzovlev, 1981). It is only by giving up the idea of true quantum trajectories and embracing instead the two-point measurement approach that the quantum exclusive fluctuation theorem Eq. (3.5) can be obtained, and has been obtained here, for the first time.

4. Microcanonical initial state

We consider next an initial microcanonical initial state of energy \( E \), that can formally be expressed as:

\[
\rho(t_0) = \bar{\rho}(t_0) = \delta(H_0 - E) / \Omega_0(E),
\]  

wherein \( \Omega_0(E) = Tr \delta(H_0 - E) \). Actually one has to replace the singular Dirac function \( \delta(x) \) by a smooth function sharply peaked around \( x = 0 \), but with infinite support. A normalized gaussian with arbitrarily small width serves this purpose well.

With this choice of initial condition, the characteristic function of work reads:

\[
\begin{align*}
G^{0}_{t_f, t_0}(E; u) &= Tr e^{iuH^0(t_f)} e^{-iuH_0} \delta(H_0 - E) / \Omega_0(E) \\
&= Tr e^{iu[H^0(t_f) - E]} \delta(H_0 - E) / \Omega_0(E)
\end{align*}
\]  

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where for completeness we listed the dependence upon $E$ among the arguments of $G_{t_f, t_0}^0$. By applying the inverse Fourier transform we obtain:

$$
\tilde{p}_{t_f, t_0}^0(E; w_0) = \text{Tr} \left( \delta(H_0^H(t_f) - E - w_0) \delta(H_0 - E) / \Omega_0(E) \right) \tag{4.3}
$$

Likewise, for the reversed protocol,

$$
\tilde{p}_{t_f, t_0}^0(E; w_0) = \text{Tr} \left( \delta(H_0^H(t_f) - E - w_0) \delta(H_0 - E) / \Omega_0(E) \right) \tag{4.4}
$$

is found.

We then find the following relation between the forward and backward work probability densities, see Appendix C.

$$
\Omega_0(E) \tilde{p}_{t_f, t_0}^0(E; w_0) = \Omega_0(E + w_0) \tilde{p}_{t_f, t_0}^0(E + w_0; -w_0) \tag{4.5}
$$

Then, the quantum microcanonical fluctuation theorem reads, within the exclusive viewpoint:

$$
\frac{\tilde{p}_{t_f, t_0}^0(E; w_0)}{\tilde{p}_{t_f, t_0}^0(E + w_0; -w_0)} = \frac{\Omega_0(E + w_0)}{\Omega_0(E)} \tag{4.6}
$$

This must be compared to the quantum microcanonical fluctuation theorem, obtained within the inclusive viewpoint (Talkner, Hänggi & Morillo, 2008)

$$
\frac{p_{t_f, t_0}(E; w)}{p_{t_f, t_0}(E + w; -w)} = \frac{\Omega_f(E + w)}{\Omega_0(E)} \tag{4.7}
$$

The difference lies in the fact that within the exclusive viewpoint the densities of states at the final energy $E + w_0$, is determined by the unperturbed Hamiltonian, i.e., $\Omega_0(E + w_0) = \text{Tr} \delta(H_0 - (E + w_0))$, whereas it results from the total Hamiltonian in the inclusive approach: $\Omega_f(E + w) = \text{Tr} \delta(H(t_f) - E - w)$.

Eq. (4.7) was first obtained within the classical framework by (Cleuren et al., 2006). It is not difficult to see that Eq. (4.6) holds classically as well.

(a) Remarks

Just as Eq. (4.5), this Eq. (4.6) is a new result that was not reported before by Bochkov & Kuzovlev (1977, 1981). It is very interesting to notice, however, that those authors already put forward a classical fluctuation theorem for the microcanonical ensemble, which can be recast in the form (Bochkov & Kuzovlev, 1981):

$$
\frac{P[\tau ; X(\tau); E]}{P[\tau ; -\dot{X}(\tau); \dot{X}(\tau); E + W_0]} = \frac{\Omega_0(E + W_0)}{\Omega_0(E)} \tag{4.8}
$$

where $P[\tau; X(\tau); E]$ is the probability density functional to observe a certain trajectory $I(\tau)$ given a certain protocol and an initial microcanonical ensemble of energy $E$. Here

$$
I(\tau) = \dot{Q}(q(0), p_0, \tau), \quad p(q_0, p_0, \tau) \tag{4.9}
$$

denotes the current. By functional integration the classical microcanonical theorem for the pdf of work

$$
\frac{p^{cl}_{t_f, t_0}(E, W_0)}{p^{cl}_{t_f, t_0}(E + W_0, W_0)} = \frac{\Omega_0(E + W_0)}{\Omega_0(E)} \tag{4.10}
$$

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is obtained from (4.8) in the same way as (3.5) follows from (3.7). However the quant-
utm version of (4.8) does not exists and the derivation of the quantum microcan-
nical fluctuation theorem (4.6) is indeed only possible if the two-point measurement
approach is adopted.

The fluctuation relations of Eqs. (4.6, 4.7) can be further expressed in terms of
entropy, according to the rules of statistical mechanics. Following Gibbs (1902) two
different prescriptions are found in textbooks to obtain the entropy associated to
the microcanonical ensemble:

\[ s(E) = k_B \ln \Omega(E) = \text{Tr} \delta(H - E) \]  
\[ S(E) = k_B \ln \Phi(E) = \text{Tr} \theta(H - E) \]  

The two definitions coincide for large systems with short range inter-
actions among their constituents, but may substantially differ if the size of the sys-
tem under study is small. It is by now clear that, of the two, only the second – cu-
stomarily called “Hertz entropy” – is the fundamentally correct one (Hertz (1910a, b);
Schlüter (1948); Pearson et al. (1985); Campisi (2005, 2008a, 2010); Dunkel & Hilbert
(2006)).

† Using the microcanonical expression for the temperature
\[ k_B T(E) = \left( \frac{\partial S(E)}{\partial E} \right)_{\Omega(E)} = \frac{\Phi(E)}{\Omega(E)} \]
we can re-express the quantum microcanonical Bochkov-Kuzovlev fluct-
uation relation in terms of entropy and temperature as:

\[ \frac{p^0_{t_f, t_0}(E; w_0)}{\tilde{p}^0_{t_f, t_0}(E + w_0; -w_0)} = \frac{T_0(E)}{T_0(E + w)} \exp\left[ \frac{S_0(E + w_0) - S_0(E)}{k_B} \right] \]  

where the subscript 0 in \( T \) and \( S \) denotes that these quantities are calculated for
the unperturbed Hamiltonian \( H_0 \). Likewise, adopting the inclusive viewpoint one
obtains:

\[ \frac{p_{t_f, t_0}(E; w)}{\tilde{p}_{t_f, t_0}(E + w; -w)} = \frac{T_f(E)}{T_f(E + w)} \exp\left[ \frac{S_f(E + w) - S_0(E)}{k_B} \right] \]  

where the subscript \( f \) in \( T \) and \( S \) denotes that these quantities are calculated for
the total final Hamiltonian \( H(t_f) \).

5. Discussion

We derived the quantum Bochkov-Kuzovlev identity as well as the quantum can-
nical and microcanonical work fluctuation theorems within the exclusive approach,
and have elucidated their relations to the original works of Bochkov & Kuzovlev
(1977, 1981). The extension of the corresponding classical theorems to the quan-
tum regime is only possible thanks to the proper definition of work as a two-time
quantum observable. We close with two comments:

1. For a cyclic process, \( X(t_f) = X(t_0) \), inclusive and exclusive work fluctuation
theorems coincide. However in no way is it true that the exclusive approach of

† It is interesting to notice that Einstein was well aware of the works of Hertz (1910a, b) which
he praised as excellent (“vortrefflich”) (Einstein 1911).

‡ If instead of the microcanonical ensemble (4.1), the modified microcanonical ensemble \( \tilde{g}(t_0) = \theta(E - H_0)/[\text{Tr} \theta(E - H_0)] \) (Ruelle 1969) would be used as the initial equilibrium state, then the
fluctuation theorem assumes the same form as in Eq. (4.14), but without the ratio of temperatures
(Talkner, Hänggi & Morillo 2008).

¶ Similar remarks were made also within the classical framework by Jarzynski (2007).
Bochkov & Kuzovlev, adopted here, is restricted to cyclic processes, as some authors have suggested (Allahverdyan & Nieuwenhuizen, 2005; Cohen & Mauzerall, 2005; Andrieux & Gaspard, 2008). As stressed in the introduction, the difference of the two approaches originates from the different definitions of work, and is not related to whether the process under study is cyclic or is not cyclic.

2. Within the inclusive approach it is natural to define the dissipated work as

\[ w_{\text{dis}} = w - \Delta F \]  

(Kawai et al., 2007; Vaikuntanathan & Jarzynski, 2009). Then, the Jarzynski equality (1.2) can be rewritten as \( \langle e^{-\beta w_{\text{dis}}} \rangle = 1 \). This might make one believe that the exclusive work \( w_0 \) coincides with the dissipated work \( w_{\text{dis}} \).

This, though, would be generally wrong. The dissipated work \( w_{\text{dis}} \) is a stochastic quantity whose statistics, given by \( p_{t_f,t_0}^{\text{dis}}(w_{\text{dis}}) = p_{t_f,t_0}(w_{\text{dis}} + \Delta F) \), in general does not coincide with the statistics of exclusive work \( w_0 \), given by \( p_{t_f,t_0}^{\beta}(w_0) \). See Appendix D for a counterexample. Only for a cyclic process, for which \( \Delta F = 0 \), does the dissipated-work \( w_{\text{dis}} \) coincide with the inclusive-work \( w \), which in turn coincides with the exclusive-work \( w_0 \).

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Appendix A. Derivation of Eq. (2.21)

The time evolution operator \( \tilde{U}_{t_f,t_0} \) can be expressed as a time ordered product (Schleich, 2001):

\[
\tilde{U}_{t_f,t_0} = \lim_{N\to\infty} e^{-i\tilde{H}(t_N)\tau} e^{-i\tilde{H}(t_{N-2})\tau} \cdots e^{-i\tilde{H}(t_1)\tau} \tag{A 1}
\]

where \( \tau = (t_f - t_0)/N \), and \( t_\nu = t_0 + \nu\tau \), for \( \nu = 0, \ldots, N \) (note that \( t_N = t_f \)). Due to Eqs. (2.15) (2.16), it is \( \tilde{H}(t) = H(t_f + t_0 - t) \), then:

\[
\tilde{U}_{t_f,t_0} = \lim_{N\to\infty} e^{-iH(t_1)\tau} e^{-iH(t_2)\tau} \cdots e^{-iH(t_N)\tau} \tag{A 2}
\]

Therefore:

\[
\Theta \tilde{U}_{t_f,t_0} \Theta^{-1} = \lim_{N\to\infty} \Theta e^{-iH(t_1)\tau} \Theta^{-1} \Theta e^{-iH(t_2)\tau} \Theta^{-1} \cdots \Theta e^{-iH(t_N)\tau} \Theta^{-1} \tag{A 3}
\]

where we inserted \( \Theta^{-1} \Theta = 1, \ldots, N - 1 \) times. Due to the property (2.20) and the antilinearity of \( \Theta \) it is

\[
\Theta e^{-iH(t)\tau} \Theta^{-1} = e^{iH(t)\tau} \tag{A 4}
\]

Using this equation, we find:

\[
\Theta \tilde{U}_{t_f,t_0} \Theta^{-1} = \lim_{N\to\infty} e^{iH(t_1)\tau} e^{iH(t_2)\tau} \cdots e^{iH(t_N)\tau} \tag{A 5}
\]

\[
= \lim_{N\to\infty} \left[ e^{-iH(t_N)\tau} \cdots e^{-iH(t_2)\tau} e^{iH(t_1)\tau} \right]^\dagger \tag{A 6}
\]

\[
= U_{t_f,t_0}^\dagger = U_{t_0,t_f} \tag{A 7}
\]
In a similar way we also obtain:

$$U_{t_f,t_0} = \Theta \tilde{U}_{t_f,t_0} \Theta^{-1}. \quad (A 8)$$

**Appendix B. Derivation of Eq. (3.4)**

The exclusive-work characteristic function reads (2.11)

$$G_{t_f,t_0}(\beta;\tau) = \text{Tr} U_{t_f,t_0} e^{i\tau H_0} U_{t_f,t_0}^\dagger e^{-i\tau H_0} e^{-\beta H_0} / Z_0. \quad (B 1)$$

Using Eqs. (A 7, A 8), we obtain:

$$G_{t_f,t_0}(\beta;\tau) = \text{Tr} \Theta \tilde{U}_{t_f,t_0} \Theta^{-1} e^{i\tau H_0} \tilde{U}_{t_f,t_0} \Theta^{-1} e^{-i\tau H_0} e^{-\beta H_0} \Theta \Theta^{-1} / Z_0$$

where we have inserted $\Theta \Theta^{-1} = \mathbb{1}$ at the right end. By multiplying Eq. (A 4) by $\Theta^{-1}$ to the left and by $\Theta$ to the right, we have (replacing $\tau$ with $u$)

$$\Theta^{-1} e^{iH(t)\tau} \Theta = e^{-iH(t)u}, \quad (B 2)$$

therefore:

$$G_{t_f,t_0}(\beta;\tau) = \text{Tr} \Theta \tilde{U}_{t_f,t_0} \Theta^{-1} e^{-i\beta H_0} \tilde{U}_{t_f,t_0} \Theta^{-1} e^{-i\beta H_0} \Theta \Theta^{-1} / Z_0. \quad (B 3)$$

The antilinearity of $\Theta$ implies, for any trace class operator $A$:

$$\text{Tr} \Theta A \Theta^{-1} = \text{Tr} A^\dagger. \quad (B 4)$$

Therefore:

$$G_{t_f,t_0}(\beta;\tau) = \text{Tr} e^{-\beta H_0} e^{-i\beta H_0} \tilde{U}_{t_f,t_0} \Theta^{-1} e^{-i\beta H_0} \Theta \Theta^{-1} / Z_0 \quad (B 5)$$

Using the cyclic property of the trace finally leads to

$$G_{t_f,t_0}(\beta;\tau) = \text{Tr} \tilde{U}_{t_f,t_0} e^{i(-u+i\beta) H_0} \tilde{U}_{t_f,t_0} e^{-i(-u+i\beta) H_0} e^{-\beta H_0} / Z_0 \quad (B 6)$$

$$= \tilde{G}_{t_f,t_0}(\beta;\tau). \quad (B 7)$$

**Appendix C. Derivation of Eq. (4.5)**

The microcanonical exclusive-work probability density function reads, Eq. (4.3):

$$p_{t_f,t_0}(E;w_0) = \text{Tr} \delta(H_0^R(t_f)-E-w_0) \delta(H_0-E) / \Omega_0(E) \quad (C 1)$$

Employing Eqs. (A 7, A 8), then leads to:

$$\Omega_0(E) p_{t_f,t_0}(E;w_0) = \text{Tr} \tilde{U}_{t_f,t_0} \Theta^{-1} \delta(H_0-E-w_0) \Theta \tilde{U}_{t_f,t_0} \Theta^{-1} \delta(H_0-E) \Theta \Theta^{-1} \quad (C 3)$$

where we have inserted $\Theta \Theta^{-1} = \mathbb{1}$ at the end. Being the Dirac delta a real function we have

$$\Theta^{-1} \delta(H_0-E) \Theta = \delta(H_0-E) \quad (C 4)$$
because $H_0$ is assumed to be invariant under time reversal. Then:

$$\Omega_0(E)\rho_{t_f,t_0}^0(E;w_0) = \text{Tr} \Theta \tilde{U}_{t_f,t_0} \delta(H_0 - E - w_0) \tilde{U}_{t_f,t_0}^\dagger \delta(H_0 - E) \Theta^{-1}. \quad (C 5)$$

Using Eq. \(B 4\), we obtain:

$$\Omega_0(E)\rho_{t_f,t_0}^0(E;w_0) = \text{Tr} \delta(H_0 - E) \tilde{U}_{t_f,t_0} \delta(H_0 - E - w_0) \tilde{U}_{t_f,t_0}^\dagger. \quad (C 6)$$

Thanks to the cyclic property of the trace one finally arrives at:

$$\Omega_0(E)\rho_{t_f,t_0}^0(E;w_0) = \text{Tr} \delta(H_0 - E) \tilde{U}_{t_f,t_0}^\dagger \delta(H_0 - E - w_0) \tilde{U}_{t_f,t_0} \delta(H_0 - E - w_0) \Theta \tilde{U}_{t_f,t_0}^\dagger \delta(H_0 - E). \quad (C 7)$$

$$= \Omega_0(E + w_0)\rho_{t_f,t_0}^0(E + w_0;w_0). \quad (C 8)$$

### Appendix D. Comparison between dissipated-work and exclusive-work pdf’s

In this appendix we provide an example that shows that the dissipated-work $w_{\text{dis}}$ and the inclusive work $w_0$ are distinct stochastic quantities with different statistical properties. To this end we show that their probability density functions may have different first and second moments. We consider a driven quantum harmonic oscillator of unit mass and unit angular frequency:

$$H(t) = \frac{p^2}{2} + \frac{q^2}{2} - X(t) q$$

For simplicity we assume $t_0 = 0$, $X(t_0) = 0$, and we chose units in such a way that $\hbar = 1$. Let $|n,t\rangle$ denote the instantaneous eigenvectors of $H(t)$ corresponding to the instantaneous eigenvalues $E_n(t) = (n + 1/2) - X^2(t)/2$.

\[(a)\] The probability density of dissipated-work

The probability density function (pdf) of inclusive work, corresponding to an initial canonical state, is

$$p_{t_0}(w) = \sum_{mn} \delta(w - m + n + X^2(t)/2)|a_{mn}|^2 \epsilon^{-\beta(n+1/2)}Z(0) \quad (D 2)$$

where $Z(0) = \sum_n \epsilon^{-\beta(n+1/2)}$ is the initial partition function, and $|a_{nm}|^2$ are the probabilities to make a transition between two eigenstates of the total Hamiltonian

$$|a_{nm}|^2 = |\langle m,t|U_{t_0}n,0\rangle|^2 \quad (D 3)$$

where we have set $t_0 = 0$ and $t_f = t$. According to Talkner, Burada & Hänggi \(2008, 2009\) the mean value and the variance of the inclusive work pdf \(D 2\) are given by

$$\langle w \rangle = \int dx \ x p_{t_0}(x) = L(t) - X^2(t)/2 \quad (D 4)$$

$$\langle \Delta w^2 \rangle = \int dx \ [x - L(t)]^2 p_{t,0}(x) = 2UL(t) \quad (D 5)$$

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where $U = \sum_n (n + 1/2)e^{-\beta(n+1/2)/Z_0}$ is the initial average energy, and

$$L(t) = C(t)^2/2 + [S(t) - X(t)]^2/2$$

(D 6)

where

$$S(t) = \int_0^t ds X(s) \sin(t-s), \quad C(t) = \int_0^t ds X(s) \cos(t-s).$$

(D 7)

The partition function of work at the final time $t$ is $Z(t) = Z(0)e^{\beta X^2(t)/2}$, therefore the free energy difference $\Delta F = -\beta^{-1}\ln Z(t)/Z_0$ is given by $\Delta F = -X^2(t)/2$ (Talkner, Burada & Hänggi 2008). Hence the dissipated work is

$$w_{\text{dis}} = w + X^2(t)/2.$$ 

(D 8)

Accordingly the dissipated work pdf is

$$p^\text{dis}_{t,0}(w_{\text{dis}}) = p_{t,0}(w) = p_{t,0}(w_{\text{dis}} - X^2(t)/2)$$

$$= \sum_{mn} \delta(w_{\text{dis}} - m + n)|a_{mn}|^2 e^{-\beta(n+1/2)/Z(0)}.$$  

(D 9)

It immediately follows that

$$\langle w_{\text{dis}} \rangle = \int dx x p^\text{dis}_{t,0}(x) = L(t)$$

(D 10)

$$\langle \Delta w^2_{\text{dis}} \rangle = \int dx |x - L(t)|^2 p^\text{dis}_{t,0}(x) = 2UL(t).$$  

(D 11)

Note that, as it should be, $\langle w_{\text{dis}} \rangle \geq 0$.

(b) The probability density of exclusive-work

The exclusive-work pdf is given by

$$p^0_{t,0}(w_0) = \sum_{mn} \delta(w_0 - m + n)|a^0_{mn}|^2 e^{-\beta(n+1/2)/Z(0)}$$

(D 12)

where $|a^0_{mn}|^2$ denotes the probability to make a transition between two states of the unperturbed Hamiltonian:

$$|a^0_{mn}|^2 = |\langle m, 0 | U_{t,0} | n, 0 \rangle|^2.$$  

(D 13)

It is known (Husimi 1953; Campisi 2008) that the transition probabilities $|a_{mn}|^2$ depend on the time $t$ at which the second measurement is performed, via the function $L(t)$, that is the $|a_{mn}|^2$ are of the form $|a_{mn}|^2 = f_{nm}[L(t)]$, for certain functions $f_{nm}$ that need not be specified here. Using Wigner functions to calculate the transition probabilities as in (Campisi 2008, Appendix), we notice that the transition probabilities are of the form

$$|a^0_{mn}|^2 = |\langle m, 0 | U_{t,0} | n, 0 \rangle|^2.$$  

(D 13)

It is a matter of elementary calculus to check that this expression coincides with Eq. (D 6).

In (Talkner, Burada & Hänggi 2008, 2009) $L$ is given as $L(t) = \int_0^t ds f(s)e^{\beta s}/Z$, where $f = -X/\sqrt{Z}$. It is a matter of elementary calculus to check that this expression coincides with Eq. (D 8).

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probabilities $|a_{mn}^0|^2$ are obtained from the same expression as of $|a_{nm}|^2$, with the only difference that $L(t)$ is replaced by

$$L_0(t) = C^2(t)/2 + S^2(t)/2 \quad (D\, 14)$$

that is $|a_{mn}^0|^2 = f_{nm}[L_0(t)]$. Therefore the exclusive-work pdf $g_{0,t}$ is obtained from the dissipated-work pdf $g_{t}$ simply by replacing $L(t)$ with $L_0(t)$. It follows immediately that

$$\langle w_0 \rangle = \int dx \, x \, p_{t,0}^0(x) = L_0(t) \quad (D\, 15)$$

$$\langle \Delta w_0^2 \rangle = \int dx [x - L_0(t)]^2 \, p_{t,0}^0(x) = 2UL_0(t) \quad (D\, 16)$$

Note that, as expected, $\langle w_0 \rangle \geq 0$.

For the specific protocol

$$X(t) = 2\sin(t) \quad (D\, 17)$$

we find

$$L(t) - L_0(t) = t \sin(2t) \quad (D\, 18)$$

which is apparently different from zero except for integer multiples of $\pi/2$. Thus for any duration $t$ of the protocol $X(t)$ that is not an integer multiple of $\pi/2$, $L_0 \neq L_0$. Accordingly the first and second moments of $p_{t,0}^{\text{dis}}$ and $p_{t,0}^0$ differ, meaning that $w_{\text{dis}}$ and $w_0$ are distinct stochastic variables with different statistical properties.

It should be stressed that analogous results are found also for a classical driven harmonic oscillator. The statistics of dissipated-work and of exclusive-work generally differ, this fact holds true both quantum-mechanically and classically.

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