Fluctuation Theorems for Continuously Monitored Quantum Fluxes

Michele Campisi,* Peter Talkner,† and Peter Hänggi‡

Institute of Physics, University of Augsburg, Universitätsstrasse 1, D-86135 Augsburg, Germany

(Received 8 June 2010; revised manuscript received 17 July 2010; published 28 September 2010)

It is shown that quantum fluctuation theorems remain unaffected if measurements of any kind and number of observables are performed during the action of a force protocol. That is, although the backward and forward probabilities entering the fluctuation theorems are both altered by these measurements, their ratio remains unchanged. This observation allows us to describe the measurement of fluxes through interfaces and, in this way, to bridge the gap between the current theory, based on only two measurements performed at the beginning and end of the protocol, and experiments that are based on continuous monitoring.

DOI: 10.1103/PhysRevLett.105.140601 PACS numbers: 05.70.Ln, 03.65.Ta, 05.30.−d, 73.63.−b

In the last decade fluctuation theorems have experienced renewed and widespread interest [1–6]. These theorems yield rigorous predictions for nonequilibrium processes beyond linear response theory. In particular, they quantify the probability of events that are forbidden by the second law of thermodynamics as being exponentially suppressed compared to the probability of a typical, allowed event.

While experimental verifications of the fluctuation theorems for classical systems were performed by different groups and for different systems, e.g., [7–9], an experiment with quantum devices was performed only very recently [10]. In that experiment the flow of electrons through a double quantum dot placed between two leads with different chemical potentials was continuously monitored and the quantum fluctuation theorem was verified for the probability $p(q)$ that a number q of electrons is exchanged by the leads in a certain interval of time $\tau = t_f - t_0$; see Fig. 1. With regard to theory, however, the existing derivations of fluctuation theorems for quantum transport do not allow for a continuous monitoring of the flux of energy and/or particles. They are rather based on two projective quantum measurements of the relevant observables (Hamiltonian, number of particles), performed at t_0 and t_f, where the exchanged energy and number of particles are obtained as differences of the outcomes of these two measurements [5,11–13]. Evidently, this scheme presents practical difficulties, as it is not clear, for example, how one could measure the macroscopically large number of electrons in a lead with the required single electron resolution. Therefore, as other authors also have pointed out (see the conclusions in Ref. [12]), it is necessary to bridge the gap between theory and experiment by extending the theory in such a way as to account for the possibility of continuously monitoring a specific quantum observable of the system.

In this Letter we develop a multiple measurement approach to quantum fluctuation theorems that extends the previous two-measurement fluctuation theorems [5,11–13] allowing for the possibility to perform measurements of any observable within the interval (t_0, t_f). Most importantly, we demonstrate how to use this approach to overcome the major problem of measuring total energies and particle numbers of large reservoirs in case of a transport problem as the one described in Ref. [10]. The salient point here is to consider the *fluxes* through an interface rather than absolute numbers of particles and amounts of energy. This provides a coherent and general theoretical framework within which the recently reported electron transport experiment of Ref. [10] can be properly analyzed and understood, and new experiments can possibly be devised.

Multiple measurements scheme.—We consider a quantum system composed of subsystems, whose mutual interaction is turned on only within the finite time interval (t_0, t_f). They are initially disconnected and separately in equilibrium at different temperatures and chemical potentials. On coupling them through a time-dependent interaction leads to energy and particle transfers. While this exchange of energy and particles occurs a quantum observable is monitored, as in the experiment reported in [10]; see Fig. 1.

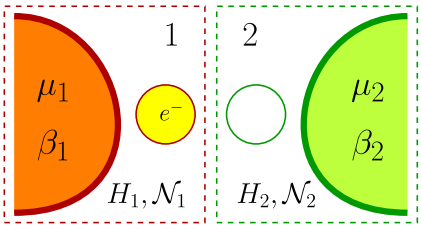

FIG. 1 (color online). Schematic representation of a bidirectional counting statistics experiment. Two leads (large semicircles) with different electronic chemical potentials ($\mu_1 \neq \mu_2$) and same inverse temperature ($\beta_1 = \beta_2$) are connected through a double quantum dot (small circles), whose quantum state is continuously monitored. The state (1,0), i.e., "one electron in the left dot, no electrons in the right dot" is depicted. The transition from this state to the state (0,1) signals the exchange of one electron from subsystem 1 to subsystem 2. $H_{1,2}$ and $\mathcal{N}_{1,2}$ denote the Hamiltonian and number of electrons operators of each subsystem, respectively.

We begin by considering only one intermediate measurement occurring at $t_1 \in (t_0, t_f)$. The total Hamiltonian $H(t)$ is: $H(t) = \sum_i H_i$ for $t \notin (t_0, t_f)$, and $H(t) = \sum_i H_i + V(t)$ for $t \in (t_0, t_f)$, where $V(t)$ is a time-dependent interaction term that couples the otherwise uncoupled subparts of the system within the time interval (t_0, t_f). Because of the time dependence of the Hamiltonian, the microreversibility principle does no longer apply in the way it does for autonomous systems. Under the assumption, however, that at each instant of time t the total Hamiltonian commutes with the antiunitary time-reversal operator Θ, $[H(t), \Theta] = 0$, a generalized form of microreversibility is obtained which jointly uses the operator Θ and the temporal inversion of the protocol $H(t)$ [12,14,15] (see the description of the reversed protocol below).

For $t \leq t_0$ each subsystem is assumed to be at equilibrium with an inverse temperature β_i and chemical potential μ_i of a particle species that may be exchanged between the subparts in presence of interaction. That is, at $t \leq t_0$ the density matrix is given by the direct product of two grand-canonical states: $\rho = \prod_i e^{-\beta_i[H_i - \mu_i \mathcal{N}_i - \Phi_i]}$, with Φ_i the grand-canonical free energy of subsystem i, and $\mathcal{N}_i$ the number operators of those particles that can be exchanged between the subsystems once the interaction is turned on. In absence of interaction the particle numbers in each subsystem are assumed to be conserved, i.e., $[H_i, \mathcal{N}_i] = 0$. In presence of interaction the total particle number $\mathcal{N} = \sum_i \mathcal{N}_i$ is still assumed to be conserved, i.e., $[H(t), \mathcal{N}] = 0$.

At time t_0 a joint quantum measurement of all H_i's and $\mathcal{N}_i$'s is performed. As a consequence the total system's wave function collapses onto a common eigenstate $|\psi_n\rangle$ of all these commuting observables, whose eigenvalues are given by: $H_i|\psi_n\rangle = E_n^i|\psi_n\rangle$ and $\mathcal{N}_i|\psi_n\rangle = N_n^i|\psi_n\rangle$. The system then evolves according to the unitary time evolution U_{t_1,t_0} governed by $H(t)$ until time t_1 when an observable A is measured. We assume for simplicity that the eigenvalues a_r of A are nondegenerate and that A commutes with the time-reversal operator Θ. As a consequence of this measurement, the system's wave function instantaneously collapses onto an eigenstate $|a_r\rangle$ of A. The evolution generated by $H(t)$ then continues until the time t_f, when a measurement of all H_i's and $\mathcal{N}_i$'s is again performed, collapsing the wave function to a joint eigenstate $|\psi_m\rangle$ of all H_i's and $\mathcal{N}_i$'s with eigenvalues E_m^i and N_m^i, respectively. Accordingly, the changes of energy $\Delta E_i = E_m^i - E_n^i$ and of particle numbers $\Delta N_i = N_m^i - N_n^i$ are recorded. During each realization of this protocol a three-state transition of the type $|\psi_n\rangle \to |a_r\rangle \to |\psi_m\rangle$ occurs with probability

$$P[n, r, m] = p(m, t_f|r, t_1)p(r, t_1|n, t_0)\rho_{nn}, \tag{1}$$

where

$$\rho_{nn} = \langle\psi_n|\rho|\psi_n\rangle = \prod_i e^{-\beta_i(E_n^i - \mu_i N_n^i - \Phi_i)} \tag{2}$$

is the probability to find the system in the state $|\psi_n\rangle$ at t_0, and $p(r, t_1|n, t_0)$ ($p(m, t_f|r, t_1)$) is the conditional probability to find the system in $|a_r\rangle$ at t_1 ($|\psi_m\rangle$ at t_f) provided that it was in $|\psi_n\rangle$ at t_0 ($|a_r\rangle$ at t_1), i.e.,

$$p(m, t_f|r, t_1) = |\langle\psi_m|U_{t_f,t_1}|a_r\rangle|^2, \tag{3}$$

$$p(r, t_1|n, t_0) = |\langle a_r|U_{t_1,t_0}|\psi_n\rangle|^2. \tag{4}$$

Accordingly, the joint forward (F) probability density function (pdf) of energy and particle number exchanges with one interruption at t_1 becomes:

$$P_F^{t_1}(\{\Delta E_i\}, \{\Delta N_i\}) = \sum_{m,n}\prod_i \delta(\Delta E_i - E_m^i + E_n^i) \times \delta(\Delta N_i - N_m^i + N_n^i)p^{t_1}(m, t_f|n, t_0)\rho_{nn}, \tag{5}$$

where

$$p^{t_1}(m, t_f|n, t_0) = \sum_r p(m, t_f|r, t_1)p(r, t_1|n, t_0) \tag{6}$$

denotes the conditional probability of finding the state $|\psi_m\rangle$ at t_f provided that the state $|\psi_n\rangle$ was found at t_0, and the observable A was measured at t_1. The sum in (6) runs over all eigenstates of A, and the superscript t_1 in Eqs. (5) and (6) indicates the measurement occurring at t_1.

We now consider the reversed protocol specified by preparing the system in the state ρ introduced above, changing the Hamiltonian in time according to the reversed schedule $\tilde{H}(t) = H(t_f + t_0 - t)$, and performing measurements of (i) all H_i's and $\mathcal{N}_i$'s, at time t_0, (ii) the observable A at $\tilde{t}_1 = t_0 + t_f - t_1$, (iii) all H_i's, $\mathcal{N}_i$'s, at time t_f. During each realization of this backward protocol a three-state transition of the type $\Theta|\psi_m\rangle \to \Theta|a_r\rangle \to \Theta|\psi_n\rangle$ is recorded with probability

$$\tilde{P}[m, r, n] = \tilde{p}(n, t_f|r, \tilde{t}_1)\tilde{p}(r, \tilde{t}_1|m, t_0)\rho_{mm}, \tag{7}$$

where $\tilde{p}(r, \tilde{t}_1|m, t_0) = |\langle a_r|\Theta^\dagger \tilde{U}_{\tilde{t}_1,t_0}\Theta|\psi_m\rangle|^2$, and $\tilde{p}(n, t_f|r, \tilde{t}_1) = |\langle\psi_n|\Theta^\dagger \tilde{U}_{t_f,\tilde{t}_1}\Theta|a_r\rangle|^2$, with $\tilde{U}_{t',t}$ the time evolution operator governed by $\tilde{H}(t)$. Thus the backward (B) pdf of energy and particle number exchanges, with an interruption at $\tilde{t}_1$ is

$$P_B^{\tilde{t}_1}(\{\Delta E_i\}, \{\Delta N_i\}) = \sum_{n,m}\prod_i \delta(\Delta E_i - E_n^i + E_m^i) \times \delta(\Delta N_i - N_n^i + N_m^i)\tilde{p}^{\tilde{t}_1}(n, t_f|m, t_0)\rho_{mm}, \tag{8}$$

where

$$\tilde{p}^{\,\tilde{t}_1}(n, t_f|m, t_0) = \sum_r \tilde{p}(n, t_f|r, \tilde{t}_1)\tilde{p}(r, \tilde{t}_1|m, t_0) \tag{9}$$

is the conditional probability to find the system in the state $\Theta|\psi_n\rangle$ at time t_f provided that it was in $\Theta|\psi_m\rangle$ at t_0 and the observable A was measured at $\tilde{t}_1$.

Using $[H(t), \Theta] = 0$, and expressing the time evolution operator as a time ordered product one finds, in a similar way as in [15], that $\Theta^\dagger \tilde{U}_{\tilde{t}_1,t_0}\Theta = U_{t_1,t_f}$, and $\Theta^\dagger \tilde{U}_{t_f,\tilde{t}_1}\Theta = U_{t_0,t_1}$. Thus $\tilde{p}(n, t_f|r, \tilde{t}_1) = p(r, t_1|n, t_0)$, $\tilde{p}(r, \tilde{t}_1|m, t_0) = p(m, t_f|r, t_1)$, and, consequently,

$$p^{t_1}(m, t_f|n, t_0) = \tilde{p}^{\tilde{t}_1}(n, t_f|m, t_0) \tag{10}$$

follows. Combining Eqs. (2), (5), (8), and (10) we obtain

$$\frac{P_F^{t_1}(\{\Delta E_i\}, \{\Delta N_i\})}{P_B^{\tilde{t}_1}(\{-\Delta E_i\}, \{-\Delta N_i\})} = \prod_i e^{\beta_i(\Delta E_i - \mu_i \Delta N_i)}, \tag{11}$$

which reads exactly as the two-measurements fluctuation theorem [12], with the major difference that now a third projective measurement is performed at t_1. Here the free energy difference does not appear in Eq. (11) because we assumed $H(t_0) = H(t_f)$. We stress that the backward and forward probabilities with intermediate measurement $P_F^{t_1}$, $P_B^{\tilde{t}_1}$, in general differ from the corresponding probabilities without intermediate measurement P_F, P_B of Ref. [12]. Remarkably, however, their ratio remains unaltered $P_F^{t_1}/P_B^{\tilde{t}_1} = P_F/P_B$. In both cases though, particle numbers and energy measurements are assumed to be performed at t_0 and t_f. As discussed in the introduction, these measurements are practically impossible when the subsystems are macroscopic reservoirs, as is the case with quantum transport problems. In the following we will show a way to get around this problem by taking advantage of the possibility opened by Eq. (11) of performing intermediate measurements.

The salient point in the derivation of Eq. (11) is that the generalized time-reversal invariance property of the transition probability without interruptions, i.e., $p(m, t_f|n, t_0) = \tilde{p}(n, t_f|m, t_0)$ [12], continues to hold, Eq. (10), even if a projective measurement is performed at t_1. Evidently the result (11) can be extended to the case of multiple intermediate quantum measurements of possibly distinct observables A_k, $k = 1 \ldots K$ occurring at times t_k during the forward protocol, and at times $\tilde{t}_k = t_0 + t_f - t_k$ during the backward protocol. Thus we conclude that the fluctuation theorem is unaffected by the action of intermediate projective quantum measurements.

Indeed a more detailed result holds. Consider the joint probability for a sequence of outcomes $n_0, \vec{n}_k, n_f = n_0, n_1 \ldots n_n, n_f$, and the joint probability of the reversed sequence $n_f, \tilde{n}_k, n_0 = n_f, n_n \ldots n_1, n_0$ for the backward protocol

$$P[n_0, \vec{n}_k, n_f] = \left[\prod_{k=0}^{K} p(n_{k+1}, t_{k+1}|n_k, t_k)\right]\rho_{n_0 n_0}, \tag{12}$$

$$\tilde{P}[n_f, \tilde{n}_k, n_0] = \left[\prod_{k=0}^{K} \tilde{p}(n_k, \tilde{t}_k|n_{k+1}, \tilde{t}_{k+1})\right]\rho_{n_f n_f}, \tag{13}$$

where $\tilde{t}_{f,0} = t_{0,f}$, and n_0, n_f label common eigenstates of $\{H_1, H_2, \mathcal{N}_1, \mathcal{N}_2\}$, and n_k labels eigenstates of A_k. Using $[H(t), \Theta] = 0$ one finds $\Theta \tilde{U}_{\tilde{t}_{k+1}, \tilde{t}_k} \Theta^\dagger = U_{t_{k+1}, t_k}$, which leads to $\tilde{p}(n_k, \tilde{t}_k|n_{k+1}, \tilde{t}_{k+1}) = p(n_{k+1}, t_{k+1}|n_k, t_k)$. Then, using Eqs. (2), (12), and (13), we readily find the following detailed fluctuation theorem

$$\frac{P[n, \vec{n}_k, m]}{\tilde{P}[m, \tilde{n}_k, n]} = \prod_i e^{\beta_i(E_m^i - E_n^i - \mu_i N_m^i + \mu_i N_n^i)}. \tag{14}$$

Notably, the right-hand side depends only on initial and final eigenvalues of H_i, $\mathcal{N}_i$, but not on the eigenvalues of A_k. Equation (14) can be regarded as the quantum mechanical version of a formula that was first put forward by Bochkov and Kuzovlev for classical trajectories [see Eq. (7) of Ref. [1]; a related result was derived recently for a classical master equation in [16]].

Application to counting statistics experiments.—In the counting statistics experiment of Ref. [10] two leads having the same inverse temperature β are connected via a double quantum dot. We identify the left reservoir plus left dot as subsystem 1, and the right reservoir plus right dot as subsystem 2; see Fig. 1. The two subsystems are coupled through a small (but important) time-independent interaction term V. At time t_0 an electric potential difference $\Delta\varphi$ is applied across the two subsystems. We model this by saying that immediately after the time t_0 the density matrix is the factorized grand-canonical state ρ specified above with $e\Delta\varphi = \mu_1 - \mu_2$. Afterwards the system follows the evolution according to $H = H_1 + H_2 + V$. This is equivalent to assuming that V is switched on instantaneously at t_0. Since V is small the energy of the total system made of the leads and the double quantum dot hardly changes, hence, $\Delta E_1 \simeq -\Delta E_2$. Because of the conservation of the total number of electrons, we have $\Delta N_1 = -\Delta N_2 \doteq q$. A time-reversal invariant observable A, detecting whether there is an excess electron in either of the two dots is continuously monitored, resulting in a signal $\vec{n}_k$. The total number of exchanged particles is calculated from $\vec{n}_k$ by means of a function $q[\vec{n}_k]$ that determines the number of transitions from the state (1,0) (i.e., one electron in the left dot and no electrons in the right dot) to the state (0,1) (i.e., no electrons in the left dot and one electron in the right dot), minus the number of opposite transitions. Evidently, $q[\vec{n}_k] = -q[\tilde{n}_k]$ holds. The only difference with the multiple measurement scheme presented above is that in the experiment no measurements of total particle numbers in the two subsystems are performed at t_0 and t_f. However, since the initial density matrix is a mixture of states with definite particle numbers in each subsystems, a measurement at t_0 can be abandoned if one is only interested in relative changes of particle numbers. By means of registration of each transition between the two subsystems, not only the flux of electrons is monitored but it is also guaranteed that during the whole process the particle numbers of each subsystem have definite values. Therefore, also at t_f, the state of the total system is a mixture of states with definite particle numbers; hence, a measurement of particle numbers need not be performed at t_f. Thus $q[\vec{n}_k]$ coincides with the value that would have been obtained from electron number measurements, i.e., $q[\vec{n}_k] = N_m^1 - N_n^1 = -N_m^2 + N_n^2$. Therefore the pdf of particle exchange is

$$P_F^{\{t_k\}}(q) = \sum_{\vec{n}_k} \delta(q - q[\vec{n}_k]) P[\vec{n}_k], \tag{15}$$

where we introduced the marginal probability $P[\vec{n}_k] = \sum_{mn} P[n, \vec{n}_k, m]$. Since the backward and forward protocols coincide in this case, we have $P_F^{\{t_k\}} = P_B^{\{\tilde{t}_k\}}$. Using Eqs. (14) and (15) with $q[\vec{n}_k] = -q[\tilde{n}_k]$, we conclude that the pdf of electron exchange indeed satisfies

$$P_F^{\{t_k\}}(q) = P_F^{\{t_k\}}(-q) e^{\beta(\mu_1 - \mu_2)q} \tag{16}$$

in good qualitative agreement for small τ, and excellent quantitative agreement for large τ, with experimental findings displayed in Fig. 2a of Ref. [10], [17]. The same experimental setup could be used to confirm the validity of the detailed fluctuation relation

$$P[\vec{n}_k] = P[\tilde{n}_k] e^{\beta(\mu_1 - \mu_2) q[\vec{n}_k]} \tag{17}$$

that follows from (14).

Remarks.— One assumption of our theory is that the initial state of the system is the direct product of two grand-canonical states. For this particular choice of the initial state the fluctuation theorems (16) and (17) do hold independently of the duration τ of the protocol. In experiments, however, unavoidable interactions may lead to correlations between the subparts of the system already before the protocol has started. These initial correlations in general will lead to transient deviations from the fluctuation theorems which then are expected to hold as steady state fluctuation theorems in the limit of large τ, as is the case with experiment [10].

The assumptions that the measured observables A_k have nondegenerate eigenvalues can be abandoned without any change of our central result formulated in Eqs. (11) and (14). We further found that our main result remains true if the quantum measurements are performed with respect to positive operator valued measures (POVM's) instead of von Neumann projection valued measures. This is not so surprising after all, being the former less invasive than the latter. The main result holds for any number of subsystems. In particular for a single closed system it generalizes the Tasaki-Crooks work fluctuation theorem [18–20]. In the case of two susbsystems with equal temperature and no matter exchange it generalizes the Tasaki-Crooks fluctuation theorem for open systems [21,22]. Multiple measurements of reservoir energies (not fluxes) were considered for this case in Ref. [23].

Far from being a problem of purely academic interest, the multiple measurement scheme pursued here, helps bridging the existing gap between theory and experiments concerning quantum fluctuation theorems. The measurement of energy and particle content of the subparts of a system may be practically impossible, especially when the subparts are macroscopic objects like the leads in an electron counting statistics experiment. On the other hand, energy and matter exchanges may be measured by monitoring the respective fluxes through interfaces, making the cumbersome measurements of total energies and particle numbers obsolete. Here we have shown that the fluctuation theorem continues to hold in this case of continuous monitoring, Eqs. (11) and (14). We further provided a theoretical explanation for an experimentally observed relation, Eq. (16), and predicted that a more detailed relation, Eq. (17), should hold in the same setup.

This work was supported by the cluster of excellence Nanosystems Initiative Munich (NIM) and the Volkswagen Foundation (project I/83902).

*michele.campisi@physik.uni-augsburg.de
†peter.talkner@physik.uni-augsburg.de
‡hanggi@physik.uni-augsburg.de

[1] G. N. Bochkov and Y. E. Kuzovlev, Zh. Eksp. Teor. Fiz. **72**, 238 (1977) [JETP **45**, 125 (1977)].
[2] D. J. Evans, E. G. D. Cohen, and G. P. Morriss, Phys. Rev. Lett. **71**, 2401 (1993).
[3] G. Gallavotti and E. G. D. Cohen, Phys. Rev. Lett. **74**, 2694 (1995).
[4] G. E. Crooks, Phys. Rev. E **60**, 2721 (1999).
[5] M. Esposito, U. Harbola, and S. Mukamel, Rev. Mod. Phys. **81**, 1665 (2009).
[6] J. Ren, P. Hänggi, and B. Li, Phys. Rev. Lett. **104**, 170601 (2010).
[7] D. Collin *et al.*, Nature (London) **437**, 231 (2005).
[8] J. Liphardt *et al.*, Science **296**, 1832 (2002).
[9] F. Douarche *et al.*, Europhys. Lett. **70**, 593 (2005).
[10] Y. Utsumi *et al.*, Phys. Rev. B **81**, 125331 (2010).
[11] C. Jarzynski and D. K. Wójcik, Phys. Rev. Lett. **92**, 230602 (2004).
[12] D. Andrieux *et al.*, New J. Phys. **11**, 043014 (2009).
[13] K. Saito and Y. Utsumi, Phys. Rev. B **78**, 115429 (2008).
[14] D. Andrieux and P. Gaspard, Phys. Rev. Lett. **100**, 230404 (2008).
[15] M. Campisi, P. Talkner, and P. Hänggi, arXiv:1003.1052.
[16] A. Altland *et al.*, arXiv:1005.4662.
[17] In Ref. [10] the measurement apparatus is not only responsible for wave function collapses but also exchanges electrons. According to [10], this exchange of electrons, that was not considered here, leads to a renormalization of the temperature in (16).
[18] H. Tasaki, arXiv:cond-mat/0009244.
[19] P. Talkner, E. Lutz, and P. Hänggi, Phys. Rev. E **75**, 050102(R) (2007).
[20] P. Talkner and P. Hänggi, J. Phys. A **40**, F569 (2007).
[21] M. Campisi, P. Talkner, and P. Hänggi, Phys. Rev. Lett. **102**, 210401 (2009).
[22] P. Talkner, M. Campisi, and P. Hänggi, J. Stat. Mech. (2009) P02025.
[23] W. De Roeck and C. Maes, Phys. Rev. E **69**, 026115 (2004).