Perturbative analysis of quantum fluctuation theorems in a driven open system

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We give a perturbative analysis of Crooks relation and Jarzynski equality in an arbitrary driven quantum system weakly coupled to a heat bath. Invoking no efficient Hamiltonian nor any restriction on the form of the coupling, we derive the first-order correction to Crooks relation and Jarzynski equality in terms of the interacting Hamiltonian. A Crooks type of relation about energy increment in the system and nonvanishing heat expenditure to the heat bath up to the second order of interaction would be given by the way. Our results tell us that deviation of the quantum fluctuation relations is mainly caused by energy trapped interaction rather than the heat flow to (or from) heat bath. We also show a way of defining work in weak coupling regime which is consistent to the work definition in closed systems and thus deepen our understanding of work in quantum realm.

Introduction.—The great success of equilibrium statistical mechanics has both deepened our understanding of nature and helped us build modern technical architecture. Though, nonequilibrium phenomenon has caught attention of physicists for a long time. Important results in this direction such as linear response theory were already given in near-equilibrium regime about half a century ago [1]. With the thriving of nanotechnology in which fluctuations are significant, development of nonequilibrium statistics has become more urgent. It stimulated a lot of investigation of statistical properties in systems arbitrarily out of equilibrium beyond linear response theory. Fluctuation theorems are among the most remarkable achievements in the field of nonequilibrium statistical mechanics [2–6]. Among them, Crooks relation and Jarzynski equality are of particular importance. Both theorems were first derived in a classical system weakly coupled to a heat bath of constant temperature $T$ [3, 4]. The former establishes a connection between a transition process and its reverse

$$p[W; \lambda]/p[-W; \tilde{\lambda}] = e^{\beta (W - \Delta F_S)}.$$  (1)

$p[W; \lambda]$ is the probability that $W$ work would be done on the system $S$ which is driven out of equilibrium through an arbitrary protocol $\lambda$ while $p[-W; \tilde{\lambda}]$ probability of extracting that much of work from $S$ during the mirrored reverse process. $\beta := 1/T$ denotes the inverse temperature and $\Delta F_S$ the change of Helmholtz free energy of system $S$. Notice that we adopt the convention of setting Boltzmann constant to one. From Crooks relation (1), one can derive Jarzynski equality directly by rearranging (1) and integrating over $W$

$$\langle e^{-\beta W}\rangle_\lambda = e^{-\beta \Delta F_S}.$$  (2)

$\langle \cdot \rangle_\lambda$ means averaging over statistical realizations of the forward process characterized by protocol $\lambda$. Many attempts were made to extent them to the quantum regime since then [5–19]. The first task to achieve this goal is defining work in the framework of quantum mechanics which is quite delicate and has caused much controversy [20–22]. Among all these definition strategies, the two-point projective energy measure (TPEM) protocol has been the most acceptable [5–8, 11–19]. By using TPEM, quantum fluctuation theorems were first obtained theoretically [7, 8, 10, 11] and verified experimentally [23, 24] for closed system under unitary time evolution. Corresponding theoretical investigation for Markovian processes [12–16] were also developed. In the case where interaction with heat bath is small and has vanishing expectation value in the eigenbasis of total Hamiltonians at both ends of the transition, quantum fluctuation theorems were also obtained [17]. This covers many important physical models such as the Jaynes-Cummings model [25] and the Rabi model [26, 27] in the weak coupling regime. For interaction of other forms such as some spin-spin coupling [28, 29] or that of sufficient magnitude, efficient Hamiltonian of the system, consisting of free Hamiltonian of the system and an additional term resulting from interaction with heat bath, has to be employed to obtain Crooks relation and Jarzynski equality [17–19].

In this Letter, we consider also a quantum system weakly coupled to a heat bath but with no further restriction on the form of interaction. The system could be driven far away from equilibrium. We invoke no efficient Hamiltonian to incorporate the coupling to the heat bath and abandon the attempt of showing a formal consistency with the exact fluctuation relations as obtained in closed system and some specialized open models. By invoking TPEM and proper definition of work, we examine Crooks (1) and Jarzynski (2) up to the first order of interaction. The work definition invoked in the weak coupling case can be proven to be consistent with that employed in a closed system [5–8, 11]. We will show with a specific numerical example that without the efficient Hamiltonian, both relations would break down and our correction would be effective to some extend even when
the interaction is comparable to the free Hamiltonian of system S. The correction turns out to be the change of average coupling Hamiltonian. An energy fluctuation relation (10) of Crooks type concerning energy increment of system S and its nonvanishing heat expenditure to heat bath would be obtained up to the second order of interaction between system S and heat bath B. In the weak coupling regime, energy trapped in interaction causes the deviation of Jarzynski equality and Crook relation while impact of heat exchange with environment is minimal.

**System setup.**—Consider the composite of system S and heat bath B

\[ H_0 = H_S(\lambda_t) + H_B + H_I, \]  

(3)

where coupling term \( H_I \) is a very small compared with \( H_S(\lambda_t) + H_B \). \( \lambda \) represents a control protocol of a (set of) parameter(s) of S with \( \lambda_t \) being its realization at time \( t \). Thus, system Hamiltonian \( H_S(\lambda_t) \) is time-dependent while heat bath Hamiltonian \( H_B \) and \( H_I \) are constant. The composite of S and B is initially a thermal state of temperature \( T \) in both forward transition and its reverse

\[ \rho(\lambda_0,\tau) := e^{-\beta H(\lambda_0,\tau)} / Z(\lambda_0,\tau), \]  

(4)

Time duration of both the forward and backward transition is \( \tau \). \( Z(\lambda_0,\tau) := \text{tr} e^{-\beta H(\lambda_0,\tau)} \) are partition functions corresponding to parameter settings \( \lambda_0 \) and \( \lambda_\tau \) respectively.

**Measurements and driving protocol.**—System S and bath B start with an energy measurement which is the first part of TPEM at the beginning of the forward transition

\[ \rho(\lambda_0) \approx \hat{\rho}(\lambda_0) \left( 1 - \beta \Delta H_1^{\lambda_0} \right), \]  

(5)

\[ p[E,Q;\lambda] = \sum_{n,k,m,\ell} \delta \left( E - E_m^{\lambda_\tau} + E_k^{\lambda_\tau} \right) \delta \left( Q - E_k^{\lambda_\tau} + E_m^{\lambda_\tau} \right) p[m\ell,nk;\lambda]. \]  

(7)

Probability \( p[m\ell,nk;\lambda] \) of obtaining \( m \) and \( \ell \) in the first projective energy measurement while \( n \) and \( k \) in the second, should be \( \text{tr} (\Pi_m^{\lambda_\tau} \otimes \Pi_k^{\lambda_\tau}) U_{\tau,0} [\lambda] (\Pi_m^{\lambda_0} \otimes \Pi_k^{\lambda_0}) \hat{\rho}(\lambda_0) U^\dagger_{\tau,0} [\lambda] \). Probability distribution \( p[-E,-Q;\lambda] \) of system energy

\[ p[m\ell,nk;\lambda] \approx \text{tr} \left( (\Pi_m^{\lambda_\tau} \otimes \Pi_k^{\lambda_\tau}) V[\lambda] (\Pi_m^{\lambda_0} \otimes \Pi_k^{\lambda_0}) V^\dagger[\lambda] \right) e^{-\beta(E_m^{\lambda_0} + E_k^{\lambda_0})} / Z_B Z_S(\lambda_0). \]  

(8)

“\( \approx \)” means “equal up to the second order of the interaction Hamiltonian \( H_1 \)”.

Decrement and heat absorption during the backward process would be of similar form. We can show that probability \( p[m\ell,nk;\lambda] \) of non-zero heat transfer namely \( k \neq \ell \) generally are of the second order of coupling Hamiltonian \( H_1 \)

\[ p[m\ell,nk;\lambda] \approx \text{tr} \left[ (\Pi_m^{\lambda_\tau} \otimes \Pi_k^{\lambda_\tau}) V[\lambda] (\Pi_m^{\lambda_0} \otimes \Pi_k^{\lambda_0}) V^\dagger[\lambda] \right] e^{-\beta(E_m^{\lambda_0} + E_k^{\lambda_0})} / Z_B Z_S(\lambda_0). \]  

(8)
rived for probability $p[nk, m\ell; \tilde{\lambda}]$ in backward process. A direct consequence of the microscopic time reversibility $U^{\dagger}_{\tau,0}[\tilde{\lambda}] = U_{\tau,0}[\lambda]$ and $U^{\dagger}_{\tau,0}[\tilde{\lambda}] = U_{\tau,0}[\lambda]$ of the composite system for both coupled and decoupled cases, is $V[\tilde{\lambda}] = V[\lambda]$. We can further use this to derive a close relation of the former pair of probabilities

$$p[m\ell, nk; \lambda] \equiv p[nk, m\ell; \tilde{\lambda}] e^{\beta(E_{m\ell}^{\lambda^*} - E_{nk}^{\lambda} + E_{m\ell}^{\lambda} - E_{nk}^{\lambda^*} - \Delta F_{3})},$$  \hspace{1cm} (9)

where $\Delta F_{3} := F_{S}(\lambda_{\tau}) - F_{S}(\lambda_{0})$ is Helmholtz free energy difference of system $S$ with parameter settings at the two ends of the forward process $F_{S}(\lambda_{0,\tau}) := -T \ln Z_{S}(\lambda_{0,\tau})$. This relation holds for any nonvanishing heat transfer where $k \neq \ell$. By substituting (9) to the expression (7), we can derive a fluctuation relation of Crooks form for energy increment $W$ and non-zero heat transfer $Q \neq 0$

$$p[E, Q; \lambda]/p[-E, -Q; \tilde{\lambda}] \approx e^{\beta(E + Q - \Delta F_{3})}. \hspace{1cm} (10)$$

Correction to Crooks relation and Jarzynski equality.— Consider the case where eigency spectrum of system $S$ is non-degenerate, or we can employ measurements of complete sets of mutually compatible observables on $S$ which including system Hamiltonian $H_{S}(\lambda_{0,\tau})$ in TPE. We can then find eigenket $|\psi_{m}^{\lambda^*}\rangle$ of the set of complete set of mutually compatible observables containing $H_{S}(\lambda_{0,\tau})$ such that $H_{S}(\lambda_{0,\tau})|\psi_{m}^{\lambda^*}\rangle = E_{m}^{\lambda^*}|\psi_{m}^{\lambda^*}\rangle$, with other degrees of freedom within the degeneracies of the energy levels incorporated to notation $m$. Given vanishing coupling, transition probability from $|\psi_{m}^{\lambda}\rangle$ to $|\psi_{m}^{\lambda^*}\rangle$ would be $p[n|m; \lambda] := |\langle \psi_{n}^{\lambda}|U_{S,\tau,0}[\lambda]|\psi_{m}^{\lambda^*}\rangle|^{2}$ in the forward driving $\lambda$. Plugging $p[n|m; \lambda]$, (5) and (6) onto (7) and invoking microscopic time reversibility $U^{\dagger}_{\tau,0}[\tilde{\lambda}] = U_{\tau,0}[\lambda]$ of the composite system, we can show that $p[E, Q; \lambda]$ and $p[-E, -Q; \tilde{\lambda}]$ are mutually related up to the first order of interaction

$$W = E_{m}^{\lambda^*} - E_{n}^{\lambda^*} + E_{m}^{\lambda} - E_{n}^{\lambda} + \Delta E_{nk, m\ell}. \hspace{1cm} (12)$$

$$p[W; \lambda] = \int dE dQ \sum_{m,n,k,\ell} \delta \left(W - E - Q - \Delta E_{nk, m\ell}\right) \delta \left(E_{m}^{\lambda^*} - E_{n}^{\lambda^*} + E_{m}^{\lambda} - E_{n}^{\lambda} \right) \delta \left(E - E_{m}^{\lambda^*} + E_{n}^{\lambda^*} - E_{m}^{\lambda} + E_{n}^{\lambda} \right) p[m\ell, nk; \lambda].$$

From energy fluctuation relation (11), one can derive a similar relation between probability distributions of work injection $p[W; \lambda]$ in forward process and extraction $p[-W; \tilde{\lambda}]$ in its reverse up to the first order of interaction

$$Z_{S}(\lambda_{0})p[W; \lambda] - e^{\beta W} Z_{S}(\lambda_{\tau})p[-W; \tilde{\lambda}] \approx \beta \left(Z_{S}(\lambda_{0})p[W; \lambda] \langle H_{1} \rangle^{\lambda_{0}} - e^{\beta W} Z_{S}(\lambda_{\tau})p[-W; \tilde{\lambda}] \langle H_{1} \rangle^{\lambda_{\tau}} \right). \hspace{1cm} (13)$$

We can obtain the first-order correction to Crooks relation from (13)

$$p[W; \lambda]/p[-W; \tilde{\lambda}] \approx e^{\beta(W - \Delta F_{3} - \Delta E_{1})}. \hspace{1cm} (14)$$
\[ \Delta E_1 := \langle H_1 \rangle_{\lambda, \tau} - \langle H_1 \rangle_{\lambda_0} \] is the difference of interaction energy of the compound system of S and B, given different parameter settings \( \lambda_0, \tau \) of S. By appropriately rearrangement of (14) and integration over \( W \), one can derive the first-order correction of Jarzynski equality

\[ \langle e^{-\beta W} \rangle_\lambda \simeq e^{-\beta(\Delta F_S + \Delta E_1)}. \] (15)

**Consistency between work definitions in the weak coupling and closed cases.** If one consider the complex of system S and heat bath B as a whole, it is closed. Its partition functions corresponding to the two configurations \( \lambda_0, \tau \) of subsystem S should be \( Z(\lambda_0, \tau) \approx Z_S(\lambda_0, \tau) Z_B(1 - \beta \langle H_1 \rangle_{\lambda_0, \tau}) \). Therefore, the change of Helmholtz free energy of the total system should be

\[ \Delta F \simeq \Delta F_S + \Delta E_1. \] (16)

Hence corrected Crooks relation (14) of subsystem S would coincide with the Crooks relation for the total system where TPEM is implemented for total Hamiltonian \( \mathbf{H}(\lambda_0, \tau) \) instead of sum of free Hamiltonians of subsystems \( \mathbf{H}_S(\lambda_0, \tau) + \mathbf{H}_B \) up the first order of \( \mathbf{H}_1 \). Analogical inference can be made to corrected Jarzynski equality (15) of S. Therefore, the work definition (12) namely the first law we employed would coincide with those employed in closed systems [7, 8, 10, 11] in weak coupling regime.

**Specific model exemplification.**—By employing a two-level system (TLS) coupled to an open-bounded regime.

\[ \langle e^{-\beta W} \rangle_\lambda \simeq e^{-\beta(\Delta F_S + \Delta E_1)}. \] (15)

**FIG. 1.** (Colored online.) \( W_0 \) with \( |D_0(W_0, \omega_c) - W_0| \geq 0.01 \) are picked out as listed on the insets of both (a). The lines in (b) and (c) correspond to the same \( W_0 \) as the lines in (a) of the same color and line type as listed on the inset on the left hand. (c) tells us that probability of nonvanishing heat transferation does not vanishing in terms of the second order of coupling though its zeroth and first order do.

\[ D_0(W_0, \omega_c) = \frac{\int_{Q \neq 0} dQp[W_0 - Q, Q; \lambda]}{\int_{Q \neq 0} dQp[-W_0, Q, -Q; \lambda]} \approx e^{\beta(W_0 - \Delta F_S)} \] (17)

which is a direct consequence of (10). For further simplification, we conduct coarse-grained division of the domain of \( W_0 \) by pieces of width \( \delta W_0 = 0.13 \text{ GHz} \). For every \( \omega_c \) between 0 and \( \omega_c \), we numerically evaluate the difference function

\[ D_0(W_0, \omega_c) := \int_{Q \neq 0} dQp[-W_0, Q, -Q, \lambda] e^{\beta(W_0 - \Delta F_S)} - p[W_0 - Q, Q, \lambda] \] which would vanish up to the second order of small coupling strength \( \omega_{\text{proc}} \) according to (17).

Our numerical simulation tells us that both \( D(W_0, \omega_c) \) (c.f. Fig. 1(a)) and its derivative (c.f. Fig. 1(b)) vanishes in the vicinity of vanishing coupling. Thus give strong numerical evidence for (17) and (10) being valid in the second order of the coupling Hamiltonian. One can infer from Fig. 1(c) that (17) and (10) are nontrivial in general. We remark that driving protocols \( \lambda \) and \( \lambda \) contain information of \( \omega_{\text{proc}} \).

Similarly, we use coarse-grained division of the domain of work \( W \) by pieces of \( \delta W = 0.34 \text{ GHz} \) in the numerical demonstration of the first-order correction of (14) to Crooks relation. We evaluate the difference function

\[ D(W, \omega_c) := p[-W, \lambda] e^{-\beta W} - p[W, \lambda] \] which would vanish in the first order of \( \omega_c \) according to (14). The simulation results shown in Fig. 2(a) confirms us the validity the
correction (14). From Fig. 2(b), we can tell that in strong coupling regime $\omega_c \geq 0.2$ GHz the probability of encountering significant deviation $|D(W, \omega_c = \omega)| \geq 4$ is very thin ($\sim 10^{-7}$). Numerical results of first-order correction (15) to Jarzynski equality are shown in Fig. 2(c). It tells us that our first-order correction (15) are not only effective near vanishing interaction regime, but can also play a significant role in strong coupling circumstances where $\omega_c \geq 0.2$ GHz is comparable to 1 GHz namely to $\omega$. Significance of the correction $e^{-\beta \Delta E f}$ to Jarzynski equality in strong coupling regime can be prominent at least in the specific model we studied numerically here. This agrees with the small probability of significant deviation from (14) as shown in Fig. 2(b).

**Discussion and outlook.**—The perturbative analysis of Crooks relation and Jarzynski equality in the limit of weak coupling shade some light on the study of fluctuation theories in an open system from a new angle. Our results shows that energy trapped in interaction between system and environment is a prominent cause of deviation of fluctuation relations from those established in closed systems as has been shown in (14) and (15). Contractively, heat transfered to (or from) environment seems a much less important cause of such a deviation according to (10). Our numerical results provide a possibility that correction with the interactive energy integrated can play a prominent role even in strong coupling regime as in the case we reported in this Letter. It is an interesting question whether in strong coupling regime such a correction would be prominent in general? If not, to what condition, would the effect of this correction extent to the strong coupling regime to a considerable degree? Further, our analysis (16) indicate that (12) should be a good definition of work in open system weakly coupled to a heat bath. It is consistent with the work definition in a closed system. The result refreshes our understanding of work in quantum regime.

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[1] R. Kubo, J. Phys. Soc. Jpn. **12**, 570 (1957).

[2] G. N. Bochkov and Y. E. Kuzolev, Sov. Phys. JETP **45**, 125 (1977).

[3] C. Jarzynski, Phys. Rev. Lett. **78**, 2690 (1997).

[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] M. Campisi, P. Hänggi, and P. Talkner, Rev. Mod. Phys. **83**, 771 (2011).

[7] H. Tasaki, arXiv:0009244.

[8] J. Kurchan, arXiv:0007360.

[9] S. Yukawa, J. Phys. Soc. Jpn. **69**, 2367 (2000).

[10] P. Talkner and P. Hänggi, J. Phys. A Math. Theor. **40**, 569 (2007).

[11] F. Liu and Z.-C. Ouyang, Chin. Phys. B **23**, 070512 (2014).

[12] S. Mukamel, Phys. Rev. Lett. **90**, 170604 (2003).

[13] W. De Roeck and C. Maes, Phys. Rev. E **69**, 026115 (2004).

[14] M. Esposito and S. Mukamel, Phys. Rev. E **73**, 046129 (2006).

[15] R. Chetrite and K. Mallick, J. Stat. Phys. **148**, 480 (2012).

[16] M. Mehboudi, A. Sanpera, and J. M. R. Parrondo, arXiv:1705.03968.

[17] P. Talkner, M. Campisi, and P. Hänggi, J. Stat. Mech. **2009**, P02025 (2009).

[18] M. Campisi, P. Talkner, and P. Hänggi, Phys. Rev. Lett. **102**, 210401 (2009).

[19] M. Campisi, P. Talkner, and P. Hänggi, J. Phys. A Math. Theor. **42**, 392002 (2009).

[20] P. Talkner, E. Lutz, and P. Hänggi, Phys. Rev. E **75**, 050102 (2007).

[21] A. E. Allahverdyan, Phys. Rev. E **90**, 032137 (2014).

[22] C. Jarzynski, H. T. Quan, and S. Rahav, Phys. Rev. X **5**, 031038 (2015).
[23] S. An, J.-N. Zhang, M. Um, D. Lv, Y. Lu, J. Zhang, Z.-Q. Yin, H. T. Quan, and K. Kim, Nat. Phys. 11, 193 (2014).

[24] T. B. Batalhão, A. M. Souza, L. Mazzola, R. Auccaise, R. S. Sarthour, I. S. Oliveira, J. Goold, G. De Chiara, M. Paternostro, and R. M. Serra, Phys. Rev. Lett. 113, 140601 (2014).

[25] B. W. Shore and P. L. Knight, J. Mod. Opt. 40, 1195 (1993).

[26] D. Braak, Phys. Rev. Lett. 107, 100401 (2011).

[27] Q.-T. Xie, S. Cui, J.-P. Cao, L. Amico, and H. Fan, Phys. Rev. X 4, 021046 (2014).

[28] P. Pfeuty, Ann. Phys. 57, 79 (1970).

[29] H. T. Quan, Z. Song, X. F. Liu, P. Zanardi, and C. P. Sun, Phys. Rev. Lett. 96, 140604 (2006).