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# Unified treatment of the quantum fluctuation theorem and the Jarzynski equality in terms of microscopic reversibility

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There are two related theorems which hold even in far from equilibrium, namely fluctuation theorem and Jarzynski equality. Fluctuation theorem states the existence of symmetry of fluctuation of entropy production, while the Jarzynski equality enables us to estimate the free energy change between two states by using irreversible processes. On the other hand, the relationship between these theorems was investigated by Crooks [Phys. Rev. E **60**, 2721 (1999)] for the classical stochastic systems. In this paper, we derive quantum analogues of fluctuation theorem and Jarzynski equality in terms of microscopic reversibility. In other words, the quantum analog of the work by Crooks is presented. Also, for the quasiclassical Langevin system, microscopically reversible condition is confirmed.

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## I. INTRODUCTION

Since the discovery by Evans *et al.* [1], many types of fluctuation theorem (FT) has been presented both for deterministic [1–3] and stochastic systems [4–10]. Experimental verification of FT was also performed [11] Though the systems, the definition of entropy production, and interpretations (such as distinction of transient or steady state) are different, FT has the following universal form:

$$\frac{P(\Delta S=A)}{P(\Delta S=-A)} \simeq e^{A/k_B}. \tag{1}$$

Here, $\Delta S$ is the entropy generated, and $P(\Delta S=A)$ is the probability for $\Delta S=A$. A few quantum analogues of FT have been proposed [12–14]. In Ref. [12] the probability for the energy change between initial and final states is considered, so we need to perform observations twice, while in Ref. [13] the spectrum distribution of work operator is considered and we need to perform only one observation. In Ref. [14], probability for heat exchanged between two systems is considered. The features of the two times measurement approach and the work operator approach are discussed in Ref. [15].

In 1997, Jarzynski presented nonequilibrium equality for free energy difference [Jarzynski equality (JE)] [16] which is a generalization of the minimum work principle. JE states that the change of free energy $\Delta F$ is calculated by taking the sample average of exponentiated work done on a system $e^{-\beta W}$ along irreversible processes which has the following form:

$$\Delta F = -\frac{1}{\beta}\ln\langle e^{-\beta W}\rangle. \tag{2}$$

Here, $\Delta F$ is the free energy difference, $\beta$ is the inverse temperature, $W$ is the work done on a system along some irreversible process, and $\langle\ \rangle$ denotes the sample average. JE was experimentally confirmed by RNA (ribonucleic acid) stretching experiment [17]. Quantum analogues of JE were proposed by Mukamel [18] and by Yukawa [19]. Both quantum analogues employ the situation where a system is initially in thermal equilibrium, and the time evolution is described by the time-dependent Hamiltonian $H(t)$. In Ref. [18] JE is derived in terms of the master equation, while in Ref. [19] the density matrix approach is considered. For classical stochastic systems, Crooks [4] derived classical FT and JE in terms of microscopic reversibility.

In this paper, we give a unified derivation of the quantum analog of FT and JE along the thought of Crooks [4]. The ratio of the probability distributions of entropy generated between time forward and time reversed process is considered, and we define the entropy production analogous to that of Ref. [4]. FT derived here is considered as a quantum extension of that of Ref. [4].

Our emphasis is on the unified treatment of these theorems in terms of microscopic reversibility. For a quasiclassical Langevin system this condition is confirmed.

## II. QUANTUM FLUCTUATION THEOREM

### A. The case of thermally isolated system

In this section, we derive a quantum analog of FT for a system which is externally driven, thermally isolated and whose Hamiltonian $H(t)$ is invariant under time reversal. In order to perform this, we derive the relation (3). In Ref. [20] for the case of the time independent system, the relation (3) was discussed, however, this relation is necessary for the derivation of FT here, we briefly show the derivation. Let the unitary time evolution operator between $t=0$ and $t=T$ be $\hat{U}_T$. And let the density matrix at $t=-0$ be $\rho(0)$ and define $\rho(T) \equiv \hat{U}_T\rho(0)\hat{U}_T^\dagger$. Consider arbitrary observables $\hat{A}$ and $\hat{B}$ whose eigenstates are $|a_n\rangle$ and $|b_m\rangle$, respectively.

Then, we perform the measurements about $\hat{A}$ at $t=+0$ and about $\hat{B}$ at $t=T$. Because the probability we find the initial state as $|a_n\rangle$ is $\langle a_n|\rho(0)|a_n\rangle$, one has

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$$\frac{P_F(|a_n\rangle \to |b_m\rangle)}{P_R(\Theta|b_m\rangle \to \Theta|a_n\rangle)}$$

$$= \frac{\langle a_n|\rho(0)|a_n\rangle|\langle b_m|U_T|a_n\rangle|^2}{\langle b_m|\overleftarrow{\Theta}\Theta\rho(T)\overleftarrow{\Theta}\Theta|b_m\rangle|\ \langle a_n|\overleftarrow{\Theta}U_T^*\Theta|b_m\rangle)|^2}$$

$$= e^{\ln\langle a_n|\rho(0)|a_n\rangle - \ln\langle b_m|\rho(T)|b_m\rangle}. \tag{3}$$

Here, $P_F(|a_n\rangle \to |b_m\rangle)$ is the probability that the forward process $\Pi$ whose observed states at time $t=0$ and $t=T$ are $|a_n\rangle$ and $|b_m\rangle$ occurs. $\Theta$ is the antilinear time reversal operator, and $P_R(\Theta|b_m\rangle \to \Theta|a_n\rangle)$ is the probability that the time reversed process $\Pi^*$ occurs, namely at $t=0$, we observe $\hat{B}$ and find the initial state as $\Theta|b_m\rangle$ according to the ensemble with the time reversed density matrix $\Theta\rho(T)\overleftarrow{\Theta}$ and at $t=T$ observe $\hat{A}$ and find the final state as $\Theta|a_n\rangle$. $\hat{U}_T^*$ is the reversed time evolution operator, $\overleftarrow{\Theta}$ express that antilinear operator $\Theta$ acts to the left, and we used the relation for the unitary operator $\langle b_m|\hat{U}_T|a_n\rangle = \langle a_n|\overleftarrow{\Theta}\hat{U}_T^*\Theta|b_m\rangle$ which results from $\Theta H(t) = H(t)\Theta$. We note that in Ref. [20] the average over $P_F(|\alpha_0\rangle \to |\alpha_T\rangle)$ of the logarithm of the ratio $P_F(|\alpha_0\rangle \to |\alpha_T\rangle)/P_R(\Theta|\alpha_T\rangle \to \Theta|\alpha_0\rangle)$ is shown to be equal to the microcanonical entropy change in the case $|\alpha_0\rangle$ and $|\alpha_T\rangle$ are macrostates provided the Hamiltonian is time independent (see also Refs. [20,21]). Then one has the quantum analogue of FT for the thermally isolated system as shown below.

We define a counterpart of the entropy production $\Delta S$ analogous to the classical one defined in Ref. [4],

$$\Delta S \equiv k_B \ln\langle a_n|\rho(0)|a_n\rangle - k_B \ln\langle b_m|\rho(T)|b_m\rangle. \tag{4}$$

Of course this definition of entropy production depends on the choice of the observables $\hat{A}$ and $\hat{B}$. The state is projected into other basis by measurement and different ways of measurements may cause different entropy productions, unlike in the classical system where all observables commute with each other and the value of the entropy production is unique. We note that $\Delta S$ is clearly considered as entropy production when these observables diagonalize the density matrices $\rho(0)$ and $\rho(T)$, respectively. For example, when the system is in equilibrium at initial and final time and we choose $\hat{A}$ and $\hat{B}$ as Hamiltonian, this condition is satisfied. We treat this case in Sec III. In such cases, $\Delta S$ is considered as entropy production in the sense of Ref. [4], namely $\Delta S \equiv -k_B \ln \rho_f + k_B \ln \rho_i - (Q/T_B)$. Here, $\rho_{i/f}$ is probability density of initial and/or final times, $Q$ is the heat transferred into the system from the heat bath, $T_B$ is the temperature of the heat bath. In comparison to the diagonal representation of von Neumann entropy $S = -\mathrm{Tr}(\rho \ln \rho) = -\Sigma_n \rho_n \ln \rho_n$, the quantity $-k_B \ln \rho_f + k_B \ln \rho_i$ is considered as entropy production of the system, and $-Q/T_B$ is the entropy production of the heat bath. In this section, from the assumption that the system is thermally isolated, heat transferred to the system $Q$ is 0.

Then, one obtains a quantum analog of FT as

$$\begin{aligned} P_F(\Delta S) &\equiv \Sigma_{n,m} P_F(|a_n\rangle \to |b_m\rangle) \\ &\quad \times \delta\{\Delta S - k_B[\ln\langle a_n|\rho(0)|a_n\rangle \\ &\quad - \ln\langle b_m|\rho(T)|b_m\rangle - \beta Q]\} \\ &= e^{\Delta S/k_B} \Sigma_{n,m} P_R(\Theta|b_m\rangle \to \Theta|a_n\rangle) \\ &\quad \times \delta\{\Delta S + k_B[\ln\langle b_m|\rho(T)|b_m\rangle \\ &\quad - \ln\langle a_n|\rho(0)|a_n\rangle + \beta Q]\} \\ &\equiv P_R(-\Delta S)e^{\Delta S/k_B}. \end{aligned} \tag{5}$$

Here, $P_F(\Delta S)[P_R(\Delta S)]$ is the probability that the counterpart of entropy production along the forward (backward) process is $\Delta S$. $Q$ is included for the convenience of the next section.

### B. The system with heat bath

In this section, we consider a system coupled to a heat bath whose total Hamiltonian is $H(t) = H_s(t) + H_B + H_i$. Here, $H_s(t)$, $H_B$, and $H_i$ are the Hamiltonian of system, heat bath and interaction, respectively. We choose $\hat{A}$ and $\hat{B}$ as system Hamiltonian $H_s(0)$ and $H_s(T)$, respectively. And we denote the $n$th eigenvalue of $H_s(t)$ as $E_n(t)$ and corresponding eigenvector as $|n,t\rangle$. We restrict ourselves to the system where the microscopic reversible condition (6) is satisfied,

$$\frac{P_F(|n,0\rangle \to |m,T\rangle)}{P_R(\Theta|m,T\rangle \to \Theta|n,0\rangle)} = e^{-\beta Q}. \tag{6}$$

Here, $P_F(|n,0\rangle \to |m,T\rangle)$ denotes the conditional probability that the system is initially in the energy level $|n,0\rangle$ and after some duration time $T$ jumps to the level $|m,T\rangle$. The distinction of time forward and reversed processes is the same as in the preceding section. $Q$ is the heat transferred to the system from the heat bath. This relation plays an essential role in Ref. [4] for the derivation of FT. Also in quantum system, there are many situations where the microscopic reversibility does hold. In the appendix, as a physically important system which satisfies this relation, we confirm this microscopic reversibility for the so-called quasiclassical Langevin system [23].

The rest of this section is devoted for the derivation of FT for the system with the heat bath in terms of the relation (6). The total probability $P_F^{\mathrm{Tot}}(|n,0\rangle \to |m,T\rangle)$ that the system is initially in $|n,0\rangle$ and after some duration $T$ found in $|m,T\rangle$ is given as the product of initial state probability and conditional probability,

$$\frac{P_F^{\mathrm{Tot}}(|n,0\rangle \to |m,T\rangle)}{P_R^{\mathrm{Tot}}(\Theta|m,T\rangle \to \Theta|n,0\rangle)} = \frac{\langle n,0|\rho_s(0)|n,0\rangle}{\langle m,T|\overleftarrow{\Theta}\Theta\rho_s(T)\overleftarrow{\Theta}\Theta|m,T\rangle} e^{-\beta Q}$$

$$= e^{\Delta S/k_B}.$$

We defined entropy production $\Delta S$ as $\Delta S/k_B \equiv \ln\langle n,0|\rho_s(0)|n,0\rangle - \ln\langle m,T|\rho_s(T)|m,T\rangle - \beta Q$. Here, we define the reduced density matrix $\rho_s(t) \equiv \mathrm{Tr}_B(\rho_{\mathrm{Tot}})$ as a system density matrix.

Then one has the quantum analog of FT for the system interacting with a heat bath as in the preceding section (5). $P_F(\Delta S) = P_R(-\Delta S)e^{\Delta S/k_B}$ which corresponds to the classical FT [4].

## III. QUANTUM JARZYNSKI EQUALITY

In this section, we derive quantum Jarzynski equality in terms of microscopic reversibility (6).

At first, we set up the framework on which we discuss here. We assume that the heat bath is large enough and interaction with the system is weak enough so that the following conditions do hold. As the system density matrix, we use the reduced density matrix $\rho_s(t) \equiv \mathrm{Tr}_B[\rho_{\mathrm{Tot}}(t)]$. In order to discuss the free energy change between two equilibrium states, we require that the density matrices of the system $\rho_s(0)$ and $\rho_s(T)$ are canonical distribution at the same temperature. We set observables $\hat{A}$ and $\hat{B}$ as the system Hamiltonian $H_s(0)$ and $H_s(T)$ as in the preceding section and denote the eigenstate of $H_s(0)$ and $H_s(T)$ as $|n,0\rangle$ and $|m,T\rangle$ with the eigenvalues $E_n(0)$ and $E_m(T)$, respectively. Then one finds

$$\langle n,0|\rho_s(0)|n,0\rangle = \frac{1}{Z_1} e^{-\beta\langle n,0|H_s(0)|n,0\rangle},$$

$$\langle m,T|\rho_s(T)|m,T\rangle = \frac{1}{Z_2} e^{-\beta\langle m,T|H_s(0)|m,T\rangle}, \tag{7}$$

where $Z_1$ and $Z_2$ are the partition function at $t=0$ and $T$,

$$\frac{P^{\mathrm{Tot}}(|n,0\rangle \to |m,T\rangle)}{P^{\mathrm{Tot}}(\Theta|m,T\rangle \to \Theta|n,0\rangle)} = e^{\beta[E_m(T)-E_n(0)-Q]-\Delta F}. \tag{8}$$

Here, $\Delta F \equiv -(1/\beta)\ln(Z_2/Z_1)$ is the change of Helmholtz free energy. We note that $W \equiv E_m(T) - E_n(0) - Q$ is considered as work externally done on the system. Thus

$$\langle e^{-\beta(W-\Delta F)}\rangle = \Sigma_{n,m} P_F^{\mathrm{Tot}}(|n,0\rangle \to |m,T\rangle) \times \frac{P_R^{\mathrm{Tot}}(\Theta|m,T\rangle \to \Theta|n,0\rangle)}{P_F^{\mathrm{Tot}}(|n,0\rangle \to |m,T\rangle)} = 1. \tag{9}$$

Here, $\langle\ \rangle$ denotes the average over the probability $P_F^{\mathrm{Tot}}(|n,0\rangle \to |m,T\rangle)$. Therefore, one has

$$\Delta F = -\frac{1}{\beta}\ln\langle e^{-\beta W}\rangle. \tag{10}$$

In summary, we derived a quantum extension of Jarzynski equality and FT in terms of microscopic reversibility (6). And this relation (6) is confirmed for quasiclassical Langevin system. This unified treatment is the quantum version of Crooks' derivation of FT and Jarzynski equality for the classical system.

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## APPENDIX: MICROSCOPIC REVERSIBILITY FOR QUASICLASSICAL LANGEVIN SYSTEM

As a physically important system which satisfies the microscopic reversibility (6), we confirm this relation for the so-called quasiclassical Langevin system [22,23]. For the classical Langevin system, microscopic reversibility was derived by Narayan and Dhar [7]. Here, we utilize their result for quantum Langevin system in the semiclassical regime. To do this, it is most convenient to use the quantum noise theory by Gardiner [22]. Because a straightforward derivation of Langevin equation is known [22,23,24], we consider a system which interacts with a harmonic reservoir. Let the total Hamiltonian be

$$H(t) = \frac{p^2}{2m} + V(q,\lambda(t)) + \frac{1}{2}\int d\lambda[(p_\lambda - \kappa_\lambda q)^2 + \omega_\lambda^2 q_\lambda^2]. \tag{A1}$$

Here, $q$,$p$ are the canonical coordinates of system and $q_\lambda$,$p_\lambda$ are those of heat bath. $\lambda(t)$ in the potential term is the control parameter corresponding to the external agent. The system is assumed to be initially uncorrelated to the heat bath and the initial density operator of the heat bath $\rho_B$ is assumed to be canonical. $[\rho(0) = \rho_s \otimes \rho_B, \rho_B = (1/Z)e^{-\beta\int d\lambda\hbar\omega_\lambda(a_\lambda^\dagger a_\lambda + 1/2)}]$. This model is standard and one of the ideal examples which describes the system interacting with the heat bath. Let $Y(t)$ be an arbitrary system operator in the Heisenberg picture. First, we define the quantity $\mu(t)$ by $\mathrm{Tr}_s[Y(t)\rho_s \otimes \rho_B] = \mathrm{Tr}_s[Y \otimes \mu(t)]\rho_B$.

The equation of motion for $\mu(t)$ (adjoint equation) is

$$\dot{\mu}(t) = A_0\mu(t) + \alpha(t)A_1\mu(t), \tag{A2}$$

where $A_0\mu(t) \equiv (i/\hbar)[H_s, \mu(t)] + (i/2\hbar)[[\gamma\dot{q}, \mu(t)]_+, q]$, $A_1\mu(t) \equiv (i/\hbar)[q, \mu(t)]$, and $\alpha(t)\mu(t) \equiv \frac{1}{2}[\xi(t), \mu(t)]_+$. $\xi(t) \equiv i\int d\lambda\kappa_\lambda\sqrt{\hbar\omega_\lambda/2}[-a_\lambda(0)e^{-i\omega_\lambda t} + a_\lambda^\dagger(0)e^{i\omega_\lambda t}]$ is the Langevin force and here we assume the density of the state is constant $2\gamma/\pi$ so that the adjoint equation is Markov (The Markov approximation). Then $\xi(t)$ satisfies the fluctuation dissipation theorem, $\langle[\xi(t), \xi(t')]_+\rangle = (2\hbar\gamma/\pi)\int_0^\infty d\omega\omega\coth(\beta\hbar\omega)\cos\omega(t-t')$, where the average $\langle\cdots\rangle \equiv \mathrm{Tr}_B(\cdots\rho_B)$ denotes the average over the bath variables. This equation is rewritten in the form of the Kramers equation. Suppose the Wigner function corresponding to $\mu(t)$ is $W(q,p,t) \equiv \int dr\langle q + (r/2)|\mu(t)|q - (r/2)\rangle e^{-i(rp/\hbar)}$. Then

$$\frac{\partial W}{\partial t} = \left[-\frac{\partial}{\partial x}\frac{p}{m} + \frac{\partial}{\partial p}\left(V'(x,\lambda(t)) + \gamma\frac{p}{m} - \alpha(t)\right)\right]W + \left[\sum_{n=1}^{\infty}\frac{1}{(2n+1)!}\left(\frac{i\hbar}{2}\right)^{2n}\frac{\partial^{2n+1}}{\partial p^{2n+1}}V^{(2n+1)}(x,\lambda(t))\right]W. \tag{A3}$$

Due to the associative nature of $\alpha(t)$, the first term of this equation is equivalent to the following $c$-number quasiclassical Langevin equation. Note that up to $O(\hbar)$ the second term is negligible and furthermore, this approximation becomes better in the large friction limit,

$$\dot{x}=\frac{p}{m},\quad \dot{p}=-V'(x,\lambda)-\gamma\frac{p}{m}+\tilde{\alpha}(t). \tag{A4}$$

Here $\tilde{\alpha}(t)$ is the $c$-number stochastic process where all the moments are equal to that of $\alpha(t)$ with respect to the thermal average. $\langle\tilde{\alpha}(t)\tilde{\alpha}(t')\rangle=\mathrm{Tr}_B[\alpha(t)\alpha(t')\rho_B]$. In particular, under the definition of thermal noise operator $\xi(t)$ and the assumption that $\rho_B$ is canonical, we can show that $\tilde{\alpha}(t)$ is the Gaussian process. Thus the mean and variance are sufficient to determine the stochastic feature of $\tilde{\alpha}(t)$, $\langle\tilde{\alpha}(t)\rangle=0$, $\langle\tilde{\alpha}(t)\tilde{\alpha}(t')\rangle=\frac{1}{2}\langle[\xi(t),\xi(t')]\rangle$.

We derived qausiclassical Langevin equation, and then we can use the result of Narayan and Dhar [7] to obtain the relation (6). From the Gaussian nature of $\tilde{\alpha}(t)$, one has the probability functional $P_{F/R}[\tilde{\alpha}(t)]$ for each realization of $\tilde{\alpha}(t)$.

$$\begin{aligned}P_{F/R}[\tilde{\alpha}(t)]&\\ &=Ce^{-1/2\int_0^t ds\int_0^t ds'\,\tilde{\alpha}(s)[(\gamma\hbar/\pi)\int_0^\infty d\omega\omega\,\coth(\beta\hbar\omega)\cos\,\omega(s-s')]^{-1}\tilde{\alpha}(s')}\\ &=Ce^{-1/2\int_0^t ds\int ds'\,\tilde{\alpha}(s)[(\beta/2\gamma)\delta(s-s')+O(\hbar^2)]\tilde{\alpha}(s')}.\end{aligned}$$

$A(s,s')^{-1}$ denote the inverse of $A(s,s')$ as a kernel. Here the distinction of time forward and reverse $F/R$ means that the time reversal of the control parameter $\lambda(t)$ is also considered. We consider up to $O(\hbar)$ (semiclassical regime) and thus the $O(\hbar^2)$ term is omitted. Then after the same discussion of Ref. [7], one has the following relation:

$$\frac{P_F(q_f,p_f|q_i,p_i)}{P_R(q_i,-p_i|q_f,-p_f)}=e^{-\beta[H_s(q_f,p_f,\lambda(t))-H_s(q_i,p_i,\lambda(0))-W]}. \tag{A5}$$

Here, $P_{F/R}(q_f,p_f|q_i,p_i)$ is the probability that the initial and final states for the equation (A4) are $(q_i,p_i)$ and $(q_f,p_f)$, respectively. $H_s(q,p,\lambda(t))=(p^2/2m)+V(q,\lambda(t))$ is the system Hamiltonian at time $t$ and $W=\int_0^t ds(\partial/\partial\lambda)V(q,\lambda(s))\dot{\lambda}(s)$ is the work externally done through the control parameter $\lambda(t)$. Then we choose the initial and final points of the phase space according to the initial and final Wigner functions. And the probability that the initial and final states are $|n,0\rangle$ and $|m,T\rangle$ is calculated as

$$\begin{aligned}P_F(|n,0\rangle\to|m,T\rangle)=\bigg\langle\int dq_i dp_i W_n(q_i,p_i,0)\int dq_f\,dp_f&\\ \times W_m(q_f,p_f,T)\delta(q_f-q(t,q_i,p_i))&\\ \times\delta(p_f-p(t,q_i,p_i))\bigg\rangle.&\end{aligned} \tag{A6}$$

Here, $q(t,q_i,p_i)$ and $p(t,q_i,p_i)$ are the solutions of the quasiclassical Langevin equation with the initial condition $(q(0),p(0))=(q_i,p_i)$ and $\langle\ \rangle$ denotes the average over $\tilde{\alpha}(t)$. $W_n(q_i,p_i,0)$ and $W_m(q_f,p_f,T)$ are the initial and final Wigner functions. In order to utilize the result for the classical system (A5), we note that the Wigner function is very localized in semiclassical regime (for the harmonic potential, proportional to $e^{-(1/\hbar)[(p^2/\sqrt{km})+\sqrt{km}q^2]}$). Therefore one may take a following view. Only the neighborhood of energy surfaces $H(q_i,p_i,\lambda(0))=E_n(0)$ and $H(q_f,p_f,\lambda(T))=E_m(T)$ does the main contribution to the integration above. The work done on the system $W$ for long enough time duration is nearly constant irrespective of the initial condition $q(0),p(0)$. And the exponent of (A5) is within this approximation independent from the path $\tilde{\alpha}(t)$. Thus one can finally confirm the microscopic reversibility (6) [25] for the quasiclassical Langevin system,

$$\frac{P_F(|n,0\rangle\to|m,T\rangle)}{P_R(\Theta|m,T\rangle\to\Theta|n,0\rangle)}=e^{-\beta[E_m(T)-E_n(0)-W+O(\hbar)]}\simeq e^{-\beta Q}. \tag{A7}$$

Here the first equality results from the above viewpoint and the $O(\hbar)$ deviation is caused by quasiclassical approximation due to the symmetrization procedure such as $qp\to\frac{1}{2}(qp+pq)$.

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