Microscopic Work Distribution of Small System in Quantum Isothermal Process

H. T. Quan, 1 S. Yang, 2 and C. P. Sun 2

1Theoretical Division, MS B213, Los Alamos National Laboratory, Los Alamos, NM, 87545, U.S.A.
2Institute of Theoretical Physics, Chinese Academy of Sciences, Beijing, 100080, China

For a two-level quantum mechanical system, we derive microscopically the exact expression for the fluctuation of microscopic work in a multi-step non-equilibrium process, and we rigorously prove that in an isothermal process, the fluctuation is vanishingly small, and the most probabilistic work just equals to the difference of the free energy. Our study demonstrates that the convergence of the microscopic work in the isothermal process is due to the nature of isothermal process rather than usual thermodynamic limit condition. Our investigation justifies the validity of “minimum work principle” formulation of the second law even for a small system far from thermodynamic limit.

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I. INTRODUCTION

Thermodynamics usually deals with the systems of infinite number of degree of freedoms, in which relative fluctuations of the observable, e.g., energy, particle number, are inversely proportional to the square root of the numbers of the particles of the system [1]. Hence for a macroscopic system consisting of infinite number of particles, the fluctuations are vanishingly small and the ensemble average can describe thermodynamic phenomena completely. However, concerning small systems, usually the fluctuations of the microscopic values of thermodynamic observable will become appreciable, and ensemble average alone can not longer give a complete description [2]. In recent years, increasing interests are drawn to the study of thermodynamics of small system, and the emphases are put on the fluctuations of the microscopic value of the observable, instead of their ensemble average. Some notable progresses have been made, examples including the Jarzynski equality [3, 4] and the Fluctuation Theorem [5]. The former connects the free energy difference of two equilibrium states with ensemble average of microscopic work in non-equilibrium process while the later illustrates the probabilistic “entropy decrease” of a closed system within short time, or transient “violation” of the second law. These studies shed new light on the understanding of non-equilibrium thermodynamical processes of biological motors in cells and promise important applications to the design of small-size machines. In all these studies, for small systems, though fluctuations of most observables are appreciable, there exists an exception – the work done during a slowest reversible equilibrium process (we use isothermal processes to replace slowest reversible processes hereafter). It has been pointed out that the fluctuation of microscopic work done by or on a macroscopic system during a slowest reversible process is vanishingly small [3, 7]. Nevertheless, though the fluctuations of microscopic work for small systems in finite-time irreversible processes has been extensively studied [9], and the vanishing fluctuation of microscopic work of classical small systems specially concerning thermodynamic isothermal process has been point out, to our best knowledge, a rigorous proof of the above result from microscopic aspect is still lacking, and its quantum mechanical generalization has not been studied yet.

In this paper, we will investigate this problem by simulating a quantum isothermal process with infinite number of infinitesimal quantum adiabatic process (QAP) and quantum isochoric process (QIP) [8-10]. We prove rigorously from microscopic aspect the above result that, for a two-level system, the fluctuations of the microscopic work during an quantum isothermal process [10] is vanishingly small. We emphasize that, different from most cases in conventional statistical mechanics, where fluctuations vanishes in the thermodynamic limit, the vanishing work fluctuations for a small system in an isothermal process is due to the intrinsic nature of isothermal process. Our study also verify the universal validity of the “minimum work principle” formulation of the second law: it holds even for a small system!

II. THE THERMODYNAMIC PROCESS IN PARAMETER SPACE

We consider a two-level quantum mechanical system with excited (ground) states |e\rangle (|g\rangle) with instantaneous eigenenergy $E_e(t)$ ($E_g(t)$) depending on time $t$. This two-level system can be modeled as a spin-1/2 in an external magnetic field. It interacts with a heat bath of inverse temperature $\beta$, which can be universally modeled as a collection of many bosons with creation (annihilation) operators $a_q^\dagger$ ($a_q$) [11]. The model Hamiltonian reads [12,13].

$$H = \Delta(t)\sigma_z + \sum_q \omega q a_q^\dagger a_q + \sum_q (\lambda q \sigma_- a_q^\dagger + h.c.),$$

(1)

where $\sigma_- = |g\rangle \langle e| = (\sigma_x - i\sigma_y)/2$ and $\sigma_z = (|e\rangle \langle e| - |g\rangle \langle g|)/2$. Initially, let the two-level system be thermalized to equilibrium. Then we alter the magnetic field slowly so that the energy level spacing $\Delta(t)$ slowly changes from $\Delta_A$ to $\Delta_B$. During the controlling process illustrated by the smooth curve $\tilde{AB}$ in Fig. 1, the work is done on the system. In the infinitely slow process, which can be alternatively regarded as a quantum isothermal process [10], the two-level system is in the thermal equilibrium at every instant, which is described by the diagonal reduced density matrix $\rho_S(t) = P_e(t)|e\rangle \langle e| + [1 - P_e(t)]|g\rangle \langle g|$, where $P_e(t) = \exp[-\Delta(t)]/(1 + \exp[-\beta\Delta(t)])$ satisfies the Gibbs
FIG. 1: Schematic illustration of a quantum isothermal process [10] AB. Here the horizontal axis $P_x$ is the occupation probability in the excited state of the two-level system, and the vertical axis indicates the level spacing of the two-level system. The smooth curve $AB$ represents the isothermal process, whose “equation of state” can be expressed as $\Delta(t) = -\beta^{-1} \ln(P_x^{-1} - 1)$. The horizontal and vertical lines represent QIC and QAP. We can use many small QAP and QIP to model the quantum isothermal process. For example, we use a “five-step staircase” path (green) $A \rightarrow C \rightarrow D \rightarrow \cdots \rightarrow B$ to simulate the smooth curve $AB$. “One-step” path (blue) and “twenty-step” path (orange) are also illustrated.

distribution. It should be pointed out that, during the isothermal process, there is a heat exchange between the two-level system and the heat bath.

For such an isothermal process, it is difficult to calculate the microscopic work distribution directly. According to Ref. [8, 9, 10], however, this process can be simulated by a series of QAP and QIP. In QAP (QIC) processes, there is only work done (heat exchange). Hence, using the changes of eigen-energies of microscopic state at instant $t = A, C, D$, we can indirectly calculate the microscopic work done (heat exchange) $dW = E_\alpha(C) - E_\alpha(A)$ ($dQ = E_\alpha(D) - E_\beta(C)$), for $\alpha, \beta = e, g$. In the parameter space, these QAP and QIP series processes are represented by the “stair” path $(A \rightarrow C \rightarrow D \rightarrow \cdots \rightarrow B)$ in Fig. 1. When every step of the “stair” path becomes infinitesimal, the “stair” path becomes equivalent to the isothermal process $AB$. In this way we simulate the quantum isothermal process with $N$ equal-height steps (see Fig. 1) with the small height $\Delta = (\Delta_B - \Delta_A)/N$ where $\Delta_A$ and $\Delta_B$ are the level spacings at point $A$ and point $B$ respectively. The level spacings of the two-level system after the $(j - 1)$-th QIC is

$$\Delta_j = \Delta_A + (j - 1)\Delta,$$

for $j = 1, 2, \cdots, N + 1$. The initial and final point $A$ and $B$ corresponds to $j = 1$ and $j = N + 1$ respectively. When we fix the initial point $A$, and the final point $B$, the jump $\Delta$ in every step decrease with the increase of the step number $N$, and $\Delta$ approaches zero when $N$ becomes infinity. Obviously, when $N \rightarrow \infty$, the “stair” path approaches its asymptotic behavior - the isothermal path (see Fig. 1). When the system reaches thermal equilibriums, the occupation probabilities obeys the Gibbs distribution defined by

$$P^j_e = e^{-\beta\Delta_j}[1 + e^{-\beta\Delta_j}]^{-1}, P^j_g = P^j_e e^\beta\Delta_j \quad (3)$$

We remark that there are three time scales in our process: $\tau_a$ for quantum adiabatic approximation, $\tau_c$ the control time of changing the magnetic field, hence the level spacing, and $\tau_r$ the relaxation of the two-level system. According to Ref. [13], $\tau_r$ is determined by the coupling strength $\lambda_g$ [1]. We consider the case that $\tau_a \ll \tau_c \ll \tau_r$ for a quantum adiabatic process where we can define the microscopic work in every realization of the process.

III. MICROSCOPIC WORK DISTRIBUTION

Having defined the “path” in the parameter space $(\Delta - P_e)$ space, we can further introduce the microscopic work and its corresponding probabilities for a given “path”. Actually, the definition of microscopic work is very similar to that in Ref. [4]. In the above path divided into many “steps”, the first step $A \rightarrow C \rightarrow D$ consists of a QAP $A \rightarrow C$, and a QIP $C \rightarrow D$. At the beginning (the point $A$ of Fig. 1), the system is initially in a thermal equilibrium state $\rho_0(A)$, which implies that the system is either in its microscopic state $|g\rangle$ or $|e\rangle$ with probabilities $P^1_g$ and $1 - P^1_g$ respectively. In the first QAP $A \rightarrow C$, the system remains in its microscopic state $|g\rangle$ ($|e\rangle$) if the system is initially in its microscopic state $|g\rangle$ ($|e\rangle$). As there is no heat exchange in the QAP, the work done by external controller is just the change of the microscopic energy $W_a = E_\alpha(C) - E_\alpha(A)$ for $\alpha = e, g$. Correspondingly the work done during $A \rightarrow C$ can be either $\Delta_e - \Delta_A$ or 0 with probabilities $P^1_g$ and $1 - P^1_g$ respectively. This also agrees with the definition of work in quantum mechanical system: work is associated with the change of the level spacing $|\Delta_e - \Delta_A|g\rangle [9, 10, 14]$.

After the QAP, a quantum isochroc process $C \rightarrow D$ (see Fig. 1) follows. Here, there is no work done according to the definition of work in quantum mechanical system $|\Delta_e - \Delta_A|g\rangle$ because there is no change in the eigenenergies. Nevertheless, there is heat exchange between the system and the bath. The QIP last long enough ($\gg \tau_r$) so that the system can reach thermal equilibrium with the heat bath. After a thermalization for long-time, the two-level system reach thermal equilibrium with the heat bath again $|\Delta_e - \Delta_B|g\rangle$ at instant $D$ indicated in Fig. 1. Then a second step $D \rightarrow F$ begins. Similarly, the microscopic work $0$ or $\Delta_3 - \Delta_2$ is done in this step with probabilities $1 - P^2_g$ and $P^2_g$. The microscopic work done and their probabilities for the remaining steps can be obtained through a similar analysis. Because in every QIP, the system is independently thermalized by the heat bath, then there should be no correlations of the probabilities distributions in every two
neighbored steps, or alternatively, this process is Markovian process. Hence, the total microscopic work done after the $N$-step is a sum of microscopic works done in all steps and the joint probabilities for the $N$-step as a whole is the product of that of all steps.

For a special example that the microscopic work done during the whole process is $W = N\Delta$ where $\Delta$ is that for each QIP step, the joint probabilities for the system keeping in $|e\rangle$ in every QIP is $P\{N\Delta\} = P^1_e P^2_e \cdots P_N^e$. The more general case with microscopic work $W = (N - k)\Delta$ corresponds to a microscopic process, in which $k$ out of $N$ QIPs ends with the system in its microscopic state $|g\rangle$. The probability $P(k) := (N - k)\Delta$ with the microscopic work $W = (N - k)\Delta$ in the $N$-step path is given by the following equation:

$$P(k) = \left(\prod_{j=1}^N P^j_e\right) \frac{\left(\sum_{l=0}^{k-1} e^{\beta\Delta_B} - e^{\beta(\Delta_A + l\Delta)}\right)}{e^{\beta\Delta_A - 1}}, \quad (4)$$

To prove the above result, we first consider the case with $k = 1$. For this case, there is one and only one out of the $N$ QIPs, in which the system ends up in the microscopic state $|g\rangle$. Then the corresponding probability can be calculated as $P(1) = (1 - P^1_e) (1 - P^2_e) \cdots (1 - P^N_e) e^{\beta\Delta_B}$ or $P(1) = \left(\prod_{j=1}^N P^j_e\right) \frac{e^{\beta\Delta_B} - e^{\beta\Delta_A}}{e^{\beta\Delta_A - 1}}, \quad (5)$

That means the Eq. (4) holds for $k = 1$. Similarly we can check the case with $k = 2$. For this case, there are two out of the $N$ QIPs in which the system ends up in the microscopic state $|g\rangle$. Hence its probability can be expressed as $P(2) = (1 - P^2_e) (1 - P^2_e) P^3_e \cdots P^N_e + (1 - P^2_e) P^1_e (1 - P^2_e) e^{\beta\Delta_B} \sum_{x_1=1}^{x_2=1} e^{-\beta\Delta_A(x_1+x_2)}$ or $P(2) = \left(\prod_{j=1}^N P^j_e\right) \frac{[e^{\beta\Delta_B} - e^{\beta\Delta_A}] [e^{\beta\Delta_B} - e^{\beta(\Delta_A + \Delta)}]}{(e^{\beta\Delta_A} - 1)(e^{2\beta\Delta_A} - 1)}, \quad (6)$

Hence Eq. (4) also holds for $k = 2$ case. In general, for an arbitrary $k$, the corresponding probability can be expressed as

$$P(k) = \left(\prod_{j=1}^N P^j_e\right) e^{k\beta\Delta_B} \chi(k), \quad (7)$$

where $\chi(k) = \sum_{x_1=1}^{x_2=1} \cdots \sum_{x_k=1}^{x_{k+1}-1} e^{-\beta\Delta(x_1+x_2+\cdots+x_k)}$. As $\chi(k), (k = 1, 2, \cdots, N)$ satisfy the following relation

$$\chi(k) = \sum_{i=1}^{k-1} \left[ \left(\prod_{j=1}^i \frac{-1}{1 - e^{-\beta\Delta}}\right) (-e^{-i\Delta}) \chi(k-i) \right] + \left(\prod_{j=1}^k \frac{-1}{1 - e^{-\beta\Delta}}\right) \left[ e^{-k(N+1)\Delta} - e^{-k\Delta} \right], \quad (8)$$

we can use the complete induction method to prove that the $\chi(k)$ can be generally expressed as

$$\chi(k) = \prod_{l=0}^{k-1} \frac{-e^{\beta N\Delta}}{e^{\beta l(1+\Delta)}} \quad (9)$$

Substituting Eq. (9) into Eq. (7), we obtain Eq. (4). Hence, by now we prove the general result given by Eq. (4).

IV. MOST PROBABILISTIC DISTRIBUTION AND FLUCTUATION

The above equation (4) can result in the main conclusion in this paper. From the above microscopic work distribution function (4), we obtain the ratio $R(k) = P(k+1)/P(k)$ of distributions for two close microscopic work, i.e.,

$$R(k) = \frac{e^{\beta\Delta_B} - e^{\beta(\Delta_A + k\Delta)}}{e^{\beta(k+1)\Delta} - 1} \quad (10)$$

Let $\tilde{k}$ maximize the probability distribution $P(k)$ for the microscopic work $[N - (\tilde{k} + 1)\Delta]$. Then $P(\tilde{k}) \geq P(\tilde{k} \pm 1)$, or $R(\tilde{k}) \leq 1$ or $R(\tilde{k} - 1) \geq 1$. For very large $\tilde{k}$, $R(\tilde{k}) \simeq 1$ that

$$\tilde{k} = \frac{1}{\beta} \ln \left[ 1 + \exp[\beta\Delta_B] \right]. \quad (11)$$

In the large $N$ limit, the above equation determines the microscopic work $W = (N - k)\Delta$ with most probabilistic distribution

$$\tilde{W} = \frac{1}{\beta} \ln \left( \frac{1 + e^{\beta\Delta_B}}{1 + e^{\beta\Delta_A}} \right) \quad (12)$$

which is just the free energy difference $\Delta F_{AB} = F_B - F_A$ where $F_j = \ln[1 + \exp(\beta\Delta_B)]/\beta$ for $j = A, B$

Next let us give a heuristic analysis of the dispersion of the work distribution (4). Because all steps in the “stair” path are independent with each other, thus the whole process can be regarded as Markovian. So the variance of total microscopic work done during the whole process equals to the sum of variance of local microscopic work in every step, i.e.,

$$\langle W^2_{AB} \rangle - \langle W_{AB} \rangle^2 = \sum_{j=1}^N \left( \langle W_j^2 \rangle - \langle W_j \rangle^2 \right), \quad \text{where } W_j \text{ is the microscopic work done during the } j \text{th QAP, and the local fluctuations}$$

$$\langle W_j^2 \rangle - \langle W_j \rangle^2 = \Delta^2 [P^j_e - (P^j_e)^2] \quad (13)$$

for different $j$ are similar. Here $\Delta$ is inversely proportional to $N$, and $\langle W_{AB} \rangle$ being independent of $N$, the relative variance of $W_{AB}$ is inversely proportional to

$$\sqrt{\frac{\langle W^2_{AB} \rangle - \langle W_{AB} \rangle^2}{\langle W_{AB} \rangle}} \propto \frac{1}{\sqrt{N}} \quad (14)$$

We numerically plot the work distribution function (see Fig.2) based on the above analytical result (4) to test the above analysis. Here we choose the step number $N$ from 1 to 10000.
For $N = 1$, the “stair” path becomes a “one-step” path consists of an QAP and an QIP (see Fig. 1). The microscopic work corresponding to the “one-step” path is either $\Delta_B - \Delta_A$ or 0 with the probability $P(W = \Delta_B - \Delta_A) = P^1_e$ or $P(W = 0) = 1 - P^1_e$. In the above figures, we choose $\exp(-\beta \Delta_A) = 1/2$, ($P^1_e = 1/3$), and the numerical result agrees well with our analysis. For $N = 5$ (see Fig. 1), the possible microscopic work can be $W = i(\Delta_B - \Delta_A)/5$, $i = 0, 1, 2, \ldots, 5$. The numerical result indicates vanishing probability for $W = \Delta_B - \Delta_A$. For $N = 20$ (see Fig. 1), the numerical result show even more vanishing probabilities of microscopic work. That is, the dispersion (fluctuation) of microscopic work decrease with the increase of $N$. Actually, from the above numerical figures, it is not difficult to find that the dispersion of the microscopic work distribution is inversely proportional to the square root of $N$. For example, the dispersion for $N = 100$ is ten times that for $N = 10000$ case (see Fig. 2). Hence, numerical results agrees well with our heuristic analysis and both they verified our main result, when $N \to \infty$, the fluctuations of microscopic work vanishes.

V. MINIMUM WORK PRINCIPLE FOR A TWO-LEVEL SYSTEM

As we have mentioned before, for small systems and within short time, the formulation “entropy never decrease for a closed system” of the second law may be transiently “violated” probabilistically due to appreciable fluctuations. A straightforward question is: will the other formulations of the second law, e.g., the minimum work principle also be transiently “violated” probabilistically for small systems? The “minimum work principle” states that “when varying the speed of a given process for an initially equilibrium system, the work is minimal for the slowest realization of the process.” In the following we will test the validity of “minimum work principle” for a two-level system by utilizing the formula we derived above. The average work over all possible realizations for a given $N$-step path can be expressed as

$$\langle W \rangle_N = \sum_{k=0}^{N} \left( \prod_{j=1}^{N} P_j^k \right) \left( \prod_{l=0}^{k-1} \frac{e^{\beta \Delta_B} - e^{\beta(\Delta_A+i\Delta)}}{e^{\beta(l+1)\Delta} - 1} \right) (N-k)\Delta,$$

(15)

FIG. 3: Averaged work $\langle W \rangle_N$ as a function of $N$ [15]. The steps $N$ chosen here are $5^0 = 1, 5^1 = 5, 5^2 = 25, 5^3 = 125, 5^4 = 625, 5^5 = 3125, \text{and} 5^6 = 15625$. It can be seen that the averaged work is a monotonically decreasing function of $N$. In the one-step path ($N = 1$), the averaged work equals to $\langle W \rangle_1 = (ln 3 - ln 2)/3 \approx 0.135155kB T$; In the $15625$-step path, the averaged work equals to $\langle W \rangle_{15} = (ln 3 - ln 2)/3 \approx 0.11784kB T$, which is very close to its asymptotic value $\Delta F_{AB} = [ln(1+1/2) - ln(1+1/3)]kB T \approx 0.117783kB T$. Thus, it can be inferred that the “minimum work principle” still holds for a two-level system.

In Fig. 3 we plot the averaged work $\langle W \rangle_N$ as a function of $N$ [15]. It can be seen that for the two-level system, $\langle W \rangle_N$ is a monotonically decreasing function of $N$ (time $t$), and when $N \to \infty$, ($t \to \infty$), the averaged work $\langle W \rangle_N$ approaches an asymptotic value, and its minimum value – the difference of the free energy. Thus, from the numerical result it can be inferred that the “minimum work principle” still holds for a two-level system.

The above proof of minimum work principle can be alternatively understood in the following way. From the above analytical and numerical result, we observed that the fluctuation of microscopic work in an isothermal process vanishes,
and then the work of the most probabilistic distribution equals to the difference of the free energy $W = \Delta F$. According to Ref. [3], $\langle W_{\text{irre}} \rangle \geq \Delta F$, where $\langle W_{\text{irre}} \rangle$ is the average work done during an irreversible process. Combining the two results, we have $\langle W_{\text{irre}} \rangle \geq W$. Thus we proved the minimum work principle for small system.

VI. DISCUSSION AND CONCLUSION

Before concluding this paper, we would like to emphasize the following points: First, the technique of simulating isothermal processes with adiabatic processes and isochoric processes are important to our proof, which enables us to establish the connection between large time limit and large N limit. Second, the calculation of exact expression of microscopic work in our paper is non-trivial because the work contributions in the different steps are not identically distributed. Hence, it is different from the law of large numbers, with time as the large number [2]. Third, we proved the “minimum work principle” formulation of the second law stands for even small system, though other formulations maybe transiently “violated” probabilistically [5]. This is not surprising because “minimum work principle” concerns infinite-long-time processes, which has no contradiction with the transient “violation” of the second law for small systems predicted by the Fluctuation Theorem. Actually, the Fluctuation Theorem does not constitute real violation of the second law, which is a statistical law and holds when averaged over different realization of the process. Fourth, the isothermal process is reversible, but the finite N “step path” is irreversible, due to the QIP (thermolization) is irreversible. We can thus expect that the work dissipation [6,7] for the finite N step path will be finite and will decrease with the increase of N, and finally vanishes when N approaches infinity.

In summary, by simulating an quantum isothermal process with infinite many infinitesimal QAP and QIP, we obtain the analytical expressions of microscopic work distribution in an isothermal process. Through both analytical and numerical analysis, we rigorously verify that the fluctuations of microscopic work distribution vanishes even for a small system in an isothermal process. This result is different from the usual fluctuations in statistical mechanics, e.g., the energy fluctuation and particle number fluctuation in canonical ensemble and grand canonical ensemble, where the fluctuations of energy and particle numbers approaches zero when the system approaches thermodynamic limit (particle number approaches infinity $N \to \infty$). Here, however, even for single particle system, we microscopically demonstrate the vanishing of microscopic work fluctuation. Because $N \to \infty$ is a must to simulate an isothermal process, we conclude that the vanishing of microscopic work fluctuations is due to the intrinsic nature of isothermal process, rather than the thermodynamic limit of the system size. We also prove that for a small system, the “minimum work principle” formulation of the second law holds though other formulations maybe transiently “violated” probabilistically. Finally we would like to point it out that our result is universal and does not depend on the specific model used here, because the technique of simulating the isothermal process with the isochoric process and the adiabatic process can be applied to any systems. Generalizations of our current discussion to other models will be given in the future.

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