Fluctuation theorems for a molecular refrigerator

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We extend fluctuation theorems to a molecular refrigeration system that consists of Brownian particles in a heat bath under feedback control of their velocities. Such control can actively remove heat from the bath due to an entropy-pumping mechanism [Phys. Rev. Lett. **93**, 120602 (2004)]. The presence of entropy pumping in an underdamped Brownian system modifies both the Jarzynski equality and the fluctuation theorems. We discover that the entropy pumping has a dual role of work and heat.

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I. INTRODUCTION

A molecular refrigerator has been proposed as a class of microscopic machines based on recent developments in nanotechnology and single-molecule manipulations [1–3]. Refrigeration can be achieved by reducing the thermal noise of nanodevices with a feedback system that detects their velocities and applies on them a corresponding frictionlike control force [1]. We have shown that *entropy pumping*, a concept originated by Schrödinger [4], is the fundamental mechanism used for thermal noise reduction in the molecular refrigeration [2]. More recently, Van den Broeck and Kawai proposed a different refrigeration mechanism which consists of two thermal reservoirs with different temperatures in the presence of a constant force on a nanodevice [3]. In this Brief Report, we focus on the former mechanism.

How efficient is such refrigeration? As a first step toward answering this question, we study the underlying thermodynamics and the second law of thermodynamics via the Jarzynski equality and the fluctuation theorems [5–15]. We propose an experiment to measure the free energy difference. We discover that entropy pumping has a dual role of work and heat. First, in its role as work, entropy pumping modifies the Jarzynski equality: one must measure not only the work done on the nanodevices but also the entropy pumping when estimating the equilibrium free energy difference under velocity-dependent feedback control (VFC) [2]. Second, in its role as heat, it changes fluctuation theorems by modifying the entropy production in nanodevices and their surrounding heat bath. Our results suggest a range of applicability of the fluctuation theorems and the Jarzynski equality.

As a model for a molecular refrigerator, we studied a Brownian particle in a heat bath under a *frictionlike* force manipulated by a VFC in [2]. As the system evolves with time, the particle eventually settles in its stationary state, where the average kinetic energy of the Brownian particle (B) is lower than the heat bath (H) since the frictionlike external force reduces the thermal fluctuations of the particle. Kinetic energy is transferred from the heat bath to the particle in the form of heat, $Q_{HB}>0$, and is ultimately absorbed by the external control agent (C) in the form of mechanical energy: $W_{BC}=Q_{HB}>0$. This fact seems to violate the second law, but this is not the case. One needs to treat both the particle and the control agent as one whole system since they are strongly coupled. In the stationary state, the entropy of the heat bath decreases due to entropy pumping by the control agent [2]: $\Delta S_H=\Delta S_p+\Delta S_{pu}<0$, with ΔS_H the entropy change in the heat bath, $\Delta S_p>0$ entropy production, and $\Delta S_{pu}<0$ entropy pumping by the control agent. We note in general that ΔS_p is positive due to the irreversible process of Brownian dynamics implying the second law, while ΔS_{pu} can take any sign.

Refrigeration by drawing heat from the bath is a unique feature of VFC. If the feedback force on the particle depends not on its velocity but on its position [we name such a control position-dependent feedback control (PFC)], the particle acts as a heater rather than a refrigerator since entropy pumping vanishes [2]. The Jarzynski equality and the fluctuation theorems for entropy production and the work functional [5–15] have been studied only in a system with PFC. Thus, a question arises: How are they modified with VFC? We formulate this question by focusing on the Jarzynski equality. The second law is mathematically an inequality: The external work done by the control agent on the particle in contact with a heat bath, $\langle W_{CB}\rangle$, is no smaller than the Helmholtz free energy change of the particle, ΔF. This inequality can be quantified through the Jarzynski equality $\langle e^{-\beta W_{CB}}\rangle=e^{-\beta\Delta F}$ [7–10,13–15], where $\beta\equiv 1/k_BT_H$ with T_H the heat bath temperature. With the presence of VFC, however, the equality needs modification due to entropy pumping. Let us consider energy balance and entropy balance: $\langle W_{CB}\rangle=\langle\Delta H\rangle+\langle Q_{BH}\rangle$ and $\Delta(S_H+S)=\Delta S_p+\Delta S_{pu}$, with ΔH the internal energy change of the system and ΔS the entropy change in the particle. Since the heat bath is in a quasistatic process [16], $\langle W_{CB}\rangle=\langle\Delta H\rangle+T_H\Delta S_H=\langle\Delta H\rangle+T_H(\Delta S_p+\Delta S_{pu}-\Delta S)$. Then, $\langle W_{CB}\rangle>\langle\Delta H\rangle+T_H(\Delta S_{pu}-\Delta S)=\Delta F+T_H\Delta S_{pu}$, where $\Delta F\equiv\langle\Delta H\rangle-T_H\Delta S$ is the free energy change. Finally, we get $\langle W_{CB}\rangle-T_H\Delta S_{pu}>\Delta F$. This implies that, with the presence of VFC, entropy pumping modifies the stochastic work functional in the Jarzynski equality. The second law can be quantitatively described by the fluctuation theorems, which are closely related to the Jarzynski equality. So they also need to be extended.

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II. EXPERIMENTAL SETUP

The extension of fluctuation theorems can be tested by modifying the experiment in [17], as shown in Fig. 1: A ferromagnetic microbead trapped in optical tweezers and a nonmagnetic microbead fixed by a pipette are connected to a polymer. The polymer is introduced only to act as a spring soft enough for the bead to be in Brownian motion but hard enough for the bead's rotation to be confined. Thus, the magnetic moment of the bead is for most of the time pointed to the nonmagnetic bead. The tweezers are shifted horizontally. A coil and a feedback circuit are added to produce a nonuniform magnetic field with the direction of current in the coil actively switched by the feedback circuit that detects the velocity of the bead. The typical range of force for the magnetic tweezers and optical tweezers is 0.01–10 pN and 0.1–100 pN, respectively. Therefore, the simultaneous control of the magnetic microbead by the coil and the tweezers is possible experimentally.

III. I-D BROWNIAN PARTICLES UNDER VFC

Without losing generality, we consider one-dimension Brownian dynamics described by the following Langevin equation:

$$\frac{dv}{dt} = -\frac{\partial H(x,v;\alpha(t))}{\partial x} - \gamma v + g(v) + \xi, \tag{1}$$

with v the velocity of a particle, γ the frictional coefficient, $g(v)$ a general VFC, and ξ Gaussian white noise satisfying $\langle \xi(t)\xi(s)\rangle = \delta(t-s)$. $H(x,v,;\alpha(t))$ is a Hamiltonian changing with a parameter $\alpha(t)$ varying with time: $H(x,v;\alpha(t))=\frac{1}{2}v^2 + U(x;\alpha(t))$. In the above proposed experiment, $g(v)=-\gamma' v$ with a positive constant γ' dependent on the applied magnetic field and $\alpha(t)$ corresponds to the center of the harmonic potential produced by the tweezers. We use unit mass and assume that the Einstein relation $T_H = 1/\gamma$ [18] holds with T_H the heat bath temperature since $g(v)$ is not a frictional force. We used $k_B=1$ unit. The corresponding Fokker-Planck equation becomes $\frac{\partial P(x,v,t)}{\partial t} = \mathcal{L}P(x,v,t)$, where

$$\mathcal{L} \equiv \partial_v^2 - \partial_v[-\{\partial_x H(x,v;\alpha(t))\} - \gamma v + g(v)] - v\partial_x, \tag{2}$$

with $\partial_v \equiv \partial/\partial v$ and $\partial_x \equiv \partial/\partial x$. We note that the Einstein relation is required for the proof of the Jarzynski equality since calculation of the free energy needs an equilibrium distribution, while the relation is not required for the fluctuation theorems (γ is arbitrary).

We first define several terms. An *internal* system is the Brownian particle together with the heat bath. An *external* system is the control agent that manipulates both the control force $g(v)$ and internal potential change due to the change of $\alpha(t)$.

IV. MESOSCOPIC THERMODYNAMICS

We define the mesoscopic heat $dQ(t)$ [2,10,19,20], mesoscopic entropy of the Brownian particle $dS(t)$ [7,10,20], and mesoscopic entropy pumping $dS_{pu}(t)$ (in the Introduction, the

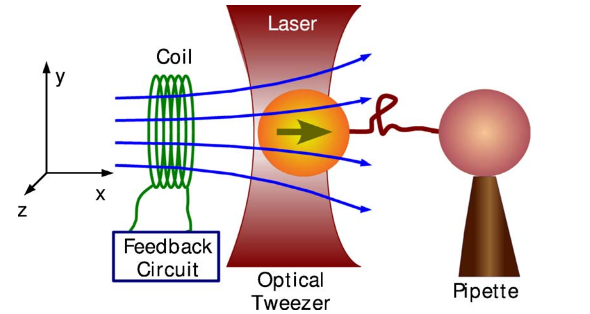

FIG. 1. (Color online) Experimental setup to test the extension of the fluctuation theorems and the Jarzynski equality.

same notations were used, but hereafter S, S_p, and S_{pu} are stochastic quantities) [2]:

$$dQ(t) \equiv -[-\gamma v_t + \xi(t)]dx_t, \tag{3}$$

$$dS(t) \equiv -d\ln P(x_t,v_t,t), \tag{4}$$

$$dS_{pu}(t) \equiv \partial_{v_t} g(v_t)dt. \tag{5}$$

These are all stochastic quantities since (x_t,v_t) has a stochastic trajectory. The entropy change in the heat bath is given due to its isothermal quasistatic nature [16],

$$dS_H(t) = \beta dQ(t),$$

with $\beta = 1/T_H$. The entropy balance, expressed as

$$dS + dS_H = dS_p + dS_{pu}, \tag{6}$$

can be considered as the definition of the entropy production dS_p. The entropy change of the internal system is due to not only entropy production but also entropy pumping. Finally, the energy balance is expressed as

$$dH = \partial_t H dt + \partial_x H dx + \partial_v H dv = \partial_t H dt + g dx - dQ, \tag{7}$$

using Eq. (1). So we define work done on the particle by the control agent

$$dW \equiv \partial_t H dt + g dx. \tag{8}$$

Note that all the above mesoscopic thermodynamic quantities are defined with a Stratonovich prescription, which is known to be physically meaningful [18].

V. NON-EQUILIBRIUM EQUALITIES

The Jarzynski equality and the equality due to the entropy production fluctuation theorem can be derived by examining the temporal behavior of the following quantity [9,10,20]:

$$f(x,v,t) = \left\langle \delta(x-x_t)\delta(v-v_t)\exp\left(\int_{s=0}^{s=t} - dS_H(s) + dS_{pu}(s) + d\ln w(x_s,v_s,s)\right)\right\rangle, \tag{9}$$

where $w(x,v,s)$ is an arbitrary weight function. $\langle\cdots\rangle$ is a path integral averaging over an arbitrary initial distribution $P(x,v,0)$: $\langle\cdots\rangle \equiv \int \lim_{N\to\infty}\Pi_{i=0}^N dx_i dv_i(\cdots)P(N|N-1)P(N-1|N-2)\cdots P(1|0)P(x_0,v_0,0)$, where $P(n|n-1)$ is the transition probability to find a particle at (x_n, v_n) after a time interval $\epsilon \equiv t/N$ given (x_{n-1}, v_{n-1}) as an initial starting point. $-dS_H + dS_{pu}$ can be expressed as $\beta dH - \beta \partial_t H dt - \beta g dx + \partial_v g dt$ using Eqs. (1), (3), and (5). Equation (9) becomes

$$f(x,v,t) = w(x,v,t) e^{\beta H(x,v;\alpha(t))} f_0(x,v,t), \tag{10}$$

where

$$f_0(x,v,t) \equiv \left\langle \frac{\delta(x-x_t)\delta(v-v_t)}{w(x_0,v_0,0)\exp[\beta H(x_0,v_0;\alpha(0))]} \exp\left\{ \int_{s=0}^{s=t} - \beta[\partial_s H(s) ds + g(v_s) dx_s] + \partial_{v_s} g(v_s) ds \right\} \right\rangle.$$

Note that $f_0(x,v,0) = \frac{P(x,v,0)}{w(x,v,0)} e^{-\beta[H(x,v;\alpha(0))]}$. The time derivative of $f_0(x,v,t)$ is expressed as

$$\partial_t f_0(x,v,t) = \mathcal{L} f_0(x,v,t) + f_0(x,v,t)[-\beta \partial_t H - \beta g v + \partial_v g].$$

Its solution becomes $f_0(x,v,t) = e^{-\beta H(x,v;\alpha(t))}$ by requiring $w(x,v,0) = P(x,v,0)$. Therefore, $f(x,v,t) = w(x,v,t)$. By integrating both sides of Eq. (9) over x and v, we get the following general equality [10]:

$$\left\langle \frac{w(x_t,v_t,t)}{P(x_0,v_0,0)} \exp[-\Delta S_H(t) + \Delta S_{pu}(t)] \right\rangle = 1, \tag{11}$$

where $w(x,v,t)$ is an arbitrary weight function with $w(x,v,0) = P(x,v,0)$.

With $w(x,v,t) = \exp[-\beta H(x,v;\alpha(t))]/Z_e(t)$, where $Z_e(t) \equiv \int dx dv \exp[-\beta H(x,v;\alpha(t))]$, Eq. (11) becomes an extended form of the Jarzynski equality:

$$\langle e^{-\beta W(t) + \Delta S_{pu}(t)} \rangle = e^{-\beta \Delta F(t)}, \tag{12}$$

where, using Eq. (8),

$$W(t) \equiv \int_0^t ds \left[\frac{\partial H(x_s,v_s;\alpha(s))}{\partial s} + g(v_s) v_s \right]$$

is the work done on the particle by the external control agent and $\Delta F(t) \equiv -\ln \frac{Z_e(t)}{Z_e(0)}$ is the free energy difference of two equilibrium states parametrized by $\alpha(0)$ and $\alpha(t)$, respectively. Note that the final probability distribution does *not* have to be in an equilibrium state parametrized by $\alpha(t)$, while the initial one does by $\alpha(0)$.

With $w(x,v,t) = P(x,v,t)$, Eq. (11) becomes an extended form of an equality related to the entropy production fluctuation theorem [10],

$$\langle \exp[-\Delta S_H(t) - \Delta S(t) + \Delta S_{pu}(t)] \rangle = \langle \exp[-\Delta S_p(t)] \rangle = 1. \tag{13}$$

Equation (13) shows that $\langle \Delta S_p(t) \rangle$ becomes positive over a finite time interval with or without VFC for an arbitrary initial distribution $P(x,v,0)$. Equation (13) implies that the entropy production fluctuation theorem holds under VFC with a proper definition of S_p, Eq. (6).

With $w(x,v,t) = w(x,v,0)$, we obtain an equality

$$\langle \exp[-\Delta S_H(t) + \Delta S_{pu}(t)] \rangle = 1 \tag{14}$$

over flat initial distribution $[P(x,v,0) = \text{const}]$. Without VFC, the average heat dissipation $\langle \Delta S_H \rangle$ becomes positive over a *finite* time interval.

We note that, for different initial probability distributions, one can get various equalities while an equality related to the entropy production fluctuation theorem is independent of initial probability distributions.

VI. FLUCTUATION THEOREMS

We now extend the entropy production fluctuation theorem

$$\frac{P(\Delta S_p(t) = a)}{P(\Delta S_p(t) = -a)} = \exp[a] \tag{15}$$

using the following path integral relation [7]:

$$\frac{P(\{x_s, v_s\}; \alpha(s))}{P(\{x_{t-s}, -v_{t-s}\}; \alpha(t-s))} = \exp[\Delta S_H(t) - \Delta S_{pu}(t)], \tag{16}$$

where $P(\{x_s, v_s\}; \alpha(s))$ is the probability to find a path $\{x_s, v_s\}$, with $0 \le s \le t$, starting from (x_0, v_0) and ending at (x_t, v_t), and $P(\{x_{t-s}, -v_{t-s}\}; \alpha(t-s))$ is the probability to find a path traced backward. The derivation of Eq. (16) is based on the following conditional probability ratio:

$$\frac{P(x,v|x',v')}{P(x',-v'|x,-v)} = \frac{\langle x,v|e^{\epsilon \mathcal{L}}|x',v'\rangle}{\langle x',-v'|e^{\epsilon \mathcal{L}}|x,-v\rangle},$$

where $\mathcal{L}$ is a Fokker-Planck operator defined as Eq. (2). To make the transition probability into a path integral form, we express $\mathcal{L}$ as a Weyl-ordered form:

$$\mathcal{L}_w(x,v,\hat{p}_x,\hat{p}_v) = -\hat{p}_v^2 - iv\hat{p}_x - \tfrac{1}{2} i\hat{p}_v[F(x,v) - \gamma v] - \tfrac{1}{2}[F(x,v) - \gamma v] i\hat{p}_v - \tfrac{1}{2}\{\partial_v[F(x,v) - \gamma v]\},$$

where $F(x,v;\alpha(t)) \equiv -\partial_x H(x,v;\alpha(t)) + g(v)$, $\hat{p}_x \equiv -i\partial_x$, and $\hat{p}_v \equiv -i\partial_v$. Then, as $\epsilon \to 0$,

$$\begin{aligned} P(x,v|x',v') &= \int dp_x dp_v \exp[\epsilon \mathcal{L}_w(\bar{x},\bar{v},p_x,p_v) + ip_x(x-x') + ip_v(v-v')] \\ &= \frac{\delta(x-x'-\epsilon\bar{v})}{\sqrt{4\pi\epsilon}} \exp\left[-\epsilon\left(\frac{\bar{F}-\gamma\bar{v}}{2} - \frac{v-v'}{2\epsilon} \right)^2 - \frac{\epsilon}{2}[\partial_{\bar{v}}(\bar{F}-\gamma\bar{v})] \right] \end{aligned}$$

and

$$P(x',-v'|x,-v) = \frac{\delta(x-x'-\epsilon\bar{v})}{\sqrt{4\pi\epsilon}} \times \exp\left[-\epsilon\left(\frac{\bar{F}+\gamma\bar{v}}{2} - \frac{v-v'}{2\epsilon} \right)^2 + \frac{\epsilon}{2}[\partial_{\bar{v}}(\bar{F}+\gamma\bar{v})] \right],$$

where $\bar{x} \equiv (x+x')/2$, $\bar{v} \equiv (v+v')/2$, and $\bar{F} \equiv F(\bar{x},\bar{v})$. Therefore,

$$\frac{P(x,v|x',v')}{P(x',-v'|x,-v)} = \exp\left[\epsilon\left(\bar{F} - \frac{v-v'}{\epsilon}\right)\gamma\bar{v} - \epsilon\partial_{\bar{v}}\bar{F}\right], = \exp[dS_H - dS_{pu}]$$

With a time-independent Hamiltonian, the work fluctuation theorem has been obtained [11,12]. Like the entropy production fluctuation theorem, the work fluctuation theorem is extended as follows. From energy balance, $\Delta S_p(t) = -\beta\Delta H(t) + \beta W(t) + \Delta S(t) - \Delta S_{pu}(t)$, where $W(t) = \int_0^t dx_s g(v_s)$ in the time-independent Hamiltonian case. $\Delta S(t)$ does not increase with sufficiently large time t on average since $\Delta S(t) = -\ln P_{ss}(x_t, v_t) + \ln P(x_0, v_0, 0)$ with P_{ss} a stationary distribution, while $W(t)$ and ΔS_{pu} monotonically change on average with the large time t. Therefore, for $t\to\infty$, $\Delta S_p(t) \to \beta W(t) - \Delta S_{pu}(t)$ and the extended work fluctuation theorem holds:

$$\frac{P(\beta W(t) - \Delta S_{pu}(t) = a)}{P(\beta W(t) - \Delta S_{pu}(t) = -a)} = \exp[a]. \tag{17}$$

The corresponding equality is derived,

$$\lim_{t\to\infty}\langle\exp[-\beta W(t) + \Delta S_{pu}(t)]\rangle = 1. \tag{18}$$

From Eq. (18), we find that $\langle W(t) - T_H\Delta S_{pu}(t)\rangle > 0$ as $t\to\infty$ with a time-independent Hamiltonian.

VII. MODIFIED WORK AND HEAT

In [2], we have found that entropy pumping is related to momentum phase-space contraction due to $g(v)$ $[dS_{pu}/dt = \partial_v g(v)]$. The extended fluctuation theorems derived above show a role of entropy pumping: *A dual role of work and heat*. The work functional W in the Jarzynski equality and the work fluctuation theorem are modified to $W' \equiv W - T_H\Delta S_{pu}$ as shown in Eqs. (12) and (17). Entropy of the heat bath S_H is modified to $S_H - S_{pu}$ as shown in Eqs. (11), (13), and (14). Thus, we define modified heat $Q' \equiv Q - T_H\Delta S_{pu}$. The reason for the duality is easy to understand from the energy balance, Eq. (7): $\Delta H = W - Q$. When Q is modified, W also needs to be modified with ΔH unchanged. We note that $\langle Q'\rangle > 0$ in the stationary state under a general VFC.

For definiteness, let us consider and compare three examples with $U(x;\alpha(t)) = 0$: (i) without VFC but only with PFC in two- or higher-dimensional systems [$g(v)$ in Eq. (1) is replaced with a nonconservative force $\vec{g}(\vec{x})$], (ii) with frictionlike VFC, $g(v) = -cv$, with $c > 0$, and (iii) with nonfrictionlike VFC with $c < 0$. Let the system be in a stationary state. In case (i), $\Delta S_{pu} = 0$ and $\langle W_{CB}\rangle > 0$ from Eq. (18), so $\langle Q_{BH}\rangle > 0$. In case (ii), $\langle W_{CB}\rangle < 0$ and $\langle Q_{BH}\rangle < 0$ [2]. However, the modified work and heat have opposite signs: $\langle W'_{CB}\rangle > 0$ and $\langle Q'_{BH}\rangle > 0$, where $\langle\Delta S_{pu}\rangle = -c\Delta t < 0$ [2]. With the modified work W' and heat Q', case (ii) becomes case (i). In case (iii), $\langle W_{CB}\rangle > 0$ and $\langle Q_{BH}\rangle > 0$. With entropy pumping modification, $\langle W'_{CB}\rangle > 0$ and $\langle Q'_{BH}\rangle > 0$.

VIII. CONCLUSIONS AND REMARKS

Nanoscale mesoscopic systems with a VFC are significantly different from the widely studied overdamped stochastic systems with a PFC [21]. The key difference is that the former involves an active entropy reduction mechanism leading to a molecular refrigeration. In this Brief Report, we showed that entropy pumping modifies the Jarzynski equality and fluctuation theorems. This modification is due to an analysis of the *subsystem*. There are other classical and quantum subsystems not governed by VFC. It will be interesting to see whether the Jarzynski equality and the fluctuation theorems can be extended to all subsystems in general, and if not, what the criteria of such an extension are. Just as a molecular motor has greatly advanced our knowledge of an overdamped small system, the molecular refrigerator will advance our knowledge of an underdamped small system.

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