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Generalized Jarzynski's Equality of Inhomogeneous Multidimensional Diffusion Processes

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Abstract Applying the well-known Feynman-Kac formula of inhomogeneous case, an interesting and rigorous mathematical proof of generalized Jarzynski's equality of inhomogeneous multidimensional diffusion processes is presented, followed by an extension of the second law of thermodynamics. Then, we explain its physical meaning and applications, extending Hummer and Szabo's work (Proc. Natl. Acad. Sci. USA 98(7):3658–3661, 2001) and Hatano-Sasa equality of steady state thermodynamics (Phys. Rev. Lett. 86:3463–3466, 2001) to the general multidimensional case.

Keywords Feynman-Kac formula · Jarzynski's equality · Inhomogeneous diffusion processes · Nonequilibrium thermodynamics · Hatano-Sasa equality

1 Introduction

Thermodynamics of irreversible systems far from equilibrium has been developed for more than thirty years since the original works by Haken [12, 13] about laser and Prigogine et al. [11, 30] about oscillations of chemical reactions. A nonequilibrium system can be regarded as an open system with positive entropy production, which means exchange of substances and energy with its environment.

At almost the same time, T.L. Hill, etc. [16–19] constructed a general mesoscopic model for the combination and transformation of biochemical polymers in vivid metabolic systems since 1966, which can be applied to explain the mechanism of muscle contraction and active transports, such as the Na and K ions actively transferring and penetrating through organic membranes in the Hodgkin-Huxley model. These contributions have been developed and summarized in [26].

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One can use stationary homogeneous Markov chains and diffusion processes as mathematical tools to model nonequilibrium steady states and cycle fluxes. Mathematical theory of nonequilibrium steady states has been developed for more than three decades since Qians' original works [34–37]. They derived the formulae for entropy production rate and circulation distribution of homogeneous Markov chains, Q-processes and diffusions. They concluded that the chain or process is reversible if and only if its entropy production rate vanishes, or iff there is no net cycle fluxes. Here, we recommend a recent book [25] for the systematic presentation of this theory.

In the past decade, a few relations that describe the statistical dynamics of driven systems have been discovered, which are valid even if the system is driven far from equilibrium. These include Jarzynski's exciting nonequilibrium work relation [2–4, 21–23], which gives equilibrium Helmholtz free energy differences in terms of nonequilibrium measurements of the work required to switch from one ensemble to another. This result has been applied to the mechanical extension of single RNA molecules in the laboratory [29]. Although the concept of Helmholtz free energy fails in nonequilibrium steady states (NESS), Hatano and Sasa [15] have generalized Jarzynski's work to the NESS described by a simple one-dimensional Langevin system, which is more relevant to motor proteins.

However, few rigorous mathematical results are derived since the emergence of Jarzynski's equality. Applying the Feynman-Kac formula, G. Hummer and A. Szabo gave a quite brief proof of Jarzynski's equality for inhomogeneous diffusive dynamics on a potential [20], and after that, Hong Qian investigated a simple two-state example of inhomogeneous Markov chains [33]. But in fact, their proofs are not mathematically rigorous, and they all misused the Feynman-Kac formula of the inhomogeneous case [27, Theorem 5.7.6], since it is quite different from the Feynman-Kac formula of the homogeneous case [27, Theorem 4.4.2] and the former is actually more difficult to apply than the latter one. Further explanation is included in Sect. 3.1. On the other hand, the Jarzynski's equality is trivial in the homogeneous case (see Remark 2.10 below), which actually implies that inhomogeneity is a necessity for Jarzynski's equality to make sense.

Recently enlightened by the work of Crooks [4], we gave a completely different and interesting rigorous derivation of the Jarzynski's equality in inhomogeneous Markov chains [8], without applying the Feynman-Kac formula. Moreover, we investigated the relationship between Jarzynski's equality and the statistical physical property in the model of inhomogeneous Markov chains, including reversibility and entropy production [9, 10].

Nevertheless, it is not easy to extend the main idea of proof in [8] to the case of inhomogeneous diffusion processes, because by this way, one needs a very general version of the Cameron-Martin-Girsanov formula similar to [39, Theorem 6.4.2], which is difficult to derive. On the other hand, physicists always believe that inhomogeneous diffusion processes can be regarded as the limit of inhomogeneous Markov chains, and in most of their works, they actually only proved the corresponding results in the case of inhomogeneous Markov chains rather than diffusion processes. However, from the mathematical point of view, inhomogeneous diffusion processes can only be regarded as the limit of inhomogeneous Markov chains *in distribution* rather than *in trajectories*. Hence, the Jarzynski's equality in inhomogeneous diffusion processes can not be directly derived as the limit in some sense of that in inhomogeneous Markov chains, and we have to appeal to the Feynman-Kac formula of inhomogeneous case.

In this paper, applying the well-known Feynman-Kac formula of inhomogeneous case, an interesting and rigorous mathematical proof of generalized Jarzynski's equality in inhomogeneous multidimensional diffusion processes is presented in Sect. 2, followed by an extension of the second law of thermodynamics. It should be mentioned that the method of proof in the present paper can also be applied to derive the same Jarzynski's equality in inhomogeneous Markov chains as [8], or even possibly to extend to general Markov processes. In Sect. 3, we explain its physical meaning and applications, extending Hummer and Szabo's work [20] and Hatano-Sasa equality of steady state thermodynamics [15] to the general multidimensional case.

In order to make the present paper accessible to a somewhat wider audience, some reasonable sufficient conditions for Jarzynski's equality of inhomogeneous diffusion processes are provided in Remark 2.7 below.

2 Mathematical Theory of Generalized Jarzynski's Equality

2.1 Basic Property of Inhomogeneous Diffusion Processes

This subsection is about the construction of inhomogeneous diffusion processes applying the fundamental solutions of partial differential equations. The conditions given here are somewhat optimal, and the readers who are not interested in technical details can directly skip to the next subsection for the proof of generalized Jarzynski's equality. We note here that most of these conditions including (A1), (A2) and (A4) are satisfied when all the coefficients belong to the smooth function set C^∞ and all the derivatives are uniformly bounded.

Denote $A_t(x) = (a_{ij}(t,x))_{d\times d}$ and $\bar{b}_t(x) = (\bar{b}_i(t,x))_{d\times 1}$, where $a_{ij}(t,x)$ and $\bar{b}_i(t,x)$ are functions defined on $[0,+\infty)\times\mathbb{R}^d$. Suppose that

(A1) $a_{ij}(t,x), \bar{b}_i(t,x)$ are uniformly bounded and uniformly continuous with respect to both x and t, and also satisfy a Hölder condition with respect to x;
(A2) $a_{ij}(t,x)$ satisfy a Hölder condition with respect to t;
(A3) $a_{ij}(t,x)$ satisfy the uniform ellipticity condition, i.e. there exists $\gamma > 0$, such that for any d-dimensional real vector $\lambda = (\lambda_1, \lambda_2, \dots, \lambda_d)$,

$$\sum_{i,j=1}^{d} a_{ij}(t,x)\lambda_i\lambda_j \geq \gamma \sum_{i=1}^{d} \lambda_i^2;$$

(A4) The derivatives $\frac{\partial a_{ij}}{\partial x_i}, \frac{\partial^2 a_{ij}}{\partial x_i \partial x_j}, \frac{\partial \bar{b}_i}{\partial x_i}$ exist, uniformly bounded and satisfy a Hölder condition with respect to x.

For simplicity, let $b_t = (b_i(t,x))_{d\times 1}$ and $b_i(t,x) = \bar{b}_i(t,x) - \frac{1}{2}\sum_{j=1}^{d} \frac{\partial a_{ij}(t,x)}{\partial x_j}$.

Theorem 2.1 below is rewritten from [6, Vol. II, Theorem 0.4, p. 227] and [7, Chap. 1, Sect. 6, Theorem 11, p. 24; Chap. 1, Sect. 8, Theorem 15], and Theorem 2.3 is rewritten from [6, Vol. I, remark of Theorem 5.11, p. 167] and [5, Chap. 4]. One can also find the same results in [27, pp. 368–369] and [39, Chap. 3].

Theorem 2.1 *If the coefficients satisfy conditions* (A1), (A2), (A3), *then the equation*

$$\frac{\partial u}{\partial s} + D_s u = 0, \tag{1}$$

where $D_s u(s,x) = \frac{1}{2}\sum_{i,j=1}^{d} \frac{\partial}{\partial x_i} a_{ij}(s,x) \frac{\partial u}{\partial x_j} + \sum_{i=1}^{d} b_i(s,x)\frac{\partial u}{\partial x_i} = (\frac{1}{2}\nabla\cdot A(s,x)\nabla + b(s,x)\cdot\nabla)u$, *has a unique fundamental solution* $p(s,t;x,y)$, *satisfying*:

(B1) $p(s,t;x,y) > 0$ *for each* s, t *and* x, y;

(B2) *In addition, if coefficients* $a_{ij}(t,x), \bar{b}_i(t,x)$ *satisfy* (A4), *then* $p(s,t;x,y)$ *satisfies the conjugate equation*:

$$\frac{\partial u}{\partial t} = \bar{D}_t^* u, \tag{2}$$

where $\bar{D}_t^* u(t,y) = \sum_{i=1}^d \frac{\partial}{\partial y_i}[\frac{1}{2}\sum_{j=1}^d a_{ij}(t,y)\frac{\partial u(t,y)}{\partial y_j} - b_i(t,y)u(t,y)] = \nabla\cdot[\frac{1}{2}A(t,y)\times \nabla u(t,y) - b(t,y)u(t,y)]$;

(B3) *For any bounded continuous function* $f(x)$, $u(s,t,x) = \int p(s,t;x,y)f(y)dy$ *satisfies* (1) *and* $\lim_{s\uparrow t} u(s,t,x) = f(x)$, *which is uniformly convergent in any bounded domain of* $\mathbb{R}^d$; $v(s,t,y) = \int p(s,t;x,y)f(x)dx$ *satisfies* (2), *and* $\lim_{t\downarrow s} v(s,t,y) = f(y)$, *which is also uniformly convergent in any bounded domain of* $\mathbb{R}^d$;

(B4) *The following inequalities are satisfied*:

$$p(s,t;x,y) \le M(t-s)^{-\frac{d}{2}} e^{-\frac{\alpha|y-x|^2}{t-s}};$$
$$\frac{\partial p(s,t;x,y)}{\partial x_i} \le M(t-s)^{-\frac{d+1}{2}} e^{-\frac{\alpha|y-x|^2}{t-s}};$$
$$\frac{\partial^2 p(s,t;x,y)}{\partial x_i \partial x_j} \le M(t-s)^{-\frac{d}{2}-1} e^{-\frac{\alpha|y-x|^2}{t-s}};$$
$$\frac{\partial p(s,t;x,y)}{\partial t} \le M(t-s)^{-\frac{d}{2}-1} e^{-\frac{\alpha|y-x|^2}{t-s}};$$
$$p(s,t;x,y) \ge M_1(t-s)^{-\frac{d}{2}} e^{-\frac{\alpha_1|y-x|^2}{t-s}} - M_2(t-s)^{-\frac{d}{2}+\lambda} e^{-\frac{\alpha_2|y-x|^2}{t-s}},$$

where M, M_1, M_2 *and* $\alpha, \alpha_1, \alpha_2$ *are all positive constants.*

(B5) *If* $f(x)$ *is a bounded function with second-order continuous derivatives, which satisfy a Hölder condition, then* $u(s,t,x) = \int p(s,t;x,y)f(y)dy$ *satisfies*

$$\lim_{s\uparrow t} \frac{\partial u}{\partial x_i} = \frac{\partial f}{\partial x_i}, \qquad \lim_{s\uparrow t} \frac{\partial^2 u}{\partial x_i \partial x_j} = \frac{\partial^2 f}{\partial x_i \partial x_j};$$

while $v(s,t,y) = \int p(s,t;x,y)f(x)dx$ *satisfies*

$$\lim_{t\downarrow s} \frac{\partial v}{\partial y_i} = \frac{\partial f}{\partial y_i}, \qquad \lim_{t\downarrow s} \frac{\partial^2 v}{\partial y_i \partial y_j} = \frac{\partial^2 f}{\partial y_i \partial y_j}.$$

Remark 2.2 Most of the following definitions of physical quantities make sense due to the basic inequalities in (B4) together with (B3) and (B5).

Theorem 2.3 *There exists a unique inhomogeneous diffusion process* $X = \{X_t : t \ge 0\}$ *on* $\mathbb{R}^d$, *whose transition probability density is* $\{p(s,t;x,y)\}$. *Moreover, X is a strong Markov process. We call X the diffusion process with infinitesimal generator D.*

Equation (1) is called the backward Kolmogorov equation of X, while (2) is called the forward Kolmogorov equation of X.

Denote the initial distribution density of X as $\{\rho_0(x) > 0 : x \in \mathbb{R}^d\}$, which is at least twice differentiable, then $\rho_t(x) = \int \rho_0(y)p(0,t;y,x)dy$ is the density function of X_t, simply denoted as ρ_t. Thus from (B3), $\rho_t(x)$ satisfies (2), which is called the Fokker-Planck equation.

Indeed, due to the condition (A3), there exists a nonsingular $d \times d$ matrix $\Gamma_t(x) = (\Gamma_{ij}(t,x))_{d\times d}$ such that $A_t(x) = \Gamma_t(x)\Gamma_t^T(x)$, where $\Gamma_t^T(x)$ is the transpose matrix of $\Gamma_t(x)$. The inhomogeneous multidimensional diffusion process $\{X_t : t \geq 0\}$ can be considered as the unique solution of the stochastic differential equation

$$dX_t = \bar{b}_t(X_t)dt + \Gamma_t(X_t)dW_t,$$

where $\{W_t\}_{t\geq 0}$ is a d-dimensional Wiener process.

2.2 Rigorous Proof of Generalized Jarzynski's Equality

Fix the time interval as $[0, T]$. In order to make the quantities in Jarzynski's equality below mathematically well-defined, we have to make another basic assumption:

(A5) The elliptic equation $\bar{D}_t^* f(x) = 0$ has a unique strong L^1 solution $\pi_t = \{\pi_t(x) : x \in \mathbb{R}^d\}$ such that $\int_{\mathbb{R}^d} \pi_t(x)dx \equiv 1$, recalling $\bar{D}_t^*\pi_t(x) = \sum_{i=1}^d \frac{\partial}{\partial x_i}[\frac{1}{2}\sum_{j=1}^d a_{ij}(t,x)\frac{\partial \pi_t(x)}{\partial x_j} - b_i(t,x)\pi_t(x)] = \nabla\cdot[\frac{1}{2}A(t,x)\nabla\pi_t(x) - b(t,x)\pi_t(x)]$. Moreover, $\pi_t(x)$ is continuously differentiable and uniformly bounded with respect to parameter $t \in [0,T]$; $\frac{\partial[\pi_t(x)]}{\partial t}$ is uniformly bounded too for $t \in [0,T]$. In addition, suppose $\pi_t(x) > 0$, $\forall x \in \mathbb{R}^d$, $t \geq 0$.

Remark 2.4 Consider a homogeneous diffusion process with infinitesimal generator $D = \frac{1}{2}\nabla \cdot A(x)\nabla + b(x)\cdot\nabla$. In the uniformly elliptic case, if $\bar{D}^*\pi(x) = 0$ has a positive strong L^1 solution such that $\int_{R^d} \pi(x)dx = 1$, it is unique. To have a solution one has to impose a sufficiently strong inward drift at infinity, or equivalently suppose that the homogeneous diffusion process is positive recurrent [14, Chap. IV].

Physicists are interested in some reasonable sufficient conditions. A particular example is the equilibrium case discussed in the next section. More generally, it is sufficient to suppose that the diffusion coefficient $A(x)$ is bounded and uniformly elliptic, $b(x)$ is bounded smooth, and for large x, it is the minus gradient of a confining potential $U(x)$, which is satisfied for many physical problems. The proof can be based on the Lyapunov function criteria for asymptotic stability of stochastic dynamic systems ([28, Theorem 11.9.1] and [14, Theorem III.5.1]):

If there exists a smooth function $V(x)$ with the properties

$$V(x) \geq 0$$

and

$$\lim_{R\to\infty} \sup_{|x|>R} DV(x) = -\infty,$$

then there exists a stationary distribution for the diffusion process.

Therefore, if the potential $U(x)$ satisfies that

$$\begin{aligned} &\sup_{|x|>R}\left[\frac{1}{2}\nabla\cdot A(x)\nabla U(x) + b(x)\cdot\nabla U(x)\right] \\ &= \sup_{|x|>R}\left[\frac{1}{2}\nabla\cdot A(x)\nabla U(x) - \|\nabla U(x)\|_2^2\right] \\ &\to -\infty \quad \text{as } R\to\infty, \end{aligned} \tag{3}$$

where $\|\vec{v}\|_2^2 = \sum_{i=1}^d v_i^2$, then $\bar{D}^*\pi(x) = 0$ has a unique positive strong L^1 solution such that $\int_{R^d} \pi(x)dx = 1$. For example, the sufficient condition (3) is satisfied when the potential $U(x)$ is a polynomial with even highest order.

Another nontrivial example is the multidimensional Ornstein-Ulenbeck process with drift coefficients $Bx = (b_{ij})_{d\times d} \cdot x$ and diffusion coefficients $\sigma = \{\sigma_{ij}\}_{d\times r}$. Its corresponding Fokker-Planck equation is

$$\frac{\partial u}{\partial t} = \frac{1}{2}\sum_{i,j=1}^d a_{ij}\frac{\partial^2 u}{\partial x_i \partial x_j} - \sum_{i=1}^d \frac{\partial}{\partial x_i}[b_i(x)u],$$

where $b_i(x) = \sum_{j=1}^d b_{ij}x_j$ and $a_{ij} = \sum_{k=1}^r \sigma_{ik}\sigma_{jk}$. It is well known that its unique stationary distribution is a multidimensional normal distribution with mean zero and variance $\Sigma = \int_0^{+\infty} e^{Bs}Ae^{B^T s}ds$, provided that the matrix $A = (a_{ij})$ is nonsingular and the real parts of all eigenvalues of $B = (b_{ij})_{d\times d}$ are negative.

According to [28, Example 11.9.2], the Ornstein-Ulenbeck semigroup determined by the above Fokker-Planck equation is asymptotically stable with limiting density of $N(0, \Sigma)$. Moreover, according to [25, Theorem 3.3.7], the stationary multidimensional Ornstein-Ulenbeck process is reversible (or say, in equilibrium state) if and only if the force $F = 2A^{-1}Bx$ is the gradient of a potential $U(x)$ satisfying $\int e^{U(x)}dx = 1$, of iff the coefficient A and B satisfy the symmetry condition $A^{-1}B = (A^{-1}B)^T$ [32]. Therefore, nonequilibrium Ornstein-Ulenbeck processes can only exist in the multidimensional ($d \geq 2$) case.

One could also require that the diffusion process is confined to some compact set or manifold (e.g. torus), and the same arguments below can also be applied to prove the Jarzynski equality in those cases without any difficulties.

π_t can be called the *quasi-invariant distribution*[1] at time t.

Fix the initial distribution density $\rho_0 = \pi_0$, which is one of the key points in Jarzynski's equality. Given an arbitrary absolutely continuous function $F(t)$, denote

$$H(t,x) = F(t) - \beta^{-1}\log \pi_t(x), \quad \forall x \in \mathbb{R}^d. \tag{4}$$

Then

$$\pi_t(x) = \frac{e^{-\beta H(t,x)}}{\int_{\mathbb{R}^d} e^{-\beta H(t,x)}dx},$$

and

$$F(t) = -\beta^{-1}\ln \int_{\mathbb{R}^d} e^{-\beta H(t,x)}dx,$$

where $\beta > 0$ is a constant. Let

$$\begin{aligned} W(\omega) &= \int_0^T \frac{\partial H}{\partial s}(s, X_s)ds = \int_0^T \frac{\partial[F(s) - \frac{1}{\beta}\log \pi_s(X_s)]}{\partial s}ds \\ &= \triangle F - \int_0^T \frac{\partial[\frac{1}{\beta}\log \pi_s](X_s)}{\partial s}ds, \end{aligned} \tag{5}$$

[1]The notion "quasi-invariant distribution" means that if one takes $A(t,x)$ and $\bar{b}(t,x)$ as the diffusion coefficient and drift coefficient of a homogeneous diffusion process respectively, $\pi_t(x)$ is just its invariant distribution.

where $\triangle F = F(T) - F(0)$. Write

$$\triangle H(\omega) = H(T, X_T) - H(0, X_0), \tag{6}$$

and

$$Q(\omega) = \triangle H(\omega) - W(\omega). \tag{7}$$

The following theorem is the basis of our proof, which is an application of the famous Feynman-Kac formula in inhomogeneous case [27, Theorem 5.7.6].

Theorem 2.5 *Under the preceding assumptions, let* $W_d = W - \triangle F = -\int_0^T \frac{\partial[\frac{1}{\beta}\log \pi_s]}{\partial s}(X_s)ds$, *and denote*

$$v(t,x) = E_{t,x} \exp\left[\int_t^T \frac{\partial[\log \pi_s]}{\partial s}(X_s)ds\right], \tag{8}$$

where $E_{t,x}$ *means the expectation is taken conditioned on the event* $\{X_t = x\}$, *then* $v(t,x)$ *satisfies the Cauchy problem*

$$\begin{cases} \frac{\partial v}{\partial t}(t,x) = -\frac{\partial[\log \pi_t(x)]}{\partial t} v(t,x) - D_t v(t,x), \\ v(T,x) = 1, \end{cases} \tag{9}$$

recalling that $D_t v(t,x) = (\frac{1}{2}\nabla \cdot A(t,x)\nabla + b(t,x)\cdot \nabla)v(t,x)$. *Moreover, such a solution is unique.*

Proof We only need to check the condition of Theorem 5.7.6 in [27].

First, according to [6, Vol. II, Theorem 0.4, p. 227], the Cauchy problem (9) has a solution $v(t,x)$ which is continuous and satisfies the exponential growth condition

$$\max_{0\le t\le T} |v(t,x)| \le M e^{\mu \|x\|^2}, \ \ x \in \mathbb{R}^d,$$

for some constants $M > 0$ and $\mu > 0$.

Moreover, due to the condition (A1), the coefficients $a_{ij}(t,x)$ and $\bar{b}_i(t,x)$ are uniformly bounded with respect to t and x.

Finally, applying [27, Theorem 5.7.6 and Problem 5.7.7], together with Theorem 2.1 and Theorem 2.3, we get the desired result. □

Now, it is time to derive the generalized Jarzynski's equality of multidimensional diffusion processes.

Theorem 2.6 *Under the preceding assumptions, suppose that*

(i) $\int \pi_t(x)v(t,x)dx < +\infty$ *for* $t \in [0,T]$, *and* $\int \frac{\partial[\pi_t(x)v(t,x)]}{\partial t}dx$ *are uniformly convergent for* $t \in [0,T]$; *and*
(ii) *for each* i *and* j,

$$\lim_{x\to\infty} \pi_t(x)b_i(t,x)v(t,x) = 0,$$

$$\lim_{x\to\infty} \pi_t(x)a_{ij}(t,x)\frac{\partial v(t,x)}{\partial x_j} = 0,$$

$$\lim_{x\to\infty} \frac{\partial \pi_t(x)}{\partial x_i} a_{ij}(t,x)v(t,x) = 0;$$

then we have

$$E^{P_{[0,T]}}[e^{-\beta W_d}] = 1, \tag{10}$$

i.e.

$$E^{P_{[0,T]}}[e^{-\beta W}] = e^{-\beta \triangle F}.$$

Proof Let $g(t) = \int \pi_t(x)v(t,x)dx$, our aim is to show that $g(t) \equiv 1$, $\forall t \in [0,T]$.

Firstly, due to the assumption (i), it holds that

$$\frac{dg(t)}{dt} = \int \frac{\partial[\pi_t(x)v(t,x)]}{\partial t}dx = \int \left[\frac{\partial \pi_t(x)}{\partial t}v(t,x) + \pi_t(x)\frac{\partial v}{\partial t}(t,x)\right]dx,$$

according to [43, Vol. III, Sect. 21.3, Theorem 4, p. 395]. And from Theorem 2.5, follows that

$$\begin{aligned}
&\frac{\partial \pi_t(x)}{\partial t}v(t,x) + \pi_t(x)\frac{\partial v}{\partial t}(t,x)dx \\
&= \frac{\partial \pi_t(x)}{\partial t}v(t,x) + \pi_t(x)\left[-\frac{\partial[\log \pi_t(x)]}{\partial t}v(t,x) - D_t v(t,x)\right] \\
&= -\pi_t(x)D_t v(t,x).
\end{aligned}$$

Then, with the assumption (ii), integrating by parts, one has

$$\frac{dg(t)}{dt} = \int [-\pi_t(x)D_t v(t,x)]dx = \int [-v(t,x)\bar{D}_t^* \pi_t(x)]dx = 0, \quad \forall t \geq 0,$$

which together with the fact that $g(T) = 1$ implies $g(t) \equiv 1$.

Therefore, $g(0) = E^{P_{[0,T]}}[e^{-\beta W_d}] = 1$. □

Remark 2.7 (Reasonable sufficient conditions) Some physicists may think that the conditions given in this paper are more general than ever needed, and they would be satisfied with reasonable sufficient conditions for the validity of the Jarzynski equality. As we have mentioned at the beginning of this section, it is sufficient to assume (a) all the coefficients are bounded, at least twice continuously differentiable and all the derivatives are uniformly bounded too; (b) the diffusion coefficients $A_t(x)$ are uniformly elliptic (see (A3)); and (c) the existence of quasi-invariant distributions (see (A5) and Remark 2.4). However, the technical requirements (i) and (ii) in Theorem 2.6 are still not easy to be verified, because we do not have exact estimation on the quantity $v(t,x)$. But we believe that the conditions (a), (b) and (c) should be able to guarantee the technical requirements (i) and (ii). Especially, in the equilibrium case (see Sect. 3.1), it would be sufficient to suppose that $H(t,x) \to \infty$ and $|\frac{\partial H(t,x)}{\partial x_i}| \to \infty$ as $x \to \infty$, and $x_i \cdot \frac{\partial H(t,x)}{\partial x_i} > 0$ for all i and sufficiently large x.

In addition, one can explicitly calculate the quantity $v(t,x)$ in some very special case. For example, consider the one-dimensional diffusion dynamics

$$dX_t = -(X_t - t)dt + \sigma dW_t$$

on a moving potential $U(t,x) = \frac{1}{2}(x-t)^2$, which is investigated by van Zon and Cohen [40–42] and mentioned in [1]. Its quasi-invariant distribution $\pi_t(x) = \frac{1}{\sqrt{\pi}\sigma}e^{-\frac{(x-t)^2}{\sigma^2}}$. Consequently, $H(t,x)=\frac{(x-t)^2}{\sigma^2}$, the Helmholtz free energy $F(t)\equiv -\log\sqrt{\pi}\sigma$, and

$$W_d = W = \int_0^T \frac{\partial H}{\partial s}(s, X_s)ds = -\frac{2}{\sigma^2}\int_0^T (X_s - s)ds.$$

The unique solution of this stochastic differential equation can be expressed as

$$X_s = e^{t-s}X_t + \left(s + e^{t-s} - te^{t-s} - 1\right) + \sigma e^{-s}\int_t^s e^u dW_u, \quad \forall s \geq t \geq 0.$$

Let

$$\begin{aligned} Y_t^T &= -\frac{2}{\sigma^2}\int_t^T (X_s - s)ds \\ &= -\frac{2}{\sigma^2}\int_t^T \left(e^{t-s}X_t + e^{t-s} - te^{t-s} - 1 + \sigma e^{-s}\int_t^s e^u dW_u\right)ds. \end{aligned}$$

Because the process $\{X_t : t \geq 0\}$ is Gaussian, one only needs to calculate the expectation and variance of the quantity Y_t^T:

$$\begin{aligned} E_{t,x}Y_t^T &= -\frac{2}{\sigma^2}\int_t^T \left(e^{t-s}x + e^{t-s} - te^{t-s} - 1\right)ds \\ &= -\frac{2}{\sigma^2}\left[(x+1-t)\left(1-e^{t-T}\right) - T + t\right], \end{aligned}$$

and

$$\begin{aligned} \mathrm{Var}_{t,x}Y_t^T &= \mathrm{Var}_{t,x}\left[\frac{2}{\sigma}\int_t^T e^{-s}\int_t^s e^u dW_u ds\right] \\ &= \mathrm{Var}_{t,x}\left[\frac{2}{\sigma}\int_t^T e^u dW_u \int_u^T e^{-s}ds\right] \\ &= \frac{4}{\sigma^2}\int_t^T \left(\int_u^T e^{u-s}ds\right)^2 du \\ &= \frac{4}{\sigma^2}\left[T - t - 2\left(1-e^{t-T}\right) + \frac{1}{2}\left(1-e^{2t-2T}\right)\right]. \end{aligned}$$

Therefore,

$$\begin{aligned} v(t,x) = E_{t,x}e^{-Y_t^T} &= \exp\left[-E_{t,x}Y_t^T + \frac{\mathrm{Var}_{t,x}Y_t^T}{2}\right] \\ &= \exp\left\{\frac{2}{\sigma^2}\left[x\left(1-e^{t-T}\right) - \frac{1}{2} - t + (t+1)e^{t-T} - \frac{1}{2}e^{2t-2T}\right]\right\}. \end{aligned}$$

Finally, it is easy to check that the process $\{X_t : t \geq 0\}$ satisfies the requirements (i) and (ii) in Theorem 2.6 and the quantity

$$\begin{aligned} g(t) &= \int \pi_t(x)v(t,x)dx \\ &= \frac{1}{\sqrt{\pi}\sigma}\int \exp\left[-\frac{(x-t-1+e^{t-T})^2}{\sigma^2}\right]dx \equiv 1. \end{aligned}$$

Definition 2.8 We call the inhomogeneous multidimensional diffusion process $X = \{X(t)\}_{0\le t\le T}$ is *unperturbed*, if π_t, $0 \le t \le T$, are all the same.

The following is an extension of the second law of thermodynamics.

Theorem 2.9 $E^{P_{[0,T]}} W_d \ge 0$, *i.e.*

$$E^{P_{[0,T]}} W \ge \triangle F. \tag{11}$$

Moreover, $E^{P_{[0,T]}} W = \triangle F$ *if and only if*

$$\int_0^T \frac{\partial \log \pi_s}{\partial s}(X_s(\omega))ds = 0, \ \ P_{[0,T]}\text{-}a.s.$$

If the inhomogeneous multidimensional diffusion process $X = \{X(t)\}_{0\le t\le T}$ *is unperturbed, then* $W(\omega) = \triangle F$ *for each trajectory* ω.

Proof By the Jazynski's equality (10) and Jensen's inequality,

$$e^{-\beta\triangle F} = E^{P_{[0,T]}}[e^{-\beta W}] \ge e^{-\beta E^{P_{[0,T]}} W},$$

i.e.

$$E^{P_{[0,T]}} W \ge \triangle F,$$

and the equality holds if and only if $W(\omega) = \triangle F \ a.s.$, i.e.

$$\int_0^T \frac{\partial \log \pi_s}{\partial s}(X_s(\omega))ds = 0, \ \ P_{[0,T]}\text{-a.s.}$$

Furthermore, if the inhomogeneous multidimensional diffusion process $X = \{X(t)\}_{0\le t\le T}$ is unperturbed, then $\frac{\partial \log \pi_s}{\partial s}(x) \equiv 0$, and consequently $W(\omega) = \triangle F$ for each trajectory ω. □

Remark 2.10 In the homogeneous(steady state) case, the diffusion process is obviously unperturbed, which yields $W_d \equiv 0$. So the theorems above become trivial.

Recently, M. Baiesi et al. [1] have proved an exact fluctuation theorem for the dissipative work W_d, i.e.

$$\frac{P_{\pi_0}(W_d = x)}{P_{\pi_0}(W_d = -x)} = e^{\beta x}, \tag{12}$$

for each x, under some condition [1, (14)], which can also give rise to the generalized Jarzynski's equality $Ee^{-\beta W_d} = 1$. They also pointed out that the diffusive dynamics on an asymmetric potential may not agree with this exact fluctuation theorem [1, Fig. 2], while the generalized Jarzynski's equality (Theorem 2.6) still holds. In addition, if the time-averaged dissipative work $\frac{W_d}{T}$ has a large deviation property with rate function $I(x)$, then (12) leads to the Gallavotti-Cohen type fluctuation theorem $I(x) = I(-x) - x$.

3 Physical Meaning and Applications

3.1 Generalization of Hummer and Szabo's Work

In Jarzynski's original work [21–24] and Crooks' recent work [2–4], they derived the Jarzynski's equality through standard derivation of statistical physics for simple stochastic models including inhomogeneous denumerable Markov chains.

Afterwards, in Hummer and Szabo's paper [20], it is shown how equilibrium free energy profiles can be extracted rigorously from repeated nonequilibrium force measurements on the basis of an extension of Jarzynski's remarkable identity between free energies and the irreversible work. But they misused the Feynman-Kac formula [27, Theorems 4.4.2, 5.7.6]. This well-known formula provides a stochastic representation for the solution $v(t,x)$ of the concerned parabolic equation through the conditional expectation of the path integration of a specific stochastic process $\xi = \{\xi_t : t \geq 0\}$, and $v(t,x)$ can be regarded intuitively as an expectation with respect to the "weighted" phase space distribution (see (8)). In the inhomogeneous case, this expectation is taken conditioned on the event $\{\xi_t = x\}$ at time t, while in the homogeneous case, x is taken to be the starting point of the trajectory of ξ, i.e. the expectation in the representation is taken conditioned on $\{\xi_0 = x\}$ at the initial time. More important, the quantity $v(t,x)$ in the Feynman-Kac formula actually should be defined by holding the final time T fixed and treating the time t as a variable, and in the inhomogeneous situation, the time interval for the path integration can not be shifted from $[t,T]$ to $[0,T-t]$ while it can in the homogeneous case, which implies that it is impossible to directly integrate the quantity $v(t,x)$ with respect to x applying the initial equilibrium distribution to derive the generalized Jarzynski's equality. Therefore, from the mathematical point of view, the right side of [20, (4)] and the quantity $g(z,t)$ defined in [22, (14)] can not satisfy the Feymann-Kac formula in the inhomogeneous case. On the other hand, although the Kolmogorov forward equation is more familiar and intuitively natural for physicists, the path integration of the specific diffusion process (i.e. the quantity $v(t,x)$) can only satisfy the elliptic equation similar to the Kolmogorov backward equation according to the standard Feynman-Kac formula [27, Theorem 5.7.6], which is just another reason why Hummer and Szabo's derivation [20] is flawed.

What they considered is the diffusive dynamics on a potential $V(t,x)$ satisfying $\int_{\mathbb{R}^d} V(t,x)dx < \infty$, whose time evolution is governed by the differential operator $L_t = \frac{1}{2}\nabla \cdot A(t,x)\nabla + b(t,x)\cdot\nabla$, in which $A(t,x) = 2D$ is the diffusion coefficient and $b(t,x) = D\nabla V(t,x)$ is the drift coefficient. Now, we can restate the results of Hummer and Szabo's work in the case of general multidimensional diffusion processes, applying the mathematical theory in the previous section. The statements below could be anticipated starting from [20], but they are here rigorously derived.

It is important to point out that in the homogeneous diffusion case, the force $2A^{-1}(x)b(x)$ has a potential $V(x)$ if and only if the steady state is an equilibrium state [25, Theorem 3.3.7]. In this case, the invariant distribution $\pi(x)$ can be expressed as the Boltzmann distribution

$$\pi(x) = \frac{e^{-V(x)}}{\int_{\mathbb{R}^d} e^{-V(x)}dx}.$$

Moreover, the stationary diffusion process with initial distribution density $\pi(x)$ is in detailed balance.

Suppose that for each $t \in [0, T]$, the force $2A^{-1}(t,x)b(t,x)$ has a potential $-\beta H(t,x)$, then the quasi-invariant distribution

$$\pi(t,x) = \frac{e^{-\beta H(t,x)}}{\int_{\mathbb{R}^d} e^{-\beta H(t,x)} dx},$$

and

$$F(t) = -\beta^{-1} \ln \int_{\mathbb{R}^d} e^{-\beta H(t,x)} dx$$

is the Helmholtz free energy of this diffusion process at time t.

Therefore, $W(\omega) = \int_0^T \frac{\partial H}{\partial s}(s, X_s) ds$ defined in the previous section is just the external work done on the system, $Q(\omega)$ in (7) is regarded as the total heat exchanged with the reservoir, and (7) is actually the extension of the first law of thermodynamics.

The reversible work, $W_r = \triangle F = F(T) - F(0)$, is the free energy difference between two equilibrium ensembles. And the dissipative work $W_d = W - W_r$, is defined as the difference between the actual work and the reversible work.

By Theorems 2.6 and 2.9, we get

Theorem 3.1 *Under the condition of Theorem 2.6, the well known Jarzynski's equality becomes*

$$E^{P_{[0,T]}}[e^{-\beta W_d}] = 1,$$

i.e.

$$E^{P_{[0,T]}}[e^{-\beta W}] = e^{-\beta \triangle F}. \tag{13}$$

And $E^{P_{[0,T]}} W_d \geq 0$, i.e. $E^{P_{[0,T]}} W \geq \triangle F$, which is an extension of the second law of thermodynamics. Moreover, $E^{P_{[0,T]}} W = \triangle F$ if and only if

$$\int_0^T \frac{\partial \log \pi_s}{\partial s}(X_s(\omega)) ds = 0, \quad P_{[0,T]}\text{-a.s.}$$

If the inhomogeneous multidimensional diffusion process $X = \{X(t)\}_{0 \leq t \leq T}$ is unperturbed, then $W(\omega) = \triangle F$ for each trajectory ω.

Remark 3.2 Although the existence of a potential for the force $2A^{-1}b(t,x)$ is not a necessity in our proof of Sect. 2, it is essential for the concept of free energy in physics, because free energy can only be defined for the equilibrium states.

3.2 Generalization of Hatano and Sasa's Work

As we have mentioned in the introduction of the present paper, Hatano and Sasa only derived their result in the case of inhomogeneous Markov chains rather than diffusion processes, and they regarded the corresponding result in the case of inhomogeneous diffusion processes be the direct limit of that in inhomogeneous Markov chain case. From the mathematical point of view, diffusion processes can be regarded as the limit of inhomogeneous Markov chains, but only in the sense of distribution rather than trajectories.

Using the phenomenological framework of steady-state thermodynamics constructed by Oono and Paniconi [31], they show that an extended form of the second law holds for transitions between steady states, relating the Shannon entropy (also accepted as the common definition of Gibbs entropy) difference to the excess heat produced in an infinitely slow operation. A generalized version of the Jarzynski work relation plays an important role in their theory [15].

In their work, they studied a simple one-dimensional stochastic model of Langevin dynamics describing nonequilibrium steady states with drift coefficient $\frac{1}{\gamma}(-\frac{\partial U(x;\alpha)}{\partial x}+f)$ and diffusion coefficient $2k_BT=\frac{2}{\beta}$. What they are concerned with is to establish the connection between the phenomena displayed by nonequilibrium steady states and thermodynamic laws. Three kinds of heats are defined: the total heat Q_{tot}, the housekeeping heat Q_{hk} and the excess heat Q_{ex}, satisfying $Q_{tot}=Q_{hk}+Q_{ex}$. By convention, they take the sign of heat to be positive when it flows from the system to the heat bath.

In the case of inhomogeneous multidimensional diffusion processes, the housekeeping heat

$$Q_{hk}(\omega)=\frac{1}{\beta}\int_0^T[2A^{-1}b(t,X_t)-\nabla\log\pi_t(X_t)]dX_t, \tag{14}$$

where dX_t is of the Stratonovich type. A simple example of this interpretation of the heat was implied in Sekimoto's work [38] and explicitly defined in Hatano and Sasa's work [15].

It has been proved that for equilibrium systems, $2A^{-1}b(t,x)=\nabla\log\pi_t(x)$ [25, Theorem 3.3.7], hence Q_{ex} reduces to the total heat Q_{tot}.

In the case of inhomogeneous multidimensional diffusion processes, the total heat is defined as

$$Q_{tot}(\omega)=\frac{1}{\beta}\int_0^T(2A^{-1}b(t,X_t))dX_t. \tag{15}$$

Since

$$\triangle H(\omega)=\int_0^T\frac{\partial H}{\partial t}(t,X_t)dt+\int_0^T\nabla H(t,X_t)dX_t$$

and

$$\nabla H(t,x)=-\frac{1}{\beta}\nabla\log\pi_t(x),$$

we find that the excess heat defined in Hatano and Sasa's paper is just

$$\begin{aligned}Q_{ex}(\omega)=Q_{tot}(\omega)-Q_{hk}(\omega)&=\int_0^T\frac{1}{\beta}\nabla\log\pi_t(X_t)dX_t\\&=-\int_0^T\nabla H(t,X_t)dX_t=-\triangle H(\omega)+\int_0^T\frac{\partial H}{\partial t}(t,X_t)dt=-Q(\omega),\end{aligned} \tag{16}$$

where $Q(\omega)$ is defined in (7).

Denote $\phi(t,x)=-\log\pi(t,x)$ to be the Gibbs entropy (also called Gibbs free energy) of state x at time t, then

$$\triangle\phi=\beta(\triangle H-\triangle F).$$

Therefore,

$$W_d=W-\triangle F=(\triangle H-Q)-\triangle F=Q_{ex}+\frac{\triangle\phi}{\beta}. \tag{17}$$

So we can get the generalized Jayzynski's equality of nonequilibrium steady states.

Theorem 3.3 *Under the condition of Theorem 2.6,*

$$E^{P_{[0,T]}}[e^{-\beta Q_{ex}-\triangle\phi}]=1. \tag{18}$$

Let $S(t)=\langle\phi(t)\rangle=-\int_{\mathbb{R}^d}\pi(t,x)\log\pi(t,x)dx$ be the Gibbs entropy (Shannon entropy) at time t, then the extension of the second law of thermodynamics in NESS becomes

Theorem 3.4

$$\triangle S=\triangle\langle\phi\rangle\geq-\beta\langle Q_{ex}\rangle. \tag{19}$$

Moreover, $-\beta\langle Q_{ex}\rangle=\triangle S$ *if and only if*

$$\int_0^T\frac{\partial\log\pi_s}{\partial s}(X_s(\omega))ds=0,\qquad P_{[0,T]}\text{-}a.s.$$

If the inhomogeneous multidimensional diffusion process $X=\{X(t)\}_{0\leq t\leq T}$ *is unperturbed, then* $-\beta\langle Q_{ex}\rangle=\triangle S$ *for each trajectory* ω.

Therefore, the generalized entropy difference $\triangle S$ between two steady states can be measured through $-\beta\langle Q_{ex}\rangle$ resulting from a slow (unperturbed) process connecting these two states, which allows one to define the generalized entropy of nonequilibrium steady states experimentally, by measuring the excess heat obtained in a slow process between any nonequilibrium steady state and an equilibrium state whose entropy is known.

As in [15], from (19) one can derive the minimum work principle for steady-state thermodynamics.

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