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Generalized integral fluctuation theorem for diffusion processes

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Using Feynman-Kac and Cameron-Martin-Girsanov formulas, we find a generalized integral fluctuation theorem (GIFT) for general diffusion processes by constructing a time-invariable integral. The existing integral fluctuation theorems can be derived as its specific cases. We interpret the origin of the GIFT in terms of time reversal of stochastic systems.

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I. INTRODUCTION

The Feynman-Kac (FK) formula, originally found by Feynman in quantum mechanics [1] and extended by Kac [2], establishes an important connection between partial differential equations (PDEs) and stochastic processes. Using this formula, one may solve certain PDE by simulating a stochastic process. Recently, the remarkable FK formula was employed by Hummer and Szabo (HS) [3] to prove the celebrated Jarzynski equality (JE) [4,5] in nonequilibrium statistics [6]. Their work not only provided a concise proof but also pointed out that intrinsic free-energy surfaces of biomolecules could be efficiently extracted by nonequilibrium single-molecule manipulation experiments.

Compared to extensive interests of applications of HS's work in single-molecule biophysics [7,8], little attention was paid to their proving method. Very recently, Ge and Jiang (GJ) reinvestigated previous derivation from a rigorously mathematical point of view [9]. They pointed out that HS misused the FK formula since it is usually applied for Kolmogorov backward equation [10] rather than Kolmogorov forward (or Fokker-Planck) equation. In fact, Chetrite and Gawedzki (CG) [11] gave a correct proof of the JE using the FK formula slightly earlier. Interestingly, however, these two works seem to be apparently distinct though they employed the same mathematical formula: GJ constructed a simple time-invariable integral (the proof of theorem 2.6 in Ref. [9]), whereas CG based on time-reversal concept.

In addition to the JE, there are several analogous equalities found in the past few years [12–18]. All of them have a type of

$$\langle \exp[-\mathcal{A}] \rangle = 1, \tag{1}$$

where $\mathcal{A}$ is a functional of the stochastic trajectory of a stochastic system and angular brackets denote an average over the ensemble of the trajectories. For instance, $\mathcal{A}$ is total entropy change along a trajectory [15]. These equalities were called integral fluctuation theorems (IFTs) to distinguish the detailed fluctuation theorems (DFTs) [13,19–22]. Due to the similarity between the IFTs with the JE in formality, one may naturally consider whether these equalities or even more general versions could be derived by GJ's approach as well. On the other hand, it should be intriguing to clarify the relationship between their approach and the others, e.g., the time reversal, which was not performed in the previous work. In this Rapid Communication, we present our results about these two questions. With the help of the FK and Cameron-Martin-Girsanov (CMG) [23,24] formulas, we obtain a generalized integral fluctuation theorem by extending GJ's idea. We find this theorem could be explained from the concept of time reversal. Because we focus on the general Markovian diffusion processes, little physics is mentioned here. The detailed discussions about the existing IFTs in literature might remedy it.

II. TIME-INVARIABLE INTEGRAL

We consider a general N-dimension stochastic system $\mathbf{x}=\{x_i\}, i=1,\ldots,N$ described by a stochastic differential equation (SDE) [25]

$$d\mathbf{x}(t) = \mathbf{A}(\mathbf{x},t)dt + \mathbf{B}^{1/2}(\mathbf{x},t)d\mathbf{W}(t), \tag{2}$$

where $d\mathbf{W}$ is an N-dimensional Wiener process, $\mathbf{A}=\{A_i\}$ denotes a N-dimensional drift vector, and $\mathbf{B}^{1/2}$ is the square root of a $N\times N$ positive definite diffusion matrix $\mathbf{B}=\{B_{ij}\}$. We use Ito's convention for stochastic differentials. Rather than directly solving Eq. (2), one usually converts the SDE into two equivalent PDEs of the conditional probability density $\rho(\mathbf{x},t|\mathbf{x}',t')(t>t')$: the forward Fokker-Planck equation (FPE) $\partial_t\rho=\mathcal{L}(\mathbf{x},t)\rho$, with

$$\mathcal{L}(\mathbf{x},t) = -\partial_{x_i}A_i(\mathbf{x},t) + \frac{1}{2}\partial_{x_i}\partial_{x_j}B_{ij}(\mathbf{x},t), \tag{3}$$

and the backward FPE $\partial_{t'}\rho=-\mathcal{L}^+(\mathbf{x}',t')\rho$, with

$$\mathcal{L}^+(\mathbf{x},t) = A_i(\mathbf{x},t)\partial_{x_i} + \frac{1}{2}B_{ij}(\mathbf{x},t)\partial_{x_i}\partial_{x_j}. \tag{4}$$

The initial conditions in both cases are $\rho(\mathbf{x},t|\mathbf{x}',t)=\delta(\mathbf{x}-\mathbf{x}')$. The two operators $\mathcal{L}$ and $\mathcal{L}^+$ are adjoint to each other. Here we use Einstein's summation convention throughout this work unless explicitly stated.

Based on the symbols and definitions above, we find that, if $u(\mathbf{x},t)$ satisfies a partial differential equation

$$\begin{aligned}\partial_{t'}u(\mathbf{x},t') = &-\mathcal{L}^+(\mathbf{x},t')u(\mathbf{x},t') - f^{-1}(\mathbf{x},t')[\partial_{t'}f(\mathbf{x},t') \\ &- \mathcal{L}(\mathbf{x},t')f(\mathbf{x},t')]u(\mathbf{x},t') + f^{-1}(\mathbf{x},t') \\ &\times[\mathcal{L}_a(\mathbf{x},t')g(\mathbf{x},t') - g(\mathbf{x},t')\mathcal{L}_a^+(\mathbf{x},t')]u(\mathbf{x},t'),\end{aligned} \tag{5}$$

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where $f(\mathbf{x},t')$ and $g(\mathbf{x},t')$ are arbitrary normalized smooth positive functions [26], $\mathcal{L}_a$ and $\mathcal{L}_a^+$ are adjoint operators such as Eqs. (3) and (4), which may be the same with or different from those of the system we focus on, we have

$$\frac{d}{dt'}\int d\mathbf{x}f(\mathbf{x},t')u(\mathbf{x},t')=0. \tag{6}$$

The proof is obvious if one makes use of the adjoint property of these operators and notes that the derivative of the integral with respect to time is

$$\frac{d}{dt'}\int d\mathbf{x}f(\mathbf{x},t')u(\mathbf{x},t')=\int d\mathbf{x}[f\partial_{t'}u(\mathbf{x},t')+u\partial_{t'}f(\mathbf{x},t')]. \tag{7}$$

This is a simple generalization of GJ's idea, where the last three terms in Eq. (5) were absent [27]. Multiplying both sides of this PDF by $f(\mathbf{x},t)$, one may see that this equation has a certain symmetry with respect to the functions f and u. In the following, we investigate Eq. (5) with the Liouville- and Fokker-Planck-type $\mathcal{L}_a$, respectively. Although one may think that the former is only a specific case of the latter (vanishing diffusion matrix), the Liouville type may be more intriguing that will be seen shortly.

III. LIOUVILLE-TYPE $\mathcal{L}_a(\mathbf{x},t)$

We assume

$$\mathcal{L}_a g(\mathbf{x},t')=2\partial_{x_i}S_i(\mathbf{x},t'), \tag{8}$$

and $\mathbf{S}=\{S_i(\mathbf{x},t)\}$ is an arbitrary vector having natural boundary condition. The coefficient 2 is for convenience in discussion. We rewrite Eq. (5)

$$\begin{aligned}\partial_{t'}u(\mathbf{x},t')=&-\mathcal{L}^+(\mathbf{x},t')u(\mathbf{x},t')-f^{-1}(\mathbf{x},t')[\partial_{t'}f(\mathbf{x},t')\\&-\mathcal{L}(\mathbf{x},t')f(\mathbf{x},t')]u(\mathbf{x},t')\\&+2f^{-1}(\mathbf{x},t')[\partial_{x_i}S_i(\mathbf{x},t')+S_i(\mathbf{x},t')\partial_{x_i}]u(\mathbf{x},t').\end{aligned} \tag{9}$$

Using the FK and CMG formulas [10], we obtain a stochastic representation of the solution $u(\mathbf{x},t)$,

$$u(\mathbf{x},t')=E^{\mathbf{x},t'}\{e^{-\mathcal{J}[f,\mathbf{S},\mathbf{x}(\cdot)]}q[\mathbf{x}(t)]\}, \tag{10}$$

where $q(\mathbf{x})$ is the final condition of $u(\mathbf{x},t)$, and the functional

$$\begin{aligned}\mathcal{J}[f,\mathbf{S},\mathbf{x}(\cdot)]=&\int_{t'}^{t}\frac{1}{f}\left[(\mathcal{L}-\partial_\tau)f+2\partial_{x_i}S_i+\frac{2}{f}S_i(\mathbf{B}^{-1})_{ij}S_j\right]d\tau\\&+\int_{t'}^{t}\frac{2}{f}S_i(\mathbf{B}^{-1})_{ij}(dx_j-A_i d\tau),\end{aligned} \tag{11}$$

where the expectation $E^{\mathbf{x},t'}$ is an average over all trajectories $\mathbf{x}(\cdot)$ determined by Eq. (2) taken conditioned on $\mathbf{x}(t')=\mathbf{x}$, and the last term is a Ito stochastic integral. Choosing $t'=0$ and combining Eqs. (6) and (10), we have

$$\int d\mathbf{x}'f(\mathbf{x}',0)E^{\mathbf{x}',0}\{e^{-\mathcal{J}[f,\mathbf{S},\mathbf{x}(\cdot)]}q[\mathbf{x}(t)]\}=\int d\mathbf{x}f(x,t)q(\mathbf{x}). \tag{12}$$

This equation has the same form as Eq. (1) for $q(\mathbf{x})=1$. We call it generalized integral fluctuation theorem (GIFT). Note other functions of $q(\mathbf{x})$ are still useful in practice.

Relationship between the GIFT and existing IFTs

Existing several IFTs are specific cases of Eq. (12). First is to choose $S_i=0$ and $f=p^{ss}(\mathbf{x},t)$, where $p^{ss}(\mathbf{x},t')$ is the transient steady-state solution

$$\mathcal{L}(\mathbf{x},t')p^{ss}(\mathbf{x},t')=0. \tag{13}$$

Then functional (11) is simply

$$\mathcal{J}=-\int_0^t\partial_\tau\ln p^{ss}[x(\tau),\tau]d\tau. \tag{14}$$

GJ analyzed this case with a final condition $q(\mathbf{x})=1$ in great details. According to the system driven by a time-dependent conservative (a gradient of a potential) or nonconservative force, Eq. (12) reduces to Jarzynski equality [4,5] and Hatano-Sasa relation [14], respectively [9]. Here we do not repeat the same derivatives. We only point out the necessity of the general final condition: one obtains key Eq. (4) in HS's work only choosing $q(\mathbf{x})=\delta(\mathbf{x}-\mathbf{z})$.

The second case is to choose S_i to be the probability current $J_i[f]$ of an arbitrary normalized function $f(\mathbf{x},t)$, where

$$J_i[f(\mathbf{x},t)]=A_i(\mathbf{x},t)f(\mathbf{x},t)-\frac{1}{2}\partial_{x_j}B_{ij}f(\mathbf{x},t). \tag{15}$$

Substituting the current into Eq. (11) and doing some derivations (the details see the Appendix), we obtain

$$\begin{aligned}\mathcal{J}=\ln\frac{f[\mathbf{x}(\mathbf{t}'),t']}{f[\mathbf{x}(\mathbf{t}),t]}+2(S)\int_{t'}^{t}\hat{A}_i(\mathbf{B}^{-1})_{ij}\\\times\left[\dot{x}_j-\frac{1}{2}(\mathbf{B}^{-1/2})_{ks}\partial_{x_k}(\mathbf{B}^{-1/2})^T_{sj}\right]d\tau,\end{aligned} \tag{16}$$

where $\hat{A}_i=A_i-\partial_{x_l}B_{il}/2$ and $\dot{x}_j=dx_j/d\tau$. Note that we used the Stratonovich's integral (denoted by "S" therein). Particularly, if the diffusion matrix is constant that is usually assumed in Langevin equation and $f(\mathbf{x},t)$ is the solution of the stochastic system with a initial condition $f(\mathbf{x},0)$, the functional is just the total entropy change $\Delta s_{\rm tot}$: the first term in Eq. (16) is the system entropy change and the second is the entropy change of environment [16]. Hence, the GIFT with Eq. (16) is the IFT of the total entropy [15,16].

The last case is for the system being in a nonequilibrium steady state with a distribution $p^{ss}(\mathbf{x})$. We choose $f=p^{ss}(\mathbf{x})$ and $S_i=J_i(p^{ss})$. Considering that $\partial_{x_i}S_i=0$, the functional becomes the housekeeping heat [17]

$$\mathcal{J}=2\int_{t'}^{t}\frac{J_i(p^{ss})}{p^{ss}}(\mathbf{B}^{-1})_{ij}\left[\dot{x}_j-\frac{1}{2}(\mathbf{B}^{-1/2})_{kl}\partial_{x_k}(\mathbf{B}^{-1/2})^T_{lj}\right]d\tau. \tag{17}$$

Of course, one can obtain this functional from Eq. (16) directly. Importantly, this IFT is even correct for time-dependent case [17]. We may indeed prove it using CMG formula only [11].

IV. FOKKER-PLANCK-TYPE $\mathcal{L}_a(\mathbf{x},t')$

If the diffusion matrix of $\mathcal{L}_a$ is not zero, we can still obtain a GIFT given

$$\mathcal{L}_a(\mathbf{x},t') = \left[-\partial_{x_i} a^i(\mathbf{x},t') + \frac{1}{2}\partial_{x_i}\partial_{x_j} b_{ij}(\mathbf{x},t') \right]. \quad (18)$$

Assuming $a^i = a_1^i(\mathbf{x},t) - 2a_2^i(\mathbf{x},t)$, we split this operator into a sum of a Fokker-Planck and Liouville operators, i.e.,

$$\mathcal{L}_a(\mathbf{x},t') = [\mathcal{L}_a^1(\mathbf{x},t') + 2\partial_{x_i} a_2^i], \quad (19)$$

where $\mathcal{L}_a^1$ is the same with Eq. (18) except that a^i is replaced by a_1^i. Substituting Eq. (19) into Eq. (5) and doing a simple rearrangement, one may obtain Eq. (9) again if we redefine the drift vector and diffusion matrix of the operator $\mathcal{L}(\mathbf{x},t')$ to be

$$A_i' = A_i(\mathbf{x},t') + a_i^1(\mathbf{x},t')g(\mathbf{x},t')/f(\mathbf{x},t'),$$

$$B_{ij}' = B_{ij}(\mathbf{x},t') + b_{ij}(\mathbf{x},t')g(\mathbf{x},t')/f(\mathbf{x},t'), \quad (20)$$

and $S_i = a_2^i(\mathbf{x},t')g(\mathbf{x},t')$. Hence, we have a GIFT [Eq. (12)] except that the expectation $E^{x,t'}$ is now an average over the trajectories generated from a new stochastic system

$$d\mathbf{x}(t) = \mathbf{A}'(\mathbf{x},t)dt + \mathbf{B}'^{1/2}(\mathbf{x},t)d\mathbf{W}(t). \quad (21)$$

From this aspect, Eq. (6) with the Fokker-Planck-type $\mathcal{L}_a$ seems to not provide additional information compared to the Liouville type [28].

V. GIFT AND TIME REVERSAL

We must emphasize that the derivation of the GIFT [Eq. (12)] is based on the adjoint characteristic of the FK operators without resorting to any physical reason. It would be intriguing to establish a relationship between the equality and time reversal similar to that done by Crook [13] in interpreting JE [4,5]. CG recently suggested that the different fluctuation relations in literature may be traced to different time reversals [11]. In the remainder of the work, we employ this viewpoint to understand the origin of Eq. (5) and the GIFT.

We first give a definition of a time reversal [11]. The variable x_i of the stochastic system is even or odd according to their rules under time reversal $t' \to t-t'$: if $x_i \to y_i = +x_i$ is even and $x_i \to y_i = -x_i$ is odd. We write them in abbreviation $x_i \to y_i = \varepsilon_i x_i$ and $\varepsilon_i = \pm 1$. The drift vector splits into "irreversible" and "reversible" parts, $\mathbf{A} = \mathbf{A}^{\rm irr} + \mathbf{A}^{\rm rev}$. Under a time reversal, these vectors are transformed into $\tilde{\mathbf{A}} = \tilde{\mathbf{A}}^{\rm irr} + \tilde{\mathbf{A}}^{\rm rev}$, where

$$\tilde{A}_i^{\rm irr}(\mathbf{x},t') = \varepsilon_i A_i^{\rm irr}(\mathbf{y},s), \quad (22)$$

$$\tilde{A}_i^{\rm rev}(\mathbf{x},t') = -\varepsilon_i A_i^{\rm rev}(\mathbf{y},s), \quad (23)$$

$\mathbf{y} = \{y_i\}$, and $s = t-t' (0 \le t' \le t)$. We also define a transformation of the diffusion matrix to be

$$\tilde{B}_{ij}(\mathbf{x},t') = \varepsilon_i \varepsilon_j B_{ij}(\mathbf{y},s). \quad (24)$$

Note that no summation over repeated indices here. Because the splitting is completely arbitrary, one may regard it as an equivalent definition of a certain time reversal.

Considering a forward FPE $\partial_s p(\mathbf{y},s) = \tilde{\mathcal{L}}(\mathbf{y},s)p(\mathbf{y},s)$ with the time-reversed drift vector and diffusion matrix,

$$\tilde{\mathcal{L}}(\mathbf{y},s) = -\partial_{y_i}\tilde{A}_i(\mathbf{y},s) + \frac{1}{2}\partial_{y_i}\partial_{y_j}\tilde{B}_{ij}(\mathbf{y},s). \quad (25)$$

Substituting

$$p(\mathbf{y},s) = f(\mathbf{x},t')v(\mathbf{x},t') \quad (26)$$

into above equation and doing some simple derivations, we obtain

$$\partial_{t'} v(\mathbf{x},t') = -\mathcal{L}^+(\mathbf{x},t')v(\mathbf{x},t') - f^{-1}(\mathbf{x},t')[\partial_{t'} f(\mathbf{x},t') - \mathcal{L}(\mathbf{x},t')f(\mathbf{x},t')]v(\mathbf{x},t') + 2f^{-1}(\mathbf{x},t') \times [\partial_{x_i} S_i^{\rm irr}(f) + S_i^{\rm irr}(f)\partial_{x_i}]v(\mathbf{x},t'), \quad (27)$$

where an irreversible probability current is defined to be

$$S_i^{\rm irr}(f) = A_i^{\rm irr}(\mathbf{x},t')f(\mathbf{x},t') - \frac{1}{2}\partial_{x_k} B_{ik}(\mathbf{x},t')f(\mathbf{x},t'). \quad (28)$$

We immediately see that Eq. (9) would be the same with Eq. (27) if $S_i = S_i^{\rm irr}(f)$. Under this circumstance, Eq. (12) is a obvious consequence of the specific time reversal of the stochastic system in that the integral of the left-hand side of Eq. (6) equals to $\int d\mathbf{y} p(\mathbf{y},s)$, which is conservative with respect to time. On the other hand, considering that the splitting of the drift vector is completely arbitrary, we can define a time reversal for a given vector S_i by

$$A^{\rm irr}(\mathbf{x},t') = \frac{1}{f(\mathbf{x},t')}\left[S_i(\mathbf{x},t') + \frac{1}{2}\partial_{x_k} B_{ik}(\mathbf{x},t')f(\mathbf{x},t') \right], \quad (29)$$

$$A^{\rm rev}(\mathbf{x},t') = A_i(\mathbf{x},t') - A^{\rm irr}(\mathbf{x},t'). \quad (30)$$

Then the GIFT for any vector $\mathbf{S}$ is a consequence of the new defined time reversal of the stochastic system. The reader should be reminded that these time reversals are not always meaningful or easily realizable in physics. Before ending the section, we give two comments about Eq. (26). First, for a homogenous diffusion process that satisfies the detailed balance, this equation has been used earlier to connect the forward FK solution $p(\mathbf{x},t)$ and backward FK solution $v(\mathbf{x},t)$, where $f(\mathbf{x},t) = p^{\rm eq}(\mathbf{x})$ [25]. Second, Eq. (26) and the stochastic representation solution $v(\mathbf{x},t')$ [Eq. (10)] are crucial for achieving the DFTs [12,13]. CG [11] gave a detailed discussion about this point (proposition 1 therein).

VI. CONCLUSION

In this Rapid Communication, we obtain a generalized integral fluctuation theorem for diffusion processes using the famous FK and CMG formulas. Although one might think that this result [Eq. (12)] is simple from the point of view of time reversal that we explained here, the derivation by constructing a time-invariable integral is novel. An apparent limit of this work is that we did not show any possible applications or new physics about the GIFT. We do not solve this point yet. We close this work by pointing out several directions that we may further improve the current results. First is to give rigorous conditions for applying FK and CMG formulas. Second, we may extend the results for the continuous diffusion processes to discrete state case, e.g., chemical master equation. Finally, the most interesting and urgent question to us is to apply the GIFT in practical problems, e.g., free-energy reconstruction of single-molecule manipulation.

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APPENDIX: DERIVATION OF THE TOTAL ENTROPY

Substituting Eq. (15) into Eq. (11) and separating the terms into f-dependent and f-independent parts, we obtain

$$\mathcal{J}=\int_{t'}^{t}[\partial_{x_i}\hat{A}_i+2\hat{A}_i(\mathbf{B}^{-1})_{ij}\hat{A}_j]d\tau+2\hat{A}_i(\mathbf{B}^{-1/2})_{ij}dW_j - \int_{t'}^{t}\left[\partial_\tau \ln f+A_i\partial_{x_i}\ln f+\frac{1}{2}B_{ij}\partial_{x_i}\partial_{x_j}\ln f\right]d\tau + (\partial_{x_i}\ln f)(\mathbf{B}^{1/2})_{ij}dW_j. \quad \text{(A1)}$$

Employing SDE (2), the second integral in above equation becomes

$$\int_{t'}^{t}\left[\partial_\tau \ln f+\frac{1}{2}B_{ij}\partial_{x_i}\partial_{x_j}\ln f\right]d\tau+\partial_{x_i}\ln f dx_i, \quad \text{(A2)}$$

which is just the system entropy change $\int_{t'}^{t} d\ln f =\ln\{f[x(t),t]/f[x(t'),t']\}$ according to the Ito formula [25]. The entropy change of environment in Eq. (16) is derived from the first integral in Eq. (A1) by converting the Ito's stochastic integral into the Stratonovich's:

$$\int_{t'}^{t}2\hat{A}_i(\mathbf{B}^{-1/2})_{ij}dW_j=(S)\int_{t'}^{t}2\hat{A}_i(\mathbf{B}^{-1/2})_{ij}dW_j - \frac{1}{2}(\mathbf{B}^{1/2})_{jk}\partial_{x_k}[2(\mathbf{B}^{-1/2})^T_{ij}\hat{A}_i]d\tau. \quad \text{(A3)}$$

An alternative method is based on time-reversal technique. Previous discussion shows that for $u(\mathbf{x},t')$ satisfying Eq. (9) and $S_i=J_i(f)$, we can define a time reversal with $\mathbf{A}^{\rm irr}=\mathbf{A}$ and $\mathbf{A}^{\rm rev}=0$. Then $f(\mathbf{x},t')u(\mathbf{x},t')=p(\mathbf{y},s)$ is a solution of FPE (25). Because f is arbitrary, we may choose $f=1$ and $w(\mathbf{x},t')=p(\mathbf{y},s)$. Hence we have $u(\mathbf{x},t')=w(\mathbf{x},t')\exp[-\ln f(\mathbf{x},t')]$. Obviously, the functional of the stochastic representation of $w(\mathbf{x},t')$ is the first integral of Eq. (A1).

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[26] This normalization requirement of these functions is inessential for our discussion.
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[28] It may be intriguing to investigate whether the solution of Eq. (5) with Eq. (18) has a stochastic representation on the original stochastic system (2).