Nonequilibrium Equalities Derived from Lebesgue’s Decomposition

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Most of the integral fluctuation theorems cannot be applied to situations in which the forward-path probability vanishes in a certain region or error-free measurements are performed under feedback control. We identify the mathematical origins of these problems based on Lebesgue’s decomposition theorem and derive new nonequilibrium equalities applicable to the above two situations. Inequalities derived from the equalities impose a stronger restriction on the averaged entropy production than the conventional second law in certain systems.

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Introduction. The last two decades have witnessed remarkable progress in nonequilibrium statistical mechanics [1–31]. In particular, Jarzynski established an integral nonequilibrium equality based on the Hamiltonian dynamics [8] and subsequently generalized it to a broader class of situations in nonequilibrium statistical mechanics [9–10]. Then, Crooks offered a general proof of the Jarzynski equality and fluctuation theorems in stochastic systems, based on a technique of comparing a thermodynamic process and its time-reversed one [11–13]. This technique is fundamentally important in evaluating thermodynamic irreversibility, i.e., entropy production. Later, a generalized Jarzynski equality under feedback control which involves mutual information obtained by measurement was shown in stochastic systems [19] and under the Hamiltonian dynamics [20]. Various types of the Jarzynski equality can be shown in stochastic systems by an appropriate choice of the so-called reference probability [24].

Possible applications of nonequilibrium fluctuation equalities range from physics to biology [26–31]. For example, the free-energy landscape of a DNA can be surveyed through nonequilibrium experiments [30] using the Hummer-Szabo equality [14]. Moreover, feedback control based on information processing as in Ref. [31] can be utilized to manipulate nanomachines subject to large thermal fluctuations such as in vivo.

Although the nonequilibrium equalities have wide applications, there are as yet uncovered important situations. The Jarzynski equality is inapplicable to free expansion of an ideal gas as discussed in Refs. [32–36]. Horowitz et al. pointed out that this is due to the fact that the forward-path probability vanishes and the backward-path probability is nonzero [22]. Such a situation arises when the initial probability distribution is confined to a restricted region in phase space or when error-free measurement is performed under feedback control [22–28]. These situations have been explicitly exempt from previous considerations, for example, in the proof of generalized Jarzynski equalities in Refs. [21–25]. Exceptions are Kawai-Parrondo-Van den Broeck-type equalities which circumvent the problem arising from the zero forward-path probability by formulation using the relative entropy [16–18]. It is natural to ask whether we can generalize Jarzynski-type equalities which are applicable to such frequently encountered situations.

In this Letter, we prove new nonequilibrium equalities which overcome the above difficulties. The previously excluded situations can be mathematically understood in terms of Lebesgue’s decomposition theorem in measure theory. Moreover, by the decomposition, we find that the conventional integral equality is inapplicable under a new situation, where a delta-function-like confinement exists in the reference probability. This corresponds, for example, to a situation in which a particle of interest is trapped in a certain narrow region such as a trapping center. It is noteworthy that the proof of the new equalities is valid regardless of the dynamics of the system and whether feedback control is performed or not.

Main equality. First, let us consider an arbitrary nonequilibrium process without feedback control. Let \( \Gamma(t) \) denote the trajectory of the system in phase space during a time interval \( 0 \leq t \leq \tau \), and let \( M[\Delta \Gamma(t)] \) denote the probability measure in phase space. Let \( M^r \) be an arbitrary reference probability measure. According to Lebesgue’s decomposition theorem [37, 38], \( M^r \) can be uniquely decomposed into two parts:

\[
M^r = M_{AC}^r + M_S^r,
\]

where \( M_{AC}^r \) is absolutely continuous with respect to \( M \) and this part can be written by the ratio of forward and backward probabilities as in previous literature [12–13]; \( M_S^r \) is the singular part corresponding to the region of the phase space in which the probability defined by \( M \) is zero while the probability defined by \( M^r \) remains nonvanishing. If \( M_S^r \) exists, the conventional Jarzynski-type equalities break down [21–25, 32–36]. Surprisingly, this purely mathematical theorem points to the distinct physical problems in the conventional fluctuation theorem and leads us to new nonequilibrium equalities as we show below.

Because \( M_{AC}^r \) is absolutely continuous with respect to \( M \), we may apply the Radon-Nikodym theorem to
obtain
\[ \mathcal{M}_{AC}[\Delta \Gamma(t)] = \left. \frac{\mathcal{D} \mathcal{M}_{AC}}{\mathcal{D} \mathcal{M}} \right|_{\Gamma(t)} \mathcal{M}[\Delta \Gamma(t)], \] (2)
where \( \mathcal{D} \mathcal{M}_{AC}/\mathcal{D} \mathcal{M} \big|_{\Gamma(t)} \) is the Radon-Nykodym derivative which is an integrable function with respect to \( \mathcal{M} \).

Let us formally define the entropy production as
\[ \sigma = -\ln \left. \frac{\mathcal{D} \mathcal{M}_{AC}}{\mathcal{D} \mathcal{M}} \right|_{\Gamma(t)}. \] (3)

If \( \mathcal{M} \) and \( \mathcal{M}_{AC} \) can be written by probability densities \( \mathcal{M}[\Delta \Gamma(t)] = \mathcal{P}[\Gamma(t)] \mathcal{D} \Gamma(t) \) and \( \mathcal{M}_{AC}[\Delta \Gamma(t)] = \mathcal{P}^r[\Gamma(t)] \mathcal{D} \Gamma(t) \), Eq. (3) can be rewritten as \( \sigma = -\ln \mathcal{P}^r[\Gamma(t)]/\mathcal{P}[\Gamma(t)] \), which is the standard definition of formal entropy production. A physical interpretation of \( \sigma \) will be discussed later.

Let \( \mathcal{F} \) denote an arbitrary functional of a path and \( \langle \cdots \rangle \) and \( \langle \cdots \rangle^r \) \( (I = AC, S) \) denote the averages over \( \mathcal{M} \), \( \mathcal{M}^r \) and \( \mathcal{M}_{r}^r \) respectively. Then, we have
\[ \langle \mathcal{F} \rangle^r_{AC} = \int \mathcal{F}[\Gamma(t)] \mathcal{M}_{AC}[\Delta \Gamma(t)] = \int \mathcal{F}[\Gamma(t)] \left. \frac{\mathcal{D} \mathcal{M}_{AC}}{\mathcal{D} \mathcal{M}} \right|_{\Gamma(t)} \mathcal{M}[\Delta \Gamma(t)] = \langle \mathcal{F} e^{-\sigma} \rangle. \] (4)

In accordance with the decomposition in Eq. (1), \( \langle \mathcal{F} \rangle^r_{AC} = \langle \mathcal{F} \rangle^r - \langle \mathcal{F} \rangle^r_S \) holds. Thus we obtain an integral fluctuation theorem:
\[ \langle \mathcal{F} e^{-\sigma} \rangle = \langle \mathcal{F} \rangle^r - \langle \mathcal{F} \rangle^r_S. \] (5)

This equality can be seen as a generalization of the master integral fluctuation theorem in Refs. [13, 24]. If we set \( \mathcal{F} = 1 \), Eq. (5) reduces to
\[ \langle e^{-\sigma} \rangle = 1 - \lambda_S, \] (6)
where \( \lambda_S = \int \mathcal{M}_{AC}[\Delta \Gamma(t)] \) is the probability of the singular part. The singular part has divergent entropy production with probability \( \lambda_S \). The right-hand side of Eq. (6) can therefore be interpreted as the probability for the absolute entropy production to be finite and mathematically well-defined. When the reference probability measure has only the absolute continuous part, i.e., \( \lambda_S = 0 \), the Jarzynski equality \( \langle e^{-\sigma} \rangle = 1 \) is reproduced. Using Jensen’s inequality: \( \langle e^{-\sigma} \rangle \geq e^{-\langle \sigma \rangle} \), we have
\[ \langle \sigma \rangle \geq -\ln(1 - \lambda_S). \] (7)

This inequality indicates that the second law of thermodynamics holds even when the singularity exists in the reference probability measure because the right-hand side is equal to or greater than zero. Moreover, when \( \lambda_S > 0 \) which is realized, for example, when the nonequilibrium process consists of free expansion, the inequality imposes a stronger restriction on the average entropy production than the second law, that is, the average entropy production must be positive in this case.

**Main equality under feedback control.** Let us now consider a nonequilibrium process with feedback control. Let \( \Lambda(t) \) denote a path in the phase space of outcomes. The control protocol can depend on \( \Lambda(t) \) at earlier times. Clearly, this protocol includes repeated discrete feedback discussed in Ref. [22]. Then, for a given \( \Lambda(t) \), we can choose an arbitrary reference probability measure \( \mathcal{M}_{AC}^r[\Lambda(t)] \), which can also be decomposed into two parts:
\[ \mathcal{M}_{AC}^r[\Lambda(t)] = \mathcal{M}_{AC}^r[\Lambda(t)] + \mathcal{M}_{S}^r[\Lambda(t)]. \] (8)

On the other hand, let \( \mathcal{M}[\Lambda(t)] \) denote the conditional probability measure for paths whose measurement outcomes are \( \Lambda(t) \). When \( \Lambda(t) \) is given, the control protocol is also fixed:
\[ \langle \mathcal{F} e^{-R[\Lambda(t)]} \rangle_{\Lambda(t)} = \langle \mathcal{F} \rangle^r_{\Lambda(t)} - \langle \mathcal{F} \rangle^r_{S[\Lambda(t)]}. \] (9)

Averaging this over the outcome \( \Lambda(t) \), we obtain
\[ \langle \mathcal{F} e^{-R[\Lambda(t)]} \rangle = \langle \mathcal{F} \rangle^r - \langle \mathcal{F} \rangle^r_S. \] (10)

The entropy production of the system should be defined by the initial probability measure and the conditional reference probability measure which reflects our knowledge acquired by measurement; then the entropy production is given by
\[ \sigma = -\ln \left. \frac{\mathcal{D} \mathcal{M}_{AC}^r[\Lambda(t)]}{\mathcal{D} \mathcal{M}^r_{\Lambda(t)} \Gamma(t)} \right|_{\Gamma(t)}. \] (11)

Then \( R[\Lambda(t)] \) can be separated into two parts:
\[ R[\Lambda(t)] = -\ln \left. \frac{\mathcal{D} \mathcal{M}_{AC}^r[\Lambda(t)]}{\mathcal{D} \mathcal{M}^r_{\Lambda(t)} \Gamma(t)} \right|_{\Gamma(t)} - \ln \left. \frac{\mathcal{D} \mathcal{M}}{\mathcal{D} \mathcal{M}^r_{\Lambda(t)} \Gamma(t)} \right|_{\Gamma(t)} = \sigma + I, \] (12)
where \( I = -\ln \mathcal{P}[\Gamma(t)]/\mathcal{P}[\Gamma(t)] \) of the system and that of the outcomes. If \( \mathcal{M} \) and \( \mathcal{M}[\Lambda(t)] \) can be written by probability densities, it reduces to the standard definition: \( I = -\ln \mathcal{P}[\Gamma(t)]/\mathcal{P}[\Gamma(t)] \). Equation (11) can be rewritten as
\[ \langle \mathcal{F} e^{-\sigma - I} \rangle = \langle \mathcal{F} \rangle^r - \langle \mathcal{F} \rangle^r_S. \] (13)

If we set \( \mathcal{F} = 1 \), Eq. (12) reduces to
\[ \langle e^{-\sigma - I} \rangle = 1 - \lambda_S. \] (14)

In particular, if the measurement is performed only once and there are no singular parts in the reference probability, the equality reproduces the original equality obtained in Refs. [19, 20]. The corresponding inequality is
\[ \langle \sigma \rangle \geq -\langle I \rangle - \ln(1 - \lambda_S). \] (15)
Thus, what determines the lower bound of entropy production is not the mutual information alone but the balance between the mutual information and the term arising from expansion and trapping. In particular, if \(<I> < -\ln(1 - \lambda_s)\), i.e., the right-hand side of the inequality is negative, the averaged entropy production may be negative.

When \(M\) can be written by the probability density, a stronger version of Lebesgue’s decomposition holds. Now, \(\mathcal{M}^r_{\Lambda(t)}\) can be uniquely decomposed into three parts:

\[
\mathcal{M}^r_{\Lambda(t)} = \mathcal{M}^r_{ac|\Lambda(t)} + \mathcal{M}^r_{sc|\Lambda(t)} + \mathcal{M}^r_{d|\Lambda(t)},
\]

where \(\mathcal{M}^r_{ac|\Lambda(t)}\) is absolutely continuous with respect to \(M\); \(\mathcal{M}^r_{sc|\Lambda(t)}\) is the singular continuous part corresponding to vanishing forward-path probability. What is new here is the discrete part \(\mathcal{M}^r_{d|\Lambda(t)}\) which represents such a reference probability distribution as a delta function.

To the best of our knowledge, no research has been conducted on this part. However, the existence of this part makes the conventional fluctuation theorem inapplicable. In accordance with Eq. (17), we obtain

\[
\langle F e^{-\sigma - I} \rangle = \langle F \rangle^r - \langle F \rangle^r_{ac} - \langle F \rangle^r_d, \tag{18}
\]

\[
\langle e^{-\sigma - I} \rangle = 1 - \lambda_{ac} - \lambda_d. \tag{19}
\]

The proof of the above nonequilibrium equalities [5, 0, 14, 15, 16, 19] can be made on a very general ground regardless of the dynamics of the system. In particular, the proof can be applied to Langevin systems and Hamiltonian systems. Physics enters the problem in the choice of the reference probability measure. Once the reference probability measure is chosen, the formal entropy production becomes the corresponding physical entropy production.

To elucidate the physical meaning of the derived formulae, let us first consider Langevin systems. By comparing the original dynamics and the time-reversed one, we can quantify the asymmetry in time reversal of a physical process. When we set the reference probability to the time-reversed path trajectory probability under the time-reversed protocol, the formal entropy production reduces to

\[
\sigma = -\ln \frac{\rho_0(\Gamma(t)|\Lambda(t))}{\rho_0(\Gamma(0))} + \Delta S^B, \tag{20}
\]

where \(\rho_0\) and \(\rho_0^\dagger\) are the initial probability distribution of the original path and that of the time-reversed one respectively, and \(\Delta S^B\) is the entropy production of the surrounding medium [24]. If we assume that the initial probability of the original path is the canonical distribution with inverse temperature \(\beta\) and set an arbitrary initial probability of the time-reversed path to the canonical distribution with the same inverse temperature \(\beta\), the entropy production becomes

\[
\sigma = \beta(W - \Delta F), \tag{21}
\]

where \(W\) is the work performed on the system and \(\Delta F\) is the free energy difference of the system [24]. Thus, the integral fluctuation theorem (15) reduces to

\[
\langle e^{-\beta(W - \Delta F) - I} \rangle = 1 - \lambda_s, \tag{22}
\]

which is the stochastic Jarzynski equality with feedback derived in Ref. [19] when \(\lambda_S = 0\). It is noteworthy that \(\lambda_S\) can be experimentally determined through measurement of the time-reversed process; \(\lambda_S\) is the total probability of backward paths with corresponding forward paths vanishing.

When we set the initial probability distribution of the time-reversed path to the final distribution of the original path, the first term in Eq. (20) is the Shannon entropy production of the system \(\Delta s\); then

\[
\sigma = \Delta s + \Delta s^B =: \Delta s^{tot} \tag{23}
\]

is the total entropy production of both the system and the bath. In this case, the integral fluctuation theorem is

\[
\langle e^{-\Delta s^{tot} - I} \rangle = 1 - \lambda_S. \tag{24}
\]

On the other hand, in deterministic systems such as Hamiltonian systems, if we fix the measurement outcome \(\Lambda(t)\) and then fix the protocol, the probability of a certain path is the same as the probability of being on the path at a certain time. In other words, the probability measures can be replaced as follows:

\[
\mathcal{M}(D\Gamma(t)) \rightarrow \mu_{t_1}(d\Gamma_{t_1}), \tag{25}
\]

\[
\mathcal{M}^r_{\Lambda(t)}(D\Gamma(t)) \rightarrow \mu^r_{t_2(\Lambda(t))}(d\Gamma_{t_2}), \tag{26}
\]

\[
\sigma \rightarrow -\ln[dp^r_{t_2(\Lambda(t))}/d\mu_{t_1}], \tag{27}
\]

where \(t_1\) and \(t_2\) are arbitrary times and \(\mu_{t_1}\) and \(\mu^r_{t_2(\Lambda(t))}\) are respectively the probability measure at time \(t_1\) and the (conditional) reference probability measure at time \(t_2\). Ordinarily, \(t_1\) and \(t_2\) are set to be the initial time 0 and the final time \(\tau\) of a nonequilibrium process. We can obtain equalities in exactly the same forms for deterministic processes with the same assumption and choice of the reference probability as in Eqs. (22) and (24).

Numerical Simulations. We demonstrate Eq. (19) by numerical simulations when \(I = 0\). First, let us consider the case of \(\lambda_d = 0\) and \(\lambda_s \neq 0\), i.e., the singular continuous part exists. We perform numerical simulations for an overdamped Langevin system confined on a one-dimensional ring, where the potential consists of \(n\) identical harmonic potential wells with stiffness \(k(t)\) (see Fig. 1(a)). The initial distribution is set to be the local equilibrium distribution in a given well and vanishes in all the other wells. We set the canonical distribution corresponding to the final potential as the reference probability. We study a nonequilibrium process during a time interval \(\tau\), in which the stiffness of potentials is decreased from \(k = K\) to 0 at a constant rate between \(t = 0\) and
FIG. 1. (a) Schematic illustration of an overdamped Langevin system consisting of $n$ harmonic potential wells on a one-dimensional ring. (b) Probability density of work for several $n$ values. The triangles indicate the points where $W = k_B T \ln n$. (c) Estimated values of $\langle \exp[-\beta W] \rangle$ at each $n$. Superposed is a fitted $1/n$ curve. (d) Estimated values of $\langle \beta W \rangle$. The line represents the minimum dissipation given by $k_B T \ln n$. Parameters are set as follows; diffusion constant $D = 10^{-12}$m$^2$/s; temperature $T = 300$K; duration of the process $\tau = 10$sec; half width of a single potential: $a = 10^{-4}$m; the initial stiffness of potential $K$ is set so as to satisfy $KA^2/2 = 5k_B T$. The nonequilibrium process is repeated $10^6$ times for each $n$.

$\tau/2$, and then increased from $k = 0$ to $n^2K$ at a constant rate between $t = \tau/2$ and $\tau$. If we assume $K$ is sufficiently large, the free-energy difference is zero in this process. Because a backward path terminates in a certain well with the equal probability, the probability that the backward path does not have the corresponding forward path is $\lambda_d = (n-1)/n$. Then the nonequilibrium integral equality reduces to $\langle e^{-\beta W} \rangle = 1/n$, and the corresponding second law is $\langle W \rangle \geq k_B T \ln n$. Figure 1(b) shows the distribution of work at different $n$ obtained by the numerical simulation. The value of $\langle e^{-\beta W} \rangle$ is confirmed to be $1/n$ (Fig. 1(c)). It is also verified that the averaged dissipation values are larger than the minima predicted by the inequality (Fig. 1(d)). When the initial probability is confined, there is diffusion to the entire phase space; then entropy production tends to be positive. Note that this process can be regarded as the information erasure process of the symmetric $n$-bit memory.

Next, let us consider the case of $\lambda_d \neq 0$ and $\lambda_s = 0$, i.e., the case in which discrete part exists. We perform numerical simulations for one-dimensional systems in which a single particle is trapped in a single harmonic potential with stiffness $k(t)$. There is a trapping point in the system and the distance between the point and the center of the harmonic potential is $x_c$. Reaching the trapping point, the particle is trapped with unit probability. The initial distribution is the equilibrium distribution of the harmonic potential. The stiffness of the potential is decreased from $k = K$ to 0 at a constant rate between $t = 0$ and $\tau/2$, and then increased from 0 to $K$ at a constant rate between $t = \tau/2$ to $\tau$. Let us denote by $p_{\text{trap}}$ the trapping probability of the final state and set the reference probability to the final probability of the process. Then the fluctuation equality (19) reduces to $\langle e^{-\Delta s^{\text{tot}}} \rangle = 1 - p_{\text{trap}}$, and the corresponding inequality is $\langle \Delta s^{\text{tot}} \rangle \geq - \ln(1 - p_{\text{trap}})$. Both formulae are consistent with the numerical simulations shown in Fig. 2.

Conclusion. By Lebesgue’s decomposition, the nonequilibrium fluctuation equalities are derived under more general condition than the conventional ones. This generalization extends the physical applicability of the fluctuation theorems. In some cases, the inequalities derived from our equalities are stronger than the conventional second law of thermodynamics. Our equalities are verified by the numerical simulations of the Langevin systems.

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