The Second Law For the Transitions Between the Non-equilibrium Steady States

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We show that the system entropy change for the transitions between non-equilibrium steady states arbitrarily far from equilibrium for any constituting process is given by the relative entropy of the distributions of these steady states. This expression is then shown to relate to the dissipation relations of both Vaikuntanathan and Jarzynski [EPL 87, 60005 (2009)] and Kawai, Parrondo and Van den Broeck [Phys. Rev. Lett. 98, 080602 (2007)] in the case of energy-conserving driving.

PACS numbers: 05.70.Ln, 05.20.-y, 05.40.-a

I. INTRODUCTION

Recently there have been many novel approaches in understanding physical systems driven away from equilibrium. Among such approaches we emphasize the work theorems of Jarzynski [1] and Crooks [2], steady-state thermodynamics [3, 4] and stochastic thermodynamics [5–8]. In this very same context, a new and profound understanding of the second law of thermodynamics has been possible through its relation to the dissipation in phase space [9–11], the relation between dissipation and lag in irreversible processes [12], and the Landauer principle and the second law [13].

An important progress concerning the second law has recently been made by Esposito and Van den Broeck [14–16] where they have shown that the total entropy production stems from adiabatic and non-adiabatic contributions. Although both of these entropy productions are non-negative, they are very different in nature, since these two distinct contributions represent two different manners of driving a system out of equilibrium. The adiabatic entropy production in a physical system occurs when the system is driven through non-equilibrium boundary conditions. It should be noted that this use of the term adiabatic does not refer to the absence of heat exchange, but to the instantaneous relaxation to the steady state as remarked in [15]. On the other hand, the non-adiabatic entropy production stems from the external driving with non-energy-conserving dynamics. Esposito and Van den Broeck have further shown that the system entropy production is the non-adiabatic entropy production minus the excess term [15].

In a related context, Speck and Seifert [17] have recently shown that the equilibrium form of the fluctuation-dissipation theorem (FDT) can be used for a colloidal particle in a periodic potential if one measures the velocity with respect to the local mean velocity. We also note that a modified fluctuation-dissipation relation for a non-equilibrium steady state has recently been experimentally verified [18]. Speck and Seifert end their discussion by asking whether some other non-velocity-like concepts can be used in their equilibrium form when one studies non-equilibrium steady states.

Considering the colloidal particle in a periodic potential studied by Speck and Seifert [17], we note that this model is particular in that its average excess heat is zero. Motivated by this observation, we aim to show that in this contribution the equilibrium form of the second law can be used for the transitions between the non-equilibrium steady states as Speck and Seifert have shown the restoration of the equilibrium FDT for the same class of transitions. However, it is worth remarking that our treatment is general and not limited only to the Brownian particles in periodic potential.

The paper is organized as follows: In the next section, we derive the formula for the entropy change associated with the transitions between non-equilibrium steady states arbitrarily far from equilibrium. Next, examples are provided to clarify the use of the previously derived main equation. The relation of the present work to the entropy change for the transitions between the equilibrium states and to the previous works derived in the case of energy-conserving dynamics is then discussed. Finally, concluding remarks are presented.

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II. THEORY

In stochastic thermodynamics, the entropy of a physical system is given by its time-dependent Shannon entropy

$$S(t) = - \sum_{m} p_{m}(t) \ln p_{m}(t)$$  \hspace{1cm} (1)

where we set the Boltzmann constant equal to unity. The general framework can then be formulated by assuming a Markovian dynamics together with a local detailed balance condition [14–16]. However, in the present work, we focus only on the non-adiabatic contribution arising from the transitions between the non-equilibrium steady states. Therefore, we consider a system initially in a normalized steady state $p_{m}^{st}(0)$ corresponding to the initial value of the control parameter $\lambda_{i}$. The external driving is represented as usual by the change of the control parameter $\lambda_{i}$ to its final value $\lambda_{f}$. Assuming that a steady state is formed for fixed values of the control parameter after an asymptotically long time, a new normalized steady state distribution $p_{m}^{st}(T)$ is reached after a long time $T$. For the transitions between the non-equilibrium steady states [15], the change in the system entropy then reads

$$\Delta S = - \sum_{m} p_{m}^{st}(T) \ln p_{m}^{st}(T) + \sum_{m} p_{m}^{st}(0) \ln p_{m}^{st}(0)$$  \hspace{1cm} (2)

and the following relation is satisfied

$$\Delta S_{na} - \Delta S_{ex} = \Delta S$$  \hspace{1cm} (3)

where the excess entropy change is

$$\Delta S_{ex} = \sum_{m} \left( p_{m}^{st}(T) - p_{m}^{st}(0) \right) \ln p_{m}^{st}(T).$$  \hspace{1cm} (4)

The non-adiabatic entropy production is due to the external driving with non-energy-conserving dynamics, and the excess contribution is what is left of the total dissipation once the adiabatic entropy production is used to maintain the non-equilibrium steady state. It is seen from Eq. (3) that the system entropy change is not equal to the non-adiabatic entropy change due to the excess entropy contribution. For a Brownian particle in a periodic potential, the excess entropy change is zero, since the steady states in this model also correspond to the equilibrium distributions once they are reached [16, 17]. However, the excess change is not zero in general. Motivated by the particular model studied by Speck and Seifert [17], we now generally include the excess entropy term in the system entropy change for the transitions between the non-equilibrium steady states. This incorporation is necessary in order to relate our results to the recent formulae derived for the transitions between equilibrium states, since the latter are formulated in terms of the system entropy change only. Note that the inclusion of the excess term into the system entropy change also makes the latter equal to the non-adiabatic entropy change as can be seen from Eq. (3). To include the excess entropy term, we treat as if it is zero and consider the result of this equality as a condition to be satisfied by the system entropy change which reads

$$\sum_{m} p_{m}^{st}(T) \ln p_{m}^{st}(T) = \sum_{m} p_{m}^{st}(0) \ln p_{m}^{st}(T)$$  \hspace{1cm} (5)

as can be easily seen from Eq. (4). Incorporating Eq. (5) into the change of the system entropy given by Eq. (2), we obtain

$$\Delta S^{st} = - \sum_{m} p_{m}^{st}(0) \ln p_{m}^{st}(T) + \sum_{m} p_{m}^{st}(0) \ln p_{m}^{st}(0)$$  \hspace{1cm} (6)

which can be rewritten as

$$\Delta S^{st} = \sum_{m} p_{m}^{st}(0) \ln \left( \frac{p_{m}^{st}(0)}{p_{m}^{st}(T)} \right),$$  \hspace{1cm} (7)
where we have denoted the system entropy change $\Delta S$ including the excess term by $\Delta S^{\text{ex}}$. Before proceeding further, one can check whether considering excess entropy term as zero in the form of a constraint and its inclusion in the system entropy change resulted any mathematical discrepancies. Note that one has initially the non-adiabatic entropy change equal to $\Delta S_{\text{na}} = \sum_{m} p_{m}^{\text{eq}}(0) \ln \left( \frac{p_{m}^{\text{eq}}(0)}{p_{m}^{\text{eq}}(T)} \right)$ as can be seen from Eqs. (2)-(4). Since we have obtained exactly the same expression in Eq. (7) for the system entropy change, it implies that the excess term is well incorporated into the system entropy without loss of generality.

We now note that the right hand side of Eq. (7) is the non-negative relative entropy expression $D \left[ p^{\text{eq}}(0) \parallel p^{\text{eq}}(T) \right]$ (also known as Kullback-Leibler distance) between the steady state distributions \[ i.e., \]

$$\Delta S = D \left[ p^{\text{eq}}(0) \parallel p^{\text{eq}}(T) \right] \geq 0 . \tag{8}$$

This is our main result, which relates the system (or, equivalently, non-adiabatic) entropy change due to the transitions between non-equilibrium steady states to the relative entropy of the initial and final steady state distributions. It shows that the equilibrium form of the second law is preserved if the excess entropy change is incorporated into the system entropy change. This can also be seen by inspecting the second law derived by Hatano and Sasa \[ i.e., \] $T \Delta S \geq -Q_{\text{ex}}$. This expression of the second law immediately gives our main result Eq. (8), once the excess heat $Q_{\text{ex}}$, being equal to $T \Delta S_{\text{ex}} = Q_{\text{ex}}$, is incorporated into the system entropy so that one now has $\Delta S^{\text{ex}} \geq 0$, since $\Delta S + \Delta S_{\text{ex}} = \Delta S^{\text{ex}}$. This is plausible, since the Hatano-Sasa form of the second law is derived for a system under nonconservative driving in contact with a single reservoir, and the excess entropy change is the same as the excess heat divided by the temperature of the reservoir under this condition.

On the other hand, it is worth noting that the result in Eq. (8) is more general than the one derived by Hatano and Sasa \[ i.e., \] the reason is that the system entropy change now possesses all the information of the non-adiabatic entropy change as a result of incorporating the excess entropy change as can be seen from Eq. (8). It is well-known that the non-adiabatic entropy change is related to the system properties, and independent of the constituting process \[ i.e., \] Therefore, our main result too, being exactly at the same level of description as the non-adiabatic entropy change, is independent of the process, which generates the steady state conditions, be it through the time-dependent driving or multiple reservoirs with different thermodynamic properties. However, the Hatano-Sasa relation is valid for the transitions between non-equilibrium steady states only under the assumption of time-dependent driving and coupling to a single reservoir. We also note that Eq. (8) is valid for any kind of transitions, be it slow or not.

Another related issue is concerning when the equality holds for the second law given by Eq. (8). In the case of the Hatano-Sasa relation, the equality holds only for slow processes so that one obtains $T \Delta S = -Q_{\text{ex}}$. Since the excess contribution is incorporated into the system entropy in Eq. (8), the Hatano-Sasa relation for slow processes becomes $\Delta S^{\text{ex}} = 0$. If the result of including excess entropy change in the system entropy change would be tantamount to rewriting the Hatano-Sasa relation, one would expect our main result i.e., Eq. (8) too to be zero for slow processes. However, being valid for arbitrary protocols and independent of the constituting process, the second law written in terms of the system entropy given by Eq. (8) is zero for two cases. i) The first trivial case is when the steady state distribution, once reached, remains unchanged despite the presence of the time-dependent driving. ii) The second case is nontrivial and a direct result of including the excess entropy term in the system entropy: the entropy change given by Eq. (8) is zero also when the steady state distribution does not change its form drastically. To understand this, consider a case where $p_{m}^{\text{eq}}(0) = \left( \frac{\alpha}{e^{\alpha m}} \right) e^{-\alpha m}$ and $p_{m}^{\text{eq}}(T) = \left( \frac{\delta}{e^{\delta m}} \right) e^{-\delta m}$ are initial and final normalized steady state distributions, respectively. The terms $\alpha$ and $\delta$ are constants which usually depend on the values of friction and diffusion, for example. Enforcing Eq. (8) i.e., incorporating the excess entropy change into the system itself, a simple calculation shows that $\alpha = \delta$, yielding zero system entropy change in Eq. (8). This provides a new insight into the physical meaning of slow processes regarding the transitions between non-equilibrium steady states. In other words, a process is slow only if the steady distribution remains the same despite the external driving or does not change its form drastically. One observes no entropy change in the physical system if an initial exponential steady state distribution is preserved exactly, or only changes its argument under the influence of external driving or due to the coupling of the physical system to multiple reservoirs with different thermodynamic properties. However, a transition from an exponential steady state distribution to a Gaussian one produces a nonzero entropy change as we see below for the Van der Pol oscillator under the influence of noise.

One might ask whether our central result Eq. (8) provides information on the entropy change when the physical system is acted on by an energy-conserving and conservative driving (or considering the generality of Eq. (8), one can have equilibrium states by keeping the thermodynamic properties of the multiple reservoirs same), so that the steady states are replaced by the equilibrium states in Eq. (8). In fact, all the steps above can be repeated in their full generality only by noting that the non-equilibrium steady states are replaced by the equilibrium ones so that one obtains the relation $\Delta S^{\text{eq}} = D \left[ p^{\text{eq}}(0) \parallel p^{\text{eq}}(T) \right] \geq 0$ as the second law for the transitions between initial and final equilibrium distributions, where the equality is satisfied for quasi-static processes.
In order to see that this is in fact the case, we consider the non-equilibrium Landauer principle in Ref. [13]. This principle in its full generality reads

$$\beta W_{diss} - D[p(T) \| p^{eq}(T)] + D[p(0) \| p^{eq}(0)] = \Delta_i S,$$

where $\beta$ is the inverse temperature. The dissipated work $W_{diss}$ is given by $\langle W \rangle - \Delta F^{eq}$, where $\langle W \rangle$ and $\Delta F^{eq}$ stand for the average work and free energy difference between the equilibrium states, respectively. In the equation above, the relative entropy terms are separately zero if the system is both initially and finally at equilibrium [13]. Moreover, $\Delta_i S$ is total entropy change and becomes equal to $\Delta S_{na}$, since the adiabatic contribution $\Delta S_a$ vanishes for a single reservoir with conservative driving or for multiple reservoirs with the same thermodynamical properties due to the local detailed balance condition [14–16]. Moreover, since the excess entropy change is incorporated into the system with conservative driving or for multiple reservoirs with the same thermodynamical properties due to the external driving, the steady states now relax to the equilibrium states (see e.g. the paragraph above Eq. (5) in Ref. [20]) so that we have

$$\beta W_{diss} = \Delta S^{eq} = D[p^{eq}(0) \| p^{eq}(T)] \geq 0.$$  \hspace{1cm} (10)

We note that $W_{diss} \geq 0$ is indeed the second law of thermodynamics, and the dissipated work too, being explicitly equal to $\langle W \rangle - \Delta F^{eq}$, relates only two equilibrium states just like the relative entropy expression $D[p^{eq}(0) \| p^{eq}(T)]$. Moreover, this equality also ensures that the relative entropy formulation of the second law attains zero only for quasistatic, reversible processes, since only then the dissipated work vanishes. By Eq. (10), one can further see why our main result Eq. (8) has been interpreted as preserving the equilibrium form of the second law.

Although we have only considered transitions between equilibrium states above, one can use the non-equilibrium Landauer principle [13] to include, for example, the transition from an initial equilibrium state to a final non-equilibrium one. Hence, one can write Eq. (9) as

$$\Delta S^{eq} - D[p(T) \| p^{eq}(T)] = \Delta_i S,$$ \hspace{1cm} (11)

where the term $D[p(0) \| p^{eq}(0)]$ is zero, since the system is initially at equilibrium. One has to keep the right hand side of the equation above as $\Delta_i S$, since the final state is not at equilibrium any more. In other words, one does not consider only the transitions between the equilibrium states, and consequently boundary terms must be included too (see in particular Eqs. (37-39), and (42) in Ref. [13]). Therefore, $\Delta_i S$ cannot be equal to $\Delta S^{eq}$ in general as we had in Eq. (10). However, the total entropy $\Delta_i S$ is always non-negative [13] i.e., $\Delta_i S \geq 0$, implying

$$\Delta S^{eq} \geq D[p(T) \| p^{eq}(T)].$$ \hspace{1cm} (12)

This expression was recently obtained by Vaikuntanathan and Jarzynski [12] for the relation between the dissipation and the lag where they have used $\beta W_{diss}$ instead of its equal $\Delta S^{eq}$ (see Eq. (10) above). Finally, following Ref. [12] (see in particular the section below Eq. (10) in Ref. [12]), one can write Eq. (12) as

$$\Delta S^{eq} \geq D[p(T) \| \tilde{p}^{eq}(0)],$$ \hspace{1cm} (13)

where tilde denotes the reverse process so that the initial state of the system during the reverse process is the final equilibrium state i.e., $p^{eq}(T) = \tilde{p}^{eq}(0)$. This last relation was obtained by Kawai et al. [9, 10], and relates the dissipation to the time reversal asymmetry. Eq. (13), on the other hand, relates the change in equilibrium entropies to the time-reversed process.

### III. EXAMPLES

Before proceeding, we would like to note that the results of the previous section are valid in the continuous case even though our formalism has been discrete so far.

As an illustrative model, we first consider a driven Brownian particle on a circle [16, 17]

$$\dot{x} = u_t + \sqrt{2D} \xi,$$ \hspace{1cm} (14)
where \( u_t \) is the time dependent drift and \( D \) is the time independent diffusion constant, also assuming \( x \in [0,1] \). This model represents a colloidal particle moving in a periodic potential, and it is used to study the violation of the fluctuation-dissipation theorem in non-equilibrium steady states with external driving [17]. Note that the force is directly proportional to the drift term in stochastic thermodynamics (see Eq. (15) in Ref. [16]). The stationary solution for the driven Brownian particle is equal to unity for any value of the control parameter i.e. \( p_{st}(0) = p_{st}(T) = 1 \) [16,17]. In other words, once the Brownian particle has relaxed to this steady state with \( p_{st} = 1 \), it remains so despite any external driving. Our main result i.e., Eq. (3), yields zero entropy change for this model. Note that this result was also observed in Ref. [16] for the non-adiabatic entropy change using the Fokker-Planck formulation of the stochastic thermodynamics once the steady state distribution is reached, rendering the excess entropy contribution redundant from there on for the transitions between the non-equilibrium steady states.

A non-trivial example is the Van der Pol oscillator subject to noise. Then the Ito-Langevin type stochastic equation reads

\[
\dot{x} = v
\]

\[
\dot{v} + (a + bE) v + x = \eta(t)
\]

where \( a \) and \( b \) are the controllable linear and fixed nonlinear friction coefficients, respectively [21]. The term \( E \) denotes, setting the mass and the angular frequency equal to unity for simplicity, the energy of the oscillation i.e., \( E = \frac{1}{2} (v^2 + x^2) \). The random noise is defined to be Gaussian with the noise intensity \( \sqrt{2D} \). i.e., \( \langle \eta(t) \rangle = 0 \), \( \langle \eta(t) \eta(t') \rangle = 2Db(t-t') \). We assume that we can control the change in the linear frictional term i.e., \( a = \gamma - \lambda \) and \( \gamma \gg \frac{bE}{2} \) where \( \lambda \) denotes the control parameter and \( \gamma \) is the linear friction coefficient by default. The most general stationary solution of the noise-driven Van der Pol oscillator then reads

\[
p_{st}(\lambda) = \exp(-\frac{aE}{D} - \frac{1}{2} \frac{bE^2}{D})
\]

apart from the appropriate normalization [21]. From here on, we use the energy representation, since it is equivalent to the phase space integration for the simple harmonic oscillator case. We now assume that the physical system initially described by the stationary distribution \( p_{st}(0) \) with zero value of the control parameter evolves into \( p_{st}(T) \) with \( \lambda_B = \gamma \) through a protocol controlled by an external agent. The initial stationary distribution corresponding to \( \lambda = 0 \) is given by

\[
p_{st}(0) = \frac{\gamma}{D} \exp\left(-\frac{\gamma E}{D}\right),
\]

where we have used \( \gamma \gg \frac{bE}{2} \). The final steady state with \( \lambda = \gamma \) reads

\[
p_{st}(T) = \sqrt{\frac{2b}{\pi D}} \exp\left(-\frac{b}{2D}E^2\right)
\]

The left hand side of Eq. (5) can now be calculated as \( \left(\frac{1}{2} \ln \left(\frac{2b}{\pi D}\right) \right) \), whereas the right hand side of the same equation yields \( \left(\frac{1}{2} \ln \left(\frac{2b}{\pi D}\right) - \frac{b}{2D}E^2\right) \). Therefore, one explicitly obtains from Eq. (5)

\[
\frac{\gamma^2}{2Db} = 1.
\]

Incorporating the relation above into Eq. (8) is tantamount to including the excess entropy change into the system entropy. Eq. (8) by itself explicitly yields

\[
\Delta S = \int_0^\infty dE \frac{\gamma}{D} \exp\left(-\frac{\gamma}{D}E\right) \ln \left[ \sqrt{\frac{2b}{\pi D}} \exp\left(-\frac{b}{2D}E^2\right) \right],
\]
which, after integration, becomes

\[ \Delta S = \ln \left( \sqrt{\pi \gamma^2 / 2Db} \right) - 1 + \frac{Db}{\gamma^2}. \]  (21)

Including the relation given by Eq. (19) which is tantamount to including the excess entropy change in the system entropy, we obtain

\[ \Delta S^{st} = \ln \left( \sqrt{\pi} \right) - 1 + \frac{1}{2}, \]  (22)

which finally yields

\[ \Delta S^{st} = D \left( p^st (0) \parallel p^st (T) \right) \approx 0.07, \]  (23)

which is indeed greater than zero, thereby indicating the irreversibility of the transition between these two nonequilibrium steady states. The smallness of this value is expected, since the departure from the compared steady state with zero control parameter is given with respect to the steady state corresponding to \( \lambda_B = \gamma \), where \( \gamma \) is itself supposed to be small in all realistic cases.

### IV. CONCLUSIONS

To summarize, the system entropy change for the transitions between non-equilibrium steady states arbitrarily far from equilibrium is obtained in terms of the relative entropy of the concomitant steady state distributions. This result is independent of the constituting process in the sense that steady states can result either due to the non-conservative driving or through the presence of multiple reservoirs with different thermodynamic properties. We also note that the same expression for the transition between non-equilibrium steady states i.e., Eq. (23), can be used for the transitions between equilibrium states only by replacing the stationary distributions with the corresponding equilibrium ones. Considering only the transitions between the equilibrium states, our result given by Eq. (10) implies the relations obtained in Refs. 9, 10, 12. However, these previous relations considered the dissipated work as a measure of the second law while we have related them to the entropy change between the equilibrium states through relative entropy expression.

It is worth noting that one should not confuse the main result of this paper given by Eq. (8) with the well-known similar expression \( D \left[ p(t) \parallel p^{st} (\lambda) \right] \) [15]. This expression can be considered as a proof of convergence to steady state and relates the actual and the corresponding steady state distributions, whereas Eq. (8) on the other hand relates two distinct steady states. In this context, we also note that a new approach has recently been introduced by defining a novel state function information free energy which also includes the adiabatic term in non-equilibrium thermodynamics [22].

Finally, we remark that the results outlined in this work can be experimentally tested e.g., by using the Van der Pol oscillator studied in this work, or a motor protein coupled to an ATP-regenerating system such that the motor protein forms a non-equilibrium steady state [23].

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