Entropy Production along a Stochastic Trajectory and an Integral Fluctuation Theorem

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For stochastic nonequilibrium dynamics like a Langevin equation for a colloidal particle or a master equation for discrete states, entropy production along a single trajectory is studied. It involves both genuine particle entropy and entropy production in the surrounding medium. The integrated sum of both $\Delta s_{\rm tot}$ is shown to obey a fluctuation theorem $\langle \exp[-\Delta s_{\rm tot}]\rangle = 1$ for arbitrary initial conditions and arbitrary time-dependent driving over a finite time interval.

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Introduction.—Can the notions appearing in the first and second law of thermodynamics consistently be applied to mesoscopic nonequilibrium processes like dragging a colloidal particle through a viscous fluid [1–4]? Concerning the first law, Sekimoto interpreted the terms in the standard overdamped Langevin equation as dynamical increments for applied work, internal energy, and dissipated heat [5]. For the second law and, in particular, entropy, a proper formulation and interpretation is more subtle. Entropy might be considered as an ensemble property and therefore not to be applicable to a single trajectory. On the other hand, the so-called fluctuation theorem [6–17] quite generally relates the probability of entropy generating trajectories to those of entropy annihilating ones which requires obviously a definition of entropy on the level of a single trajectory. While for a colloidal particle immersed in a heat bath it is pretty clear what the entropy change of the bath is, it is less obvious whether or not one should assign an entropy to the particle itself as well.

The purpose of this Letter is to show that consequent adaption of a previously introduced stochastic entropy [11,18] to the trajectory of a colloidal particle together with the present original discussion of its equation of motion yields a consistent interpretation of entropy production along a single stochastic trajectory. Moreover, it leads to a lucid and concise identification of boundary terms in fluctuation relations. In fact, we show for arbitrary time-dependent driving that the total entropy production obeys an integral fluctuation theorem which is related to but different from Jarzynski's nonequilibrium work relation [19]. The present definition of entropy also implies that the known steady-state fluctuation theorem holds for finite times rather than in the long-time limit only as previously in stochastic dynamics [9,10]. In a final step, this approach is generalized to arbitrary driven stochastic dynamics governed by a master equation with time-dependent rates.

Entropy along a trajectory.—As a paradigm, we consider overdamped motion $x(\tau)$ of a particle with mobility μ along a one-dimensional coordinate in the time-interval $0 \le \tau \le t$ subject to a force

$$F(x, \lambda) = -\partial_x V(x, \lambda) + f(x, \lambda). \tag{1}$$

This force can arise from a conservative potential $V(x, \lambda)$ and/or be applied to the particle directly as $f(x, \lambda)$. Both sources may be time dependent through an external control parameter $\lambda(\tau)$ varied according to some prescribed experimental protocol from $\lambda(0) \equiv \lambda_0$ to $\lambda(t) \equiv \lambda_t$. The motion is governed by the Langevin equation

$$\dot{x} = \mu F(x, \lambda) + \zeta, \tag{2}$$

with stochastic increments modeled as Gaussian white noise with $\langle \zeta(\tau)\zeta(\tau')\rangle = 2D\delta(\tau - \tau')$, where D is the diffusion constant. In equilibrium, D and μ are related by the Einstein relation $D = T\mu$, where T is the temperature of the surrounding medium. We assume this relation to persist even in a nonequilibrium situation. Throughout the Letter we set Boltzmann's constant to unity such that entropy becomes dimensionless.

For a definition of entropy along the trajectory, we consider first the corresponding Fokker-Planck equation for the probability $p(x, \tau)$ to find the particle at x at time τ as

$$\partial_\tau p(x, \tau) = -\partial_x j(x, \tau) = -\partial_x(\mu F(x, \lambda) - D\partial_x)p(x, \tau). \tag{3}$$

This partial differential equation must be augmented by a normalized initial distribution $p(x, 0) \equiv p_0(x)$. It will become crucial to distinguish the dynamical solution $p(x, \tau)$ of this Fokker-Planck equation, which depends on this given initial condition, from the solution $p^s(x, \lambda)$ for which the right-hand side of Eq. (3) vanishes at any fixed λ. The latter corresponds either to a steady state for $f \neq 0$ or to equilibrium for $f = 0$, respectively.

The common definition of a nonequilibrium Gibbs entropy

$$S(\tau) \equiv -\int dx\, p(x, \tau) \ln p(x, \tau) \equiv \langle s(\tau)\rangle \tag{4}$$

suggests to define a trajectory-dependent entropy for the particle (or "system")

$$s(\tau) = -\ln p(x(\tau), \tau), \tag{5}$$

where the probability $p(x, \tau)$ obtained by solving the Fokker-Planck equation is evaluated along the stochastic trajectory $x(\tau)$. Obviously, for any given trajectory $x(\tau)$, the entropy $s(\tau)$ depends on the given initial data $p_0(x)$ and thus contains information on the whole ensemble. For an equilibrium Boltzmann distribution at fixed λ, this definition assigns an entropy

$$s(x) = [V(x, \lambda) - \mathcal{F}(\lambda)]/T, \tag{6}$$

with the free energy $\mathcal{F}(\lambda) \equiv -T \ln \int dx \exp[-V(x, \lambda)/T]$. The definition (5) has been used previously by Crooks for stochastic microscopically reversible dynamics [11] and by Qian for stochastic dynamics of macromolecules [18]. Neither work, however, discusses the equation of motion for this stochastic entropy.

Entropy production.—The rate of change of the entropy of the system (5) is given by

$$\begin{aligned} \dot{s}(\tau) &= -\frac{\partial_\tau p(x, \tau)}{p(x, \tau)}\bigg|_{x(\tau)} - \frac{\partial_x p(x, \tau)}{p(x, \tau)}\bigg|_{x(\tau)} \dot{x} \\ &= -\frac{\partial_\tau p(x, \tau)}{p(x, \tau)}\bigg|_{x(\tau)} + \frac{j(x, \tau)}{D p(x, \tau)}\bigg|_{x(\tau)} \dot{x} \\ &\quad - \frac{\mu F(x, \lambda)}{D}\bigg|_{x(\tau)} \dot{x}. \end{aligned} \tag{7}$$

The first equality identifies the explicit and the implicit time dependence. The second one uses the Fokker-Planck Eq. (3) for the current. The third term in the second line can be related to the rate of heat dissipation in the medium

$$\dot{q}(\tau) = F(x, \lambda)\dot{x} \equiv T \dot{s}_{\mathrm{m}}(\tau), \tag{8}$$

where we identify the exchanged heat with an increase in entropy of the medium s_{m} at temperature $T = D/\mu$. Then (7) can be written as a balance equation for the trajectory-dependent total entropy production

$$\begin{aligned} \dot{s}_{\mathrm{tot}}(\tau) &= \dot{s}_{\mathrm{m}}(\tau) + \dot{s}(\tau) \\ &= -\frac{\partial_\tau p(x, \tau)}{p(x, \tau)}\bigg|_{x(\tau)} + \frac{j(x, \tau)}{D p(x, \tau)}\bigg|_{x(\tau)} \dot{x}, \end{aligned} \tag{9}$$

which is our first central result. The first term on the right-hand side signifies a change in $p(x, \tau)$ which can be due to a time-dependent $\lambda(\tau)$ or, even at fixed λ, due to relaxation from a nonstationary initial state $p_0(x) \neq p^s(x, \lambda_0)$.

Upon averaging, the total entropy production rate $\dot{s}_{\mathrm{tot}}(\tau)$ has to become positive as required by the second law. This ensemble average proceeds in two steps. First, we average over all trajectories which are at time τ at a given x leading to

$$\langle \dot{x} | x, \tau \rangle = j(x, \tau)/p(x, \tau). \tag{10}$$

Second, with $\int dx \partial_\tau p(x, \tau) = 0$ due to probability conservation, averaging over all x with $p(x, \tau)$ leads to

$$\dot{S}_{\mathrm{tot}}(\tau) \equiv \langle \dot{s}_{\mathrm{tot}}(\tau) \rangle = \int dx \frac{j(x, \tau)^2}{D p(x, \tau)} \geq 0, \tag{11}$$

where equality holds in equilibrium only. Averaging the increase in entropy of the medium along similar lines leads to

$$\dot{S}_{\mathrm{m}}(\tau) \equiv \langle \dot{s}_{\mathrm{m}}(\tau) \rangle = \langle F(x, \tau)\dot{x} \rangle / T \tag{12}$$

$$= \int dx F(x, \tau) j(x, \tau)/T. \tag{13}$$

Hence upon averaging, the increase in entropy of the system itself becomes $\dot{S}(\tau) \equiv \langle \dot{s}(\tau) \rangle = \dot{S}_{\mathrm{tot}}(\tau) - \dot{S}_{\mathrm{m}}(\tau)$. On the ensemble level, this balance equation for the averaged quantities has previously been derived directly from the ensemble definition (4) [18]. The key point of our approach is that we have defined entropy production (or annihilation) along a single stochastic trajectory splitting it up into a medium part and a part of the particle (system). Beyond the conceptual advantage, this identification facilitates a discussion of fluctuation theorems.

Fluctuation theorem.—Fluctuation theorems derive from the behavior of the weight of a trajectory under "time reversal" which associates with each protocol $\lambda(\tau)$ a reversed one $\tilde{\lambda}(\tau) \equiv \lambda(t - \tau)$ and a reversed trajectory $\tilde{x}(\tau) \equiv x(t - \tau)$. For a given initial value $x_0 \equiv x(0) = \tilde{x}(t) \equiv \tilde{x}_t$ and final value $x_t \equiv x(t) = \tilde{x}(0) \equiv \tilde{x}_0$, the ratio of probabilities of the forward path $p[x(\tau)|x_0]$ and of the backward path $\tilde{p}[\tilde{x}(\tau)|\tilde{x}_0]$ can easily be calculated in the path integral representation of the Langevin equation as [9]

$$\ln \frac{p[x(\tau)|x_0]}{\tilde{p}[\tilde{x}(\tau)|\tilde{x}_0]} = \int_0^t F(x, \tau)\dot{x} d\tau / T = \Delta s_{\mathrm{m}}. \tag{14}$$

If this quantity is combined with arbitrary normalized distributions for initial and final value $p_0(x_0)$ and $p_1(\tilde{x}_0) = p_1(x_t)$, respectively, according to

$$R[x(\tau), \lambda(\tau); p_0, p_1] \equiv \ln \frac{p[x(\tau)|x_0] p_0(x_0)}{\tilde{p}[\tilde{x}(\tau)|\tilde{x}_0] p_1(\tilde{x}_0)} \tag{15}$$

$$= \Delta s_{\mathrm{m}} + \ln \frac{p_0(x_0)}{p_1(x_t)}, \tag{16}$$

one easily derives the integral fluctuation relation [14]

$$\begin{aligned} \langle e^{-R} \rangle &\equiv \sum_{x(\tau), x_0} p[x(\tau)|x_0] p_0(x_0) e^{-R} \\ &= \sum_{\tilde{x}(\tau), \tilde{x}_0} \tilde{p}[\tilde{x}(\tau)|\tilde{x}_0] p_1(\tilde{x}_0) = 1. \end{aligned} \tag{17}$$

Here, the average is over both initial values drawn from the (in principle arbitrary) initial distribution $p_0(x_0)$ and trajectories $x(\tau)$ determined by the noise history $\zeta(\tau)$. Since the normalized distribution $p_1(x)$ is arbitrary, there are, even for fixed $p_0(x)$, an infinity of choices for R which obey the constraint (17) and its implication $\langle R \rangle \geq 0$.

At least two choices of p_1 have physical meaning in the present context. First, for $p_1(x_t) = p(x, t)$ which is the solution of the Fokker-Planck equation for the given initial distribution $p_0(x_0)$, the definition (5) implies that the last term in (16) becomes the entropy change of the particle Δs along the trajectory. Hence (17) implies the integral fluctuation theorem

$$\langle e^{-\Delta s_{\rm tot}} \rangle = 1, \tag{18}$$

which is our second main result. This integral theorem for $\Delta s_{\rm tot}$ is truly universal since it holds for any kind of initial condition (not only for $p_0(x_0) = p^s(x_0, \lambda_0)$), any time dependence of force and potential, with (for $f = 0$) and without (for $f \neq 0$) detailed balance at fixed λ, and any length of trajectory t without the need for waiting for relaxation. Crucial for this universality is our identification of the boundary term in (16) as the change in entropy of the particle.

As a second important choice, for $f = 0$ and $p_{0,1}(x) = p^s(x, \lambda_{0,t}) = \exp[-V(x, \lambda_{0,t}) - \mathcal{F}(\lambda_{0,t})]$, one recovers Jarzynski's relation $\langle \exp[-w_d/T] \rangle = 1$ [19] since in this case

$$\begin{aligned} R &= \Delta s_{\rm m} + [V(x_t, \lambda_t) - V(x_0, \lambda_0) - \mathcal{F}(\lambda_t) + \mathcal{F}(\lambda_0)]/T \\ &= w_d/T, \end{aligned} \tag{19}$$

where w_d is the part of the work which is irreversibly lost as dissipated into the medium. The difference between the two choices for $p_1(x)$ is subtle but important. In the first case, the fluctuation theorem holds for the total entropy change along the trajectory evaluated at the very end of the protocol. For Jarzynski's relation, one takes the distribution corresponding to equilibrium at the final value of λ. The difference arises from relaxation of the system towards the final equilibrium state at constant λ_t which further increases the averaged entropy of the particle. In fact, $p_1(x) = p(x, t)$ is the one choice which leads to the smallest $\langle R \rangle$ among all possible $p_1(x)$.

For a steady state at constant λ and constant force $f \neq 0$ like for motion along a ring with periodic boundary conditions, by choosing $p_0(x) = p_1(x) = p^s(x)$ in (16), one obtains the stronger fluctuation relation [11,14]

$$p(-R)/p(R) = e^{-R}. \tag{20}$$

Since with the definition (5) the last term in (16) is again the change in entropy of the system Δs, the quantity R becomes the total entropy change $\Delta s_{\rm tot} = \Delta s_{\rm m} + \Delta s$. Hence, one recovers the fluctuation theorem for the total change in entropy as

$$p(-\Delta s_{\rm tot})/p(\Delta s_{\rm tot}) = e^{-\Delta s_{\rm tot}} \tag{21}$$

even for a finite length t of the trajectories. In contrast, previous derivations of this genuine fluctuation theorem within stochastic dynamics [9,10] hold in the long-time limit only since they implicitly ignore what we call Δs and consider only $\Delta s_{\rm m}$. Since the former is bounded for finite potentials, the latter will always win in the long run.

Generalizations.—It is obvious that the present discussion holds as well for systems with more than one degree of freedom obeying overdamped coupled Langevin equations. Rather than spelling out the notational details, we will now discuss a more general stochastic dynamics on a discrete set $\{n\}$ of states. Again, we aim at a consistent definition of an entropy along a trajectory without having available any *a priori* notion of heat contrary to the colloidal case above which facilitated the identification of entropy production in the medium there.

Let a transition between discrete states m and n occur with a rate $w_{mn}(\lambda)$, which depends on an externally controlled time-dependent parameter $\lambda(\tau)$. The master equation for the time-dependent probability $p_n(\tau)$ then reads

$$\partial_\tau p_n(\tau) = \sum_{m \neq n} [w_{mn}(\lambda) p_m(\tau) - w_{nm}(\lambda) p_n(\tau)]. \tag{22}$$

For a solution, an initial distribution $p_n(0)$ must be specified as well. As above, the system is driven externally from λ_0 to λ_t according to a protocol $\lambda(\tau)$. For any fixed λ, there is a steady state $p_n^s(\lambda)$ which may or may not obey detailed balance $p_n^s(\lambda) w_{nm}(\lambda) = p_m^s(\lambda) w_{mn}(\lambda)$.

A stochastic trajectory $n(\tau)$ starts at n_0 and jumps at times τ_j from n_j^- to n_j^+ ending up at n_t. As entropy along this trajectory, we define

$$s(\tau) \equiv -\ln p_{n(\tau)}(\tau), \tag{23}$$

where $p_{n(\tau)}(\tau)$ is the solution $p_n(\tau)$ of the master equation (22) for a given initial distribution $p_n(0)$ taken along the specific trajectory $n(\tau)$. As in the colloidal case, this entropy will depend on the chosen initial distribution.

The equation of motion for the system entropy $s(\tau)$ becomes

$$\dot{s}(\tau) = -\frac{\partial_\tau p_n(\tau)}{p_n(\tau)}\bigg|_{n(\tau)} - \sum_j \delta(\tau - \tau_j) \ln \frac{p_{n_j^+}(\tau_j)}{p_{n_j^-}(\tau_j)}. \tag{24}$$

The second term arises from the jumps at τ_j. The first term contributes along the time intervals during which the system remains in one state. For constant λ, this term is nonzero if the initial distribution $p_n(0)$ is not the stationary one $p_n^s(\lambda)$. For a time-dependent $\lambda(\tau)$, the entropy changes during these intervals due to the time dependence of $p_n(\tau)$. We now split up the right-hand side of (24) into a total entropy production and one of the medium as follows

$$\dot{s}_{\rm tot}(\tau) \equiv -\frac{\partial_\tau p_n(\tau)}{p_n(\tau)}\bigg|_{n(\tau)} - \sum_j \delta(\tau - \tau_j) \ln \frac{p_{n_j^+} w_{n_j^+ n_j^-}}{p_{n_j^-} w_{n_j^- n_j^+}} \tag{25}$$

and

$$\dot{s}_{\rm m}(\tau) \equiv -\sum_j \delta(\tau - \tau_j) \ln \frac{w_{n_j^+ n_j^-}}{w_{n_j^- n_j^+}} \tag{26}$$

such that the balance $\dot{s}_{\rm tot}(\tau) = \dot{s}(\tau) + \dot{s}_{\rm m}(\tau)$ holds [20]. Here, and in the remainder, we suppress notationally the time dependence of both $p_n(\tau)$ and the rates $w_{nm}(\tau)$ in the jump terms.

The rational behind the identification (26) for the increase in entropy of the medium becomes clear after averaging over many trajectories. For this average, we need the probability for a jump to occur at $\tau = \tau_j$ from n_j^- to n_j^+ which is $p_{n_j^-}(\tau_j) w_{n_j^- n_j^+}(\tau_j)$. Hence, one gets

$$\dot{S}_{\rm m}(\tau) \equiv \langle \dot{s}_{\rm m}(\tau) \rangle = \sum_{n,k} p_n w_{nk} \ln \frac{w_{nk}}{w_{kn}}, \tag{27}$$

$$\dot{S}_{\rm tot}(\tau) \equiv \langle \dot{s}_{\rm tot}(\tau) \rangle = \sum_{n,k} p_n w_{nk} \ln \frac{p_n w_{nk}}{p_k w_{kn}}, \tag{28}$$

and

$$\dot{S}(\tau) \equiv \langle \dot{s}(\tau) \rangle = \sum_{n,k} p_n w_{nk} \ln \frac{p_n}{p_k} \tag{29}$$

such that the global balance $\dot{S}_{\rm tot}(\tau) = \dot{S}_{\rm m}(\tau) + \dot{S}(\tau)$ with $\dot{S}_{\rm tot}(\tau) \geq 0$ is valid. By averaging our stochastic expressions, we thus recover and generalize established results for the nonequilibrium ensemble entropy balance available so far for the steady state only [10,21,22].

For the fluctuation theorems, the stochastic quantity R is derived from the probability $P[n(\tau)|n_0]$ of a trajectory $n(\tau)$ to occur under the protocol $\lambda(\tau)$ and the probability $\tilde{P}[\tilde{n}(\tau)|\tilde{n}_0]$ for the reversed trajectory $\tilde{n}(\tau) \equiv n(t-\tau)$ to occur under the reversed protocol $\tilde{\lambda}(\tau) \equiv \lambda(t-\tau)$. With an arbitrary initial distribution p_n^0 and an arbitrary final distribution p_n^1 it becomes

$$R[n(\tau), \lambda(\tau); p_n^0, p_n^1] \equiv \ln \frac{P[n(\tau)|n_0] p_{n_0}^0}{\tilde{P}[\tilde{n}(\tau)|\tilde{n}_0] p_{\tilde{n}_0}^1} = \Delta s_{\rm m} + \ln \frac{p_{n_0}^0}{p_{n_t}^1}. \tag{30}$$

From the infinity of possible fluctuation relations $\langle \exp[-R] \rangle = 1$, we choose two important ones. First, for $p_n^0 = p_n^s(\lambda_0)$ and $p_n^1 \equiv p_n(t)$, the last term in (30) becomes the increase in system entropy and $R = \Delta s_{\rm tot}$ the total entropy change. Hence, we have again the integral theorem (18). Second, the choice of p_n^1 that corresponds to Jarzynski's relation in the colloidal case above is implied in the theorem derived in Ref. [23]. Finally, in a steady state for time-independent rates, by choosing $p_n^0 = p_n^1 = p_n^s$, one has the detailed version (21) for the total entropy change for any finite length of the trajectory as exemplified for a molecular motor or enzyme in Ref. [16].

Summarizing perspective.—We have expressed the entropy production along a single stochastic trajectory as a sum of an entropy production of the system and of the medium both for a colloidal particle and for general stochastic dynamics obeying a master equation. The total entropy production obeys an integral fluctuation theorem for arbitrary time-dependent driving, for arbitrary initial conditions, and any length of trajectories. This theorem and the Jarzynski relation are both shown to be special cases of an infinity of possible fluctuation relations. With the present definition of entropy, the detailed fluctuation theorem valid in steady states for the total entropy production holds even for trajectories of finite length.

The trajectory-dependent entropy of the particle could be measured experimentally for a time-dependent protocol by first recording over many trajectories the probability distribution $p(x, \tau)$ from which the entropy $s(\tau)$ of each trajectory can be inferred. With such data, one could also test the new integral fluctuation theorem (18) and compare it to Jarzynski's relation for the same protocol. It will be interesting to derive, both experimentally and theoretically, the probability distribution of these entropy changes and to see, e.g., whether they are Gaussian for slow driving as is the dissipated work appearing in Jarzynski's relation [24].

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