RAPID COMMUNICATIONS

PHYSICAL REVIEW E VOLUME 61, NUMBER 5 MAY 2000

Fluctuation theorems for entropy production in open systems

Lamberto Rondoni,[1] Tamás Tél,[2] and Jürgen Vollmer[3,*]
[1]Dipartimento di Matematica, Politecnico di Torino, Corso Duca degli Abruzzi 24, I-10129 Torino, Italy
[2]Institute for Theoretical Physics, Eötvös University, P.O. Box 32, H-1518 Budapest, Hungary
[3]Centre for Nonlinear Phenomena and Complex Systems, Université Libre de Bruxelles, Code Postal 231, B-1050 Brussels, Belgium
(Received 25 June 1999)

We derive a fluctuation theorem to describe entropy fluctuations in steady states of systems with density gradients due to open boundaries. The fluctuations are related to the growth rate of the phase-space density, instead of the phase-space contraction rate. Explicit derivations are presented for a multibaker map, but the arguments are rather general, and should hold for a much wider class of dynamical systems. A comparison with recent results for stochastic systems is also given.

PACS number(s): 05.70.Ln, 05.45.Ac, 05.20.−y, 05.40.−a

The connection between nonequilibrium statistical physics and chaotic microscopic dynamics has become a subject of vivid interest [1]. Particular attention [2–5] has been devoted to the observation [6] that in steady states there are fluctuating quantities, like the phase-space contraction rate [2,3] or the displacement of trajectories [4], whose average α along trajectory segments of fixed duration t occurs with a probability distribution Π_t fulfilling

$$\frac{1}{t}\ln\left[\frac{\Pi_t(\alpha)}{\Pi_t(-\alpha)}\right]=\alpha. \tag{1}$$

The Gallavotti-Cohen fluctuation theorem (GCFT) [3] states that for sufficiently large (in the sense of large deviation theory) t, Eq. (1) is a generic property of time-reversible, strongly chaotic systems, where α is the average σ of the local phase-space contraction rate $\sigma(x,p)$ along trajectory segments of length t. Here, (x,p) is a point in phase space. The physical relevance of the theorem is based on the identification of the average of $\sigma(x,p)$ with the rate of specific irreversible entropy production $\sigma^{(irr)}$ in steady states [7]. Then, σ is the contribution of trajectory segments of length t to the entropy production. In this sense, the GCFT extends the Onsager relations away from equilibrium, and yields the Green-Kubo relations for weak driving [8].

We note that although the concept of entropy often is not clearly defined in this context, the notion of entropy production is widely used. This apparent contradiction has been resolved in the framework of multibaker models [9–11] by introducing a *coarse-grained* (CG) entropy, which formally fulfills an entropy-balance equation in the form known from thermodynamics. Starting with the area-preserving example of Gaspard [9] explicit calculations [10–13] have revealed that the identification of the phase-space contraction with the irreversible entropy production is not justified in general.

Fluctuation theorems hold for steady states. We distinguish two classes of steady states: *macroscopic* steady states, characterized by stationary CG densities, i.e., stationary average densities in regions of macroscopically small extent; and the usual invariant measures of dynamical systems. For smooth initial distributions, the usual, or *exact*, phase-space density $\varrho_t(x,p)$ never stops evolving in time, while CG densities quickly reach their stationary values.

In macroscopic steady states of open systems with CG density gradients, the irreversible entropy production $\sigma^{(irr)}$ per particle was shown to be the average of the growth rate σ_ϱ of $\varrho_t(x,p)$ [11] (rather than σ). The quantity σ_ϱ is defined by

$$\sigma_\varrho(x,p)\equiv\frac{1}{\tau}\ln\frac{\varrho_\tau(x,p)}{\varrho_0(x,p)}, \tag{2}$$

where τ is a short time interval. This *local* expression holds irrespective of boundary conditions. Besides phase-space contraction, σ_ϱ characterizes the mixing of regions with different macroscopic densities. Hence, it does not vanish in boundary-driven, volume-preserving systems. Only in systems with macroscopically homogeneous steady states $\sigma_\varrho(x,p)$ reduces to $\sigma(x,p)$ [see Eq. (4) with m-independent ϱ_m]. In the following, we show how the validity of Eq. (1) can be extended to time-reversible systems with open boundaries by taking $\alpha=\sigma_\varrho$. Subsequently, we discuss the connection between the GCFT derived on the basis of deterministic dynamics, and fluctuation theorems for stochastic processes [4].

Here, we consider multibaker maps, which model certain aspects of transport processes [9–11], even though their physical limitations are still a matter of discussion. Multibakers are appealing, being the simplest spatially extended dynamical systems with the possibility for a biased time evolution and nontrivial boundary conditions. At present they are the only analytically tractable models allowing us to explore the structure of fluctuation theorems for steady states of systems with density gradients. The phase space of the model consists of a strip of size $aN\times b$ in the (x,p) plane, which is divided into N identical cells of size $a\times b$ labeled by the index m. After each time unit τ, every cell is divided into three vertical columns (cf. Fig. 1): the rightmost (leftmost) column of width ra (la) of every cell is squeezed and stretched into a strip of width a and height $\tilde{r}b$ ($\tilde{l}b$) in the cell to the right (left). They are responsible for transport in one

*Present addresses: Fachbereich Physik, Universitat-GH Essen, D-45117 Essen, Germany and MPI for Polymer Research, Ackermannweg 9, D-55128 Mainz, Germany.

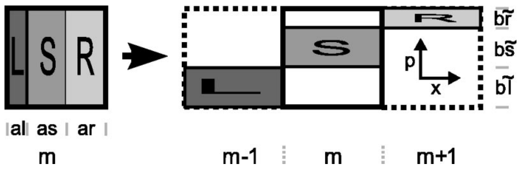

FIG. 1. Graphical illustration of the action of the multibaker map on a unit cell m. The three columns in the cell are uniformly squeezed and stretched leading to the shown deformation of the tags L, S, and R.

time step. The middle column of width sa remains in the cell, modeling motion that does *not* contribute to transport. It is mapped onto a strip of width a and height sb so that the internal dynamics is area preserving. Motivated by more general models, global phase-space conservation is required, which implies $s+l+r=1=s+\tilde{l}+\tilde{r}$.

The details of the transport process described by the model depend on the boundary conditions [11], since they determine the distribution of the CG density ϱ_m (i.e., the average density in every cell), which we assume to be known in the steady state [14,21]. Results that are consistent with thermodynamics are obtained in the *macroscopic limit* $a,\tau \to 0$, in which the density evolves according to an advection-diffusion equation with constant drift v and diffusion coefficient D. In taking the limit, the parameters r and l of the map scale with a and τ like

$$v=a(r-l)/\tau, \quad D=a^2(r+l)/(2\tau). \tag{3}$$

The total length of the chain is fixed to $L=aN$, and the physical time is $t=\tau n$.

In the multibaker model, the single time step contribution of a trajectory to the growth rate σ_ϱ of the phase-space density is [recall Eq. (2) and Fig. 1]

$$\begin{cases} \frac{1}{\tau}\ln\left[\frac{r\varrho_m}{\tilde{r}\varrho_{m+1}}\right] & \text{for a step from cell } m \text{ to } m+1, \\ 0 & \text{whenever it stays in cell } m, \\ \frac{1}{\tau}\ln\left[\frac{l\varrho_m}{\tilde{l}\varrho_{m-1}}\right] & \text{for a step from cell } m \text{ to } m-1. \end{cases} \tag{4}$$

Here ϱ_j denotes the CG density [15] in cell j, which is time-independent in a steady state. The density ratios enter Eq. (4) because of the mixing in cell m of different CG densities coming from the neighboring cells $m-1$, $m+1$. [For comparison, the phase-space contraction rate σ is like (4), but without the ϱ factors.] In the present paper, we only consider stationary CG density distributions ϱ_m, so that the expressions given in Eq. (4) can be interpreted as contributions to the entropy production [cf. Eq. (2) and Ref. [11]]. Consequently, the average entropy production of trajectories of length n, which make n_l (n_r) steps to the left (right) and are centered at the half-integer cell index μ, i.e., which start in cell $m=\mu-\Delta n/2$ and end in $\mu+\Delta n/2$, where $\Delta n \equiv n_r - n_l$ is the displacement, is

$$\sigma_\varrho^{(\mu)}(n_l,n_r)=\frac{1}{n\tau}\ln\left[\left(\frac{r}{\tilde{r}}\right)^{n_r}\left(\frac{l}{\tilde{l}}\right)^{n_l}\frac{\varrho_{\mu-\Delta n/2}}{\varrho_{\mu+\Delta n/2}}\right]. \tag{5}$$

It is independent of the details of the sequence of steps.

First, we choose $r=\tilde{l}$, $l=\tilde{r}$, which leads to a dissipative, time-reversal symmetric dynamics, as in thermostatted systems [10,11]. Later we drop this condition to study *improperly thermostated* multibaker models.

For the time-reversible model, the rate $\sigma_\varrho^{(\mu)}(n_l,n_r)$ only depends on n_l and n_r through Δn. Hence, the probability $\Pi_t^{(\mu)}(\sigma_\varrho)$ of finding a value $\sigma_\varrho \equiv \sigma_\varrho^{(\mu)}(\Delta n)$ for trajectory segments of length $t=n\tau$ centered at μ is

$$\Pi_t^{(\mu)}[\sigma_\varrho^{(\mu)}(\Delta n)]=\frac{\varrho_{\mu-\Delta n/2}}{\sum_{m=1}^{N}\varrho_m}\sum_{\substack{n_l,n_s,n_r\\ n_r+n_s+n_l=n\\ n_r-n_l=\Delta n}}\mathcal{N}_t^{(\mu)}(n_l,n_r) \times l^{n_l}s^{n_s}r^{n_r}, \tag{6}$$

with normalization $\Sigma_{\mu,\sigma_\varrho}\Pi_t^{(\mu)}(\sigma_\varrho)=1$ [16]. The first factor accounts for the probability that the trajectory segment starts in cell $\mu-\Delta n/2$, while the second factor is the probability to find a segment with displacement Δn; $\mathcal{N}_t^{(\mu)}(n_l,n_r)$ denotes the number of trajectories of length $n=t/\tau$, which are centered at μ, make n_l (n_r) steps to the left (right), and never leave the chain. Here, consistently with the piecewise-linear character of the multibaker dynamics, these probabilities are taken to be independent. Comparing trajectories with their time-reversed counterparts, we find that $\mathcal{N}_t^{(\mu)}(n_l,n_r)=\mathcal{N}_t^{(\mu)}(n_r,n_l)$. In particular, $\mathcal{N}_t^{(\mu)}(n_l,n_r)=n!/(n_l!\,n_s!\,n_r!)$, if all trajectories stay inside the chain.

To extend the validity of Eq. (1) to distributions like $\Pi_t^{(\mu)}$, observe that time-reversed trajectory segments produce the same entropy up to a change of sign, $\sigma_\varrho^{(\mu)}(\Delta n)=-\sigma_\varrho^{(\mu)}(-\Delta n)$, as seen by exchanging n_r and n_l in (5). Thus,

$$\frac{\Pi_t^{(\mu)}[\sigma_\varrho^{(\mu)}(\Delta n)]}{\Pi_t^{(\mu)}[\sigma_\varrho^{(\mu)}(-\Delta n)]}=\frac{\varrho_{\mu-\Delta n/2}\sum_{\substack{n_l,n_s,n_r\\ n_r+n_s+n_l=n\\ n_r-n_l=\Delta n}}\mathcal{N}_t^{(\mu)}(n_l,n_r)\,l^{n_l}s^{n_s}r^{n_r}}{\varrho_{\mu+\Delta n/2}\sum_{\substack{n_l,n_s,n_r\\ n_r+n_s+n_l=n\\ n_r-n_l=\Delta n}}\mathcal{N}_t^{(\mu)}(n_l,n_r)\,l^{n_r}s^{n_s}r^{n_l}}=\frac{\varrho_{\mu-\Delta n/2}}{\varrho_{\mu+\Delta n/2}}\left(\frac{r}{l}\right)^{\Delta n}. \tag{7}$$

After taking the logarithm of both sides, the right-hand side gives $t=n\tau$ times $\sigma_\varrho^{(\mu)}(\Delta n)$. Therefore, Eq. (7) constitutes a *local* fluctuation theorem [17], in the sense that it takes the form of Eq. (1) with $\alpha=\sigma_\varrho^{(\mu)}(\Delta n)$, but concerns trajectories of given center μ and finite length $n\tau$.

In order to obtain a *global* fluctuation theorem one needs the probability $\Pi_t(\sigma_\varrho)$ of finding a value σ_ϱ irrespective of the position of the trajectory. This is obtained by summing up the contributions of all $\Pi_t^{(\mu)}(\sigma_\varrho), \mu=1,\frac{3}{2},2,\frac{5}{2}, \ldots ,N$, and observing that μ contributes to $\Pi_t(\sigma_\varrho)$ if and only if there is a displacement Δn_μ with $\sigma_\varrho=\sigma_\varrho^{(\mu)}(\Delta n_\mu)$. Consequently,

$$\begin{aligned}\Pi_t(\sigma_\varrho)&=\sum_\mu \Pi_t^{(\mu)}[\sigma_\varrho^{(\mu)}(\Delta n_\mu)]\\ &=\sum_\mu \frac{\varrho_{\mu-\Delta n_\mu/2}}{\varrho_{\mu+\Delta n_\mu/2}}\left(\frac{r}{l}\right)^{\Delta n_\mu}\Pi_t^{(\mu)}[\sigma_\varrho^{(\mu)}(-\Delta n_\mu)]\\ &=\exp(t\sigma_\varrho)\Pi_t(-\sigma_\varrho),\end{aligned} \tag{8}$$

where we used Eqs. (5) and (7) with Δn_μ to obtain the second equality. We hereby have derived our key result: Eq. (1) holds with $\alpha=\sigma_\varrho$ in the stationary distribution ϱ_m of the time-reversible system. Moreover, the result does not depend on the particular choice of boundary conditions.

To recover continuum relations with a form known from thermodynamics, we take the macroscopic limit of the growth rate σ_ϱ. Provided the density difference between the two ends of trajectories is small, i.e., $\varrho_{\mu+\Delta n/2}-\varrho_{\mu-\Delta n/2} \ll \varrho_\mu$, we obtain

$$\sigma_\varrho(x;u)=\frac{uj}{\rho(x)D}, \tag{9}$$

where $u\equiv a\Delta n/t$ is the average velocity of the considered trajectory, $\rho(x)=b\varrho_m$ is the density at position $x=am$, and $j=v\rho(x)-D\partial_x\rho(x)$ denotes the current density, which is independent of x in a steady state. Since the expectation value of u is the local streaming velocity $j/\rho(x)$, the expectation value for the growth rate is $\sigma^{(irr)}(x)=j^2/[\rho(x)^2D]$. Thus, we formally recover the well-known relation between entropy production and Joule's heat by using (cf. [11]) the "Einstein relation" $\rho D=\sigma_{el}T$, where σ_{el} represents the electric conductivity and T the temperature.

Let us consider now the improperly thermostatted multibaker model (i.e., we no longer require $\tilde r=l$, $\tilde l=r$). The growth rate depends then on n_l and n_r separately [cf. Eq. (5)], while the probability $\Pi_t^{(\mu)}[\sigma_\varrho^{(\mu)}(n_l,n_r)]$ is unchanged. Since $\sigma_\varrho^{(\mu)}$ is now no longer a function of n_r-n_l only, it does no longer change sign when comparing a trajectory segment with its time-reversed image. Therefore, the argument leading to Eq. (7) no longer applies, and we cannot derive a relation like Eq. (1). We conclude, that fluctuation theorems for deterministic systems can only hold for time reversible dynamics.

Finally, we turn to a comparison with fluctuation theorems for systems with stochastic dynamics. In Ref. [4] Lebowitz and Spohn (LS) consider *jump processes* described by a master equation. It is assumed that the direct and the reversed transition probabilities between any two states are either both vanishing or both nonzero, but no assumption on time-reversal invariance is made. Their basic variable, the action functional $W(t)$, is proportional to the current of particles integrated over the time t. It is a function of solely the transition probabilities along the stochastic trajectory. For $t\to\infty$, $w=W/t$ becomes a continuous random variable whose probability distribution $\Pi_t(w)$ fulfills Eq. (1) with $\alpha=w$ [4].

In our setting W corresponds to a quantity proportional to the particle displacement Δn. Since the multibaker map is a discrete time model, we first write a large deviation theory for Δn in the spirit of [4], but keep the time unit τ finite, as considered in [18] for random walks without bias. The dynamics along the x axis is equivalent to a Markov process with transition rates $t_{j,j+1}=r$, $t_{j,j-1}=l$, and $t_{j,j}=s$, over time τ, i.e., it generates a biased random walk. The moment generating function $e(\lambda)$ of the random walk is defined via $\langle\exp(-a\lambda\Delta n)\rangle\sim\exp[-te(\lambda)]$ for large times t. Due to the translation invariance of the transition rates, it takes the form $e(\lambda)=-(1/\tau)\ln[l\exp(a\lambda)+s+r\exp(-a\lambda)]$. Note that $e(\lambda)$ only contains the transition rates and not the parameters $\tilde r,\tilde l$ of the multibaker. It characterizes the random walk along the x axis, but does not concern phase-space dynamics.

The probability to find an average velocity $u=a\Delta n/t$ along a trajectory of length t can be written as $\Pi_t(u)\sim\exp[-tg(u)]$ for large t. A saddle point approximation shows [4,18] that $g(u)$ is the Legendre transform of $e(\lambda)$. For the multibaker map one obtains

$$g(u)=-\frac{1}{\tau}\ln\left[\frac{a(R+as)}{a^2-(u\tau)^2}\right]-\frac{u}{a}\ln\left[\frac{R-s(u\tau)}{2l(a+u\tau)}\right], \tag{10}$$

where $R\equiv\{s^2(u\tau)^2+4lr[a^2-(u\tau)^2]\}^{1/2}$ [19]. In this case Eq. (1) with $\alpha=u$ only holds in a modified form: the right-hand side of Eq. (1) is no longer α but the more general expression $A=cu$ with $c=(1/a)\ln(r/l)$. Although for the variable $\alpha=w=W/t=cu$ considered in [4] this is of the form of Eq. (1) and $A=w$, the spirit is completely different: the fluctuating quantity w is not necessarily related to entropy production. Equation (10) is valid for every random walk, in particular for those described by one-dimensional maps, where the concept of phase-space contraction, needed to obtain the GCFT and Eq. (8), does not apply. Hence, the LS fluctuation theorem for the velocity u of an improperly thermostatted system is still of the modified form of Eq. (1) with $A=uc$, irrespective of the choice of $\tilde r$ and $\tilde l$. After all, the LS approach is based on the transition rates r and l only. Thus, the GCFT and the LS fluctuation theorem are different. Not even formally can they be identified for non time-reversible dynamical systems.

In conclusion, we obtained strong evidence that both local and global entropy-related fluctuation theorems hold also in macroscopically inhomogeneous steady states of time-reversible systems, and clarified the relation between fluctuation theorems for chaotic and stochastic systems. For deterministic dynamics these theorems should be based on the growth rate of the phase-space density. The essential new ingredients of our derivation are a contribution to the entropy production due to density gradients, and the assignment of an *a priori* probability for a trajectory segment to start at a given position. Although derived for a particular model, we believe that our results hold quite generally, since baker-type maps are known to be paradigms of chaotic systems [20]. Because of a slower convergence of more general models, the theorems can, however, only be expected to hold when evaluated for sufficiently long trajectory segments. Moreover, for open macroscopic systems of finite size L the time t should be limited from above by the typically much larger value L^2/D in order to ensure a representative sampling of phase space, i.e., to avoid escape of trajectories during observation. Altogether, this still implies that entropy related fluctuation theorems hold for a much wider class of steady states than previously thought. In particular, our version of the theorem also applies to area preserving systems (i.e., an unbiased, time-reversal symmetric dynamics) driven by appropriate nonequilibrium boundary conditions, where there is no phase-space contraction. The local formulation of the fluctuation theorem also allows us to focus on the finite time entropy production of a selected number of particles residing in a finite region of spatially extended systems.

We are grateful to F. Bonetto, E. G. D. Cohen, J. R. Dorfman, D. J. Evans, G. Gallavotti, L. Mátyás, and G. Nicolis for enlightening discussions. Support from GNFM-CNR (Italy), the EC (Contract No. ERBCHRXCT940460), the Hungarian Science Foundation (Grant No. OTKA T17493, T19483), the German-Hungarian D125 Cooperation, and the TMR network *Spatially Extended Dynamics*, is acknowledged.

[1] Chaos **8** (2) (1998), focus issue on chaos and irreversibility.

[2] D.J. Evans and D.J. Searles, Phys. Rev. E **50**, 1645 (1994); **52**, 5839 (1995); **53**, 5808 (1996).

[3] G. Gallavotti and E.G.D. Cohen, Phys. Rev. Lett. **74**, 2694 (1995); J. Stat. Phys. **80**, 931 (1995).

[4] J.L. Lebowitz and H. Spohn, J. Stat. Phys. **95**, 333 (1999).

[5] S. Lepri, R. Livi, and A. Politi, Physica D **119**, 140 (1998); N. Chernov and J.L. Lebowitz, J. Stat. Phys. **86**, 953 (1997); F. Bonetto, N. Chernov, and J.L. Lebowitz, Chaos **8**, 823 (1998).

[6] D.J. Evans, E.G.D. Cohen, and G.P. Morriss, Phys. Rev. Lett. **71**, 2401 (1993).

[7] $\sigma^{(irr)}$ differs from the rate of entropy production per unit volume only by a factor: the particle density.

[8] G. Gallavotti, Phys. Rev. Lett. **77**, 4334 (1996).

[9] P. Gaspard, J. Stat. Phys. **89**, 1215 (1997); *Chaos, Scattering and Statistical Mechanics* (Cambridge University Press, Cambridge, England, 1998).

[10] J. Vollmer, T. Tél, and W. Breymann, Phys. Rev. Lett. **79**, 2759 (1997).

[11] W. Breymann, T. Tél, and J. Vollmer, Chaos **8**, 396 (1998); J. Vollmer, T. Tél, and W. Breymann, Phys. Rev. E **58**, 1672 (1998).

[12] G. Nicolis and D. Daems, J. Phys. Chem. **100**, 19 187 (1996); C. Wagner, R. Klages, G. Nicolis, Phys. Rev. E **60**, 1401 (1999).

[13] E.G.D. Cohen and L. Rondoni, Chaos **8**, 357 (1998).

[14] A proper way to implement open boundary conditions is to consider the system as part of a chain of infinitely many cells. A normalized invariant measure can then be given on the phase space (a subset of the power set of the chain) starting from an invariant measure on the chain. This leads to a Poisson suspension on the original dynamical system [21]. The CG entropy is defined for the Poisson suspension, and differs from the usual CG Gibbs entropy only by a term negligible in the macroscopic limit (cf. [9]).

[15] Due to the piecewise linearity of the baker dynamics, the CG phase-space and the particle densities are proportional. We need not distinguish them here.

[16] This normalization is valid in the large system limit. Indeed, $\Sigma_{\sigma_\varrho}\Pi_t^{(\mu)}=\varrho_{\mu-\Delta n/2}/\Sigma_m\varrho_m$ for all μ, with $0\leqslant\mu-\Delta n/2\leqslant N$, making the correction to the normalization of Eq. (6) only a boundary effect. In any case, the normalization factor does not enter our calculations, and drops out in Eq. (7).

[17] This local fluctuation relation is of different nature than the one in G. Gallavotti, Physica A **263**, 39 (1999).

[18] P. Gaspard, in *From Phase Transitions to Chaos*, edited by G. Györgyi *et al.* (World Scientific, Singapore, 1991), p. 322.

[19] In the limit $\tau\to 0$, Eq. (10) becomes equivalent to example (A.4) of [4], showing that in the master equation limit the large deviation theory described here and that of LS are identical. If in addition $a\to 0$, Eq. (10) yields $g(u)=(u-v)^2/(4D)$.

[20] E. Ott, *Chaos in Dynamical Systems* (Cambridge University Press, Cambridge, England, 1993).

[21] I.P. Cornfeld, S.V. Fomin, and Ya.G. Sinai, *Ergodic Theory* (Springer-Verlag, Berlin, 1982).