Chaotic Hypothesis, Fluctuation Theorem and singularities

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Recently the "Fluctuation theorem" has been criticized and incorrect contents have been attributed to it. Here we reestablish and comment the original statements. We also discuss aspects of the extension of the chaotic hypothesis and of the fluctuation relation to singular systems.

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I. ANOSOV SYSTEMS

Fluctuations

Mathematically the *Fluctuation theorem* is a property of the phase space contraction of a *time reversible* Anosov map S, modeling a *time evolution.* The possible connection between the fluctuation theorem and Physics is a different matter that we will not discuss here: there are many places where this is attempted, [1–3].

We shall denote by Ω the *phase space* (a smooth finite boundaryless Riemannian manifold) and by $\sigma(x)$ the volume *contraction*

$$\sigma(x) = -\log|\det \partial_x S(x)| \tag{1}$$

Time reversal is defined as an isometry $I:\Omega\longleftrightarrow\Omega$ with

$$IS = S^{-1}I, \qquad \sigma(Ix) = -\sigma(x) \tag{2}$$

For Anosov maps, existence of a unique invariant probability distribution μ, called the *SRB distribution* and describing the long–time statistics of the motions whose initial data are chosen randomly with respect to the volume measure, is established, [4, 5]. It has the property that, with the exception of points $x\in\Omega$ in a set of 0–volume, it is

$$\lim_{\tau\to\infty}\frac{1}{\tau}\sum_{t=0}^{\tau-1}F(S^t x)\stackrel{def}{=}\langle F\rangle=\int_\Omega F(y)\mu(dy) \tag{3}$$

for all smooth observables F defined on phase space.

It is intuitive that "phase space cannot expand"; this is expressed by the following result of Ruelle, [6],

Proposition: *If* $\sigma_+\stackrel{def}{=}\langle\sigma\rangle$ *it is* $\sigma_+\ge 0$

Clearly if S is volume preserving it is $\sigma_+=0$. If $\sigma_+>0$ the system does not admit any stationary distribution of the form $\mu(dx)=\rho(x)dx$, with density with respect to the volume measure dx (often called *absolutely continuous* with respect to the volume).

This motivates calling systems for which $\langle\sigma\rangle>0$ *dissipative* and *conservative* the others.

For Anosov systems which are *transitive* (*i.e.* with a dense orbit), reversible and dissipative one can define the dimensionless phase space contraction, a quantity often related to entropy creation rate (see [7]), averaged over a time interval of size τ. This is

$$p(x)=\frac{1}{\sigma_+\tau}\sum_{-\tau/2}^{\tau/2-1}\sigma(S^k x) \tag{4}$$

provided *of course* $\sigma_+>0$.

Then for such systems the probability with respect to the stationary state, *i.e.* to the SRB distribution μ, that the variable $p(x)$ takes values in $\Delta=[p,p+\delta p]$ can be written as $\Pi_\tau(\Delta)=e^{\tau\max_{p\in\Delta}\zeta(p)+O(1)}$, where $\zeta(p)$ is a suitable function and, for any fixed choice of Δ contained in an open interval $(-p^*,p^*)$, $p^*\ge1$, the correction term at the exponent is $O(1)$ with respect to τ^{-1}, as $\tau\to\infty$ (this is often informally expressed as $\lim_{\tau\to\infty}\frac{1}{\tau}\log\Pi_\tau(p)=\zeta(p)$ for $-p^*<p<p^*$). The function $\zeta(p)$ is called in probability theory the *rate function* for the large deviations of p.

The function $\zeta(p)$ is analytic in p and *convex* in the interval of definition $(-p^*,p^*)$. Analyticity and convexity of large deviation rates are general properties, established by Sinai and valid for the SRB-averages of smooth observables (in Anosov systems), [5, 8, 9]. In fact more can be said for the specific case of the large deviation rate of the observable p, and one can prove the following *fluctuation theorem*:

Proposition: *In transitive time reversible dissipative Anosov systems the rate function* $\zeta(p)$ *for the dimensionless phase space contraction* $p(x)$ *defined in (4) is analytic and strictly convex in an interval* $(-p^*,p^*)$ *with* $+\infty>p^*\ge1$ *and* $\zeta(p)=-\infty$ *for* $|p|>p^*$. *Furthermore*

$$\zeta(-p)=\zeta(p)-p\sigma_+, \qquad \text{for} \qquad |p|<p^* \tag{5}$$

which is called the "fluctuation relation".

Strict convexity follows from a theorem of Griffiths and Ruelle which shows that the only way strict convexity could fail is if $\sigma(x)=\varphi(Sx)-\varphi(x)+c$ where

$\varphi(x)$ is a smooth function (typically a Lipschitz continuous function) and c is a constant, see propositions (6.4.2) and (6.4.3) in [5]. The constant c vanishes if time reversal holds and $\sigma(x) = \varphi(Sx) - \varphi(x)$ contradicts the assumption that $\sigma_+ > 0$, because $\tau^{-1}\sum_{-\tau/2}^{\tau/2-1}\sigma(S^k x) = \tau^{-1}\big[\varphi(S^{\tau/2-1}x) - \varphi(S^{-\tau/2}x)\big] \to 0$ as $\tau \to \infty$.

The value of p^* must be $p^* \geq 1$ otherwise the average of p could not be 1 (as it is by its very definition): it is defined, adopting as in [10, 11] and in the proposition above the natural convention that $\zeta(p) = -\infty$ for the values of p whose probability goes to 0 with τ faster than exponentially, as the infimum of the $p > 0$ for which $\zeta(p) = -\infty$. Alternatively $\pm p^*$ are the asymptotic slopes as $\lambda \to \pm\infty$ of the Laplace transform $\log\langle e^{\lambda p}\rangle_{SRB}$ [12].

The fluctuation relation was discovered in a numerical experiment, [13], dealing with a non smooth system (hence not Anosov). The formulation and proof of the above proposition is in [10] and in the context of Anosov systems the relation (5) is properly called the *fluctuation theorem* (a name later given to other very different relations with remarkable confusion, [14]). The theorem can be extended to Anosov flows (*i.e.* to systems evolving in continuous time), [15].

Alternative formulations

Sometimes, *e.g.* in [16, 17], rather than the above p the quantity $a = \tau^{-1}\sum_{j=-\tau/2}^{\tau/2-1}\sigma(S^j x)$ is considered and eq.(5) becomes

$$\widetilde{\zeta}(-a) = \widetilde{\zeta}(a) - a, \qquad \text{for } |a| < a^* \equiv p^*\sigma_+ \tag{6}$$

where $\widetilde{\zeta}(a)$ is trivially related to $\zeta(p)$. This form dangerously suggests that in systems with $\sigma_+ = 0$ the distribution of a is asymmetric (because the extra condition $|a| < p^*\sigma_+$ might be forgotten, see [18]).

Note that p^* is certainly $< +\infty$ because the variable $\sigma(x)$ is bounded (being continuous on the bounded manifold on which the Anosov map is defined).

However no confusion should be made between $p^*\sigma_+$ and $\sigma_{max} \stackrel{def}{=} \max|\sigma(x)|$: unlike σ_{max} the quantity p^* is a *non trivial* dynamical quantity, independent on the metric used on phase space to measure distances, hence volume. This point has not been always understood and confusion has appeared in the published literature with nefast consequences. In fact it is very easy to build examples of Anosov systems in which $p^*\sigma_+ < \sigma_{max}$: still, this does not mean that fluctuation relation is violated for such systems. Some explicit examples are discussed in next Section.

Conservative systems and the equilibrium limit

Considering more closely the cases $\sigma_+ = 0$ it follows that $\sigma(x) = \varphi(Sx) - \varphi(x)$ (again by the above mentioned result of Griffiths and Ruelle), with φ a smooth function of phase space. Hence the variable

$$a = \frac{1}{\tau}\sum_{j=-\tau/2}^{\tau/2-1}\sigma(S^j x) \equiv \frac{\varphi(S^{-\frac{\tau}{2}}x) - \varphi(S^{\frac{\tau}{2}}x)}{\tau} \tag{7}$$

is *bounded* and tends to 0 *uniformly*. One could repeat the theory developed for p when $\sigma_+ > 0$ but one would reach the conclusion that $\widetilde{\zeta}(a) = -\infty$ for $|a| > 0$ and we see that the result is trivial. In fact in this case it follows that the system admits an absolutely continuous SRB distribution. The distribution of a is symmetric (trivially by time reversal symmetry) and becomes a delta function around 0 as $\tau \to \infty$.

Nevertheless the fluctuation relation is *non trivial* in cases in which the map S depends on parameters $\underline{E} = (E_1, \ldots, E_n)$ and becomes volume preserving ("conservative") as $\underline{E} \to \underline{0}$: in this case $\sigma_+ \to 0$ as $\underline{E} \to \underline{0}$ and one has to rewrite the fluctuation relation in an appropriate way to take a meaningful limit.

The result is that the limit as $\underline{E} \to \underline{0}$ of the fluctuation relation in which both sides are divided by $\underline{E}^2$ makes sense and yields (in the case considered here of transitive Anosov dynamical systems) relations which are non trivial and that can be interpreted as giving Green–Kubo formulae and Onsager reciprocity for transport coefficients, [19, 20].

In fact the very definition of the duality between currents and fluxes so familiar in nonequilibrium thermodynamics since Onsager can be set up in such systems using as generating function the σ_+ regarded as a function of $\underline{E}$. Note that the fluxes are usually "currents" divided by the temperature: therefore via the above interpretation one can try to define the temperature even in nonequilibrium situations, [7, 21, 22].

II. SINGULAR SYSTEMS

The fluctuation relation has been proved only for Anosov systems. However, a *Chaotic Hypothesis* has been proposed, which states that, *for the purpose of studying the physically interesting observables*, a chaotic dynamical system can be considered as an Anosov system, [3, 10, 11].

In applying the chaotic hypothesis to singular systems, *e.g.* a system of particles interacting via a Lennard-Jones potential (which is infinite in the origin), one might encounter apparent difficulties. We will discuss them in the following.

A simple example: Anosov flows

The simplest example (out of many) is provided by the simplest conservative system which is strictly an Anosov transitive system and which has therefore an SRB distribution: this is the geodesic flow S_t on a surface of constant negative curvature, [23]. We discuss here an evolution in continuous time because the matter is considered in the literature for such systems, [18] (even simpler examples are possible for time evolution maps).

The phase space M is compact, time reversal is just momentum reversal and the natural metric, induced by the Lobatchevsky metric $g_{ij}(q)$ on the surface, is time reversal invariant: the SRB distribution is the Liouville distribution and $\sigma(x) \equiv 0$. However one can introduce a function $\Phi(x)$ on M which is very large in a small vicinity of a point x_0, arbitrarily selected, constant outside a slightly larger vicinity of x_0 and positive everywhere. A new metric could be defined as $g_{new}(x) = (\Phi(x) + \Phi(Ix))g(x) \stackrel{def}{=} e^{-F(x)}g(x)$: it is still time reversal invariant but its volume elements *will no longer be invariant* under the time evolution S_t associated with the geodesic flow with respect to the Lobatchevsky metric. The rate of change of phase space volume in the new metric will be $\sigma_{new}(x) = \frac{d}{dt}F(x)$. Then the phase space contraction $\sigma_{new}(x)$ takes values that not only are not identically 0 but which can in general be arbitrarily large, depending on the specific choice of $\Phi(x)$. The distribution of $a = \frac{1}{\tau}\int_0^\tau \sigma_{new}(S_t x)dt = \tau^{-1}\big[F(S_\tau x) - F(x)\big]$, at any finite time, will violate (6), simply because it is symmetric around 0, by time reversal.

In the limit $\tau \to \infty$, as long as $F(x)$ is bounded, $a = \tau^{-1}\big[F(S_\tau x) - F(x)\big] \xrightarrow[\tau\to\infty]{} 0$ uniformly in x, as in the corresponding map case, and the SRB distribution of a will tend to a delta function centered in 0 (hence $\widetilde\zeta(a) = -\infty$ for $a \neq 0$). However, if $F(x)$ is not bounded (*e.g.* if it is allowed to become infinite in x_0) the distribution of a could be different from a delta function centered in 0 also for conservative systems, yielding a finite distribution $\widetilde\zeta(a)$. One might be tempted to apply the fluctuation relation to it, but misleading results could be obtained, as will be discussed in the following.

The effect of singular boundary terms

One can realize that terms of the form $\tau^{-1}\big[F(S_\tau x) - F(x)\big]$ with $F(x)$ not bounded can affect the large fluctuations of $\sigma(x)$, at least if the probability of an arbitrarily large value of F is not too small, *i.e.* if asymptotically for big values of F it is exponentially small in F (or larger), *e.g.* it is of the form $\sim e^{-\kappa F}$, for some constant $\kappa > 0$. This is a valuable and interesting remark brought up for the first time, and correctly interpreted, already in [24] and in the following papers [25, 26]. The analysis of [24–26] applies to cases where the unbounded fluctuations are driven by an external white noise. In the following we extend the theoretical analysis in [25, 26] to cases in which the unbounded fluctuations do not arise from a Gaussian noise but from a deterministic evolution like the ones in [16, 17]: this is a simple extension of the main idea and method of [25] and provides an alternative interpretation to the analysis in [16, 17].

Two important examples (also considered in [16, 17]) to which our analysis can be applied are systems of particles interacting via an unbounded potential (like a Lennard–Jones or a Week–Chandler potential), driven by an external field and subject to an isokinetic or a Nosé–Hoover thermostat. To be definite one can consider system of N particles in d dimensions, described by evolution equations $\dot{\mathbf{p}}_i = \mathbf{E} - \partial_{\mathbf{q}_i}\Phi - \alpha\mathbf{p}_i$, $\dot{\mathbf{q}}_i = \mathbf{p}_i$. For an isokinetic thermostat, α is a function of $\mathbf{p}_i$, chosen so to keep the total kinetic energy fixed to $\sum_i \mathbf{p}_i^2 = Nd\beta^{-1}$. For a Nosé–Hoover thermostat $\alpha(t)$ is a variable independent of $\mathbf{q}_i(t), \mathbf{p}_i(t)$ and satisfying the evolution equation $\dot\alpha = \frac{1}{Q}\big[\sum_i \mathbf{p}_i^2 - Nd\beta^{-1}\big]$, with $Q, \beta > 0$ parameters.

In both cases the phase space contraction $\sigma(x)$ has the form $\sigma_0(x) - \beta\frac{d}{dt}V(x)$, where β has the interpretation of inverse temperature. In the isokinetic case, $\sigma_0(x)$ is bounded, and $V = \Phi$. In the Nosé–Hoover case $\sigma_0(x)$ has, in the SRB distribution, a fast decaying tail (Gaussian at equilibrium, and likely to remain such in presence of external forcing) and $V = \sum_i \frac{\mathbf{p}_i^2}{2} + \Phi(\mathbf{q}) + Q\frac{\alpha^2}{2}$, [27, 28].

In both cases, *in equilibrium*, the SRB probability of V has an exponential tail $\sim e^{-\beta V}$ (possibly with power-law corrections). *For the purpose of illustration* we assume, from now on, that the same happens in presence of the force $\mathbf{E}$. This is an *essential and far from obvious assumption* useful, as discussed below, to understand the possible role of the singularities, but it should not be assumed lightly as it is well known that the SRB distributions may have very peculiar E dependence and, at the moment, a not intuitive character, [29, 30]. Nevertheless, in preliminary numerical simulations, it seems approximately correct, at least within the accuracy of the numerical data and for $|\mathbf{E}|$ not too large; furthermore the analysis that follows can be naturally adapted to more general assumptions on the tails.

In such cases the non normalized variable a (introduced before Eq. (6)) has the form $a_0 + \frac{\beta}{\tau}(V_i - V_f)$ where V_i, V_f are the values of $V(x)$ at the initial and final instants of the time interval of size τ on which a is defined, and $a_0 \stackrel{def}{=} \frac{1}{\tau}\int_0^\tau \sigma_0(S_t x)dt$:

$$a = \frac{1}{\tau}\int_0^\tau \sigma(S_t x)dt \equiv a_0 + \frac{\beta}{\tau}(V_i - V_f) \tag{8}$$

If the system is chaotic and τ is large, the variables a_0, V_i, V_f can be regarded as independently distributed and the distribution of $V = V_i$ or $V = V_f$ is essentially $\sim e^{-\beta V}dV$ to leading order as $V \to \infty$, as discussed above. Therefore the rate function of the variable a can be computed as

$$\begin{aligned}&\lim_{\tau\to\infty}\frac{1}{\tau}\log\int_{-p^*\sigma_+}^{p^*\sigma_+} da_0 \int_0^\infty dV_i \int_0^\infty dV_f\,\cdot\\ &\cdot\, e^{\tau\widetilde\zeta_0(a_0) - \beta V_i - \beta V_f}\,\delta[\tau(a - a_0) + \beta V_i - \beta V_f] \\ &= \lim_{\tau\to\infty}\frac{1}{\tau}\log\int_{-p^*\sigma_+}^{p^*\sigma_+} da_0\, e^{\tau\widetilde\zeta_0(a_0) - \tau|a - a_0|}\end{aligned} \tag{9}$$

where $\widetilde\zeta_0(a_0)$ is the rate function of a_0; thus

$$\widetilde\zeta(a) = \max_{a_0\in[-p^*\sigma_+, p^*\sigma_+]}\Big[\widetilde\zeta_0(a_0) - |a - a_0|\Big] \tag{10}$$

Defining $a_\mp$ by $\widetilde{\zeta}_0'(a_\mp) = \pm 1$, by the strict convexity of $\widetilde{\zeta}_0(a_0)$ it follows

$$\widetilde{\zeta}(a) = \begin{cases} \widetilde{\zeta}_0(a_-) - a_- + a \ , & a < a_- \\ \widetilde{\zeta}_0(a) \ , & a \in [a_-, a_+] \\ \widetilde{\zeta}_0(a_+) + a_+ - a \ , & a > a_+ \end{cases} \tag{11}$$

If we assume that $\widetilde{\zeta}_0(a_0)$ satisfies FR (as expected from the chaotic hypothesis, see below), then $\widetilde{\zeta}_0(a_0) = \widetilde{\zeta}_0(-a_0) + a_0$ and by differentiation it follows that $a_- = -\sigma_+$, where σ_+ is the location of the maximum of $\widetilde{\zeta}_0$, *i.e.* is the average of a.

It follows that, if $\widetilde{\zeta}_0(a_0)$ satisfies FR up to $a = p^*\sigma_+$, then $\widetilde{\zeta}(a)$ satisfies FR only in the interval $|a| < |a_-| = \sigma_+$. Outside this interval $\widetilde{\zeta}(a)$ does not satisfy the FR and in particular for $a \geq a_+$ it is $\widetilde{\zeta}(a) - \widetilde{\zeta}(-a) = const.$, as already described in [25]. Translated into the normalized variables $p_0 = a_0/\sigma_+$ and $p = a/\sigma_+$, this means that, even if the rate function of p_0 satisfies FR up to $p^* > 1$, the rate function of p verifies FR only for $|p| \leq 1$. This is the effect due to the presence of the singular boundary term.

An example of $\widetilde{\zeta}(a)$ is reported in Fig. 1: it is a simple stochastic model for the FT (taken from Sect. 5 in [31], see also the extensions in [32, 33]). The example is the Ising model without interaction in a field h, *i.e.* a Bernoulli scheme with symbols $\pm$ with probabilities $p_\pm = \frac{e^{\pm h}}{2\cosh h}$. Defining $a_0 = \frac{1}{\tau}\sum_{i=0}^{\tau-1} 2h\sigma_i$, so that $\sigma_+ = \langle a_0 \rangle = 2h\tanh h$, and setting $x \stackrel{def}{=} \frac{1+a_0/(2h)}{2}$, and $s(x) = -x\log x - (1-x)\log(1-x)$, one computes $\widetilde{\zeta}_0(a_0) = s(x) + \frac{1}{2}a_0 + const$ which is *not Gaussian* and it is defined in the interval $[-a^*, a^*]$ with $a^* = 2h$. In this case the large deviation function $\widetilde{\zeta}_0(a_0)$ satisfies FR for $|a_0| \leq a^*$. If a singular term $V = -\log(\sum_{i=0}^{\infty} 2^{-i-1}\frac{\sigma_i+1}{2})$ is added to a_0, defining $a = a_0 + \beta(V_i - V_f)$ (with $\beta = \log_2(1+e^{2h})$ so that the probability distribution of V is $\sim e^{-\beta V}$ for large V), the resulting $\widetilde{\zeta}(a)$ does not verify FR for $a > \langle a \rangle = 2h\tanh h$. In particular, for $h \to 0$, the interval in which the FR is satisfied vanishes.

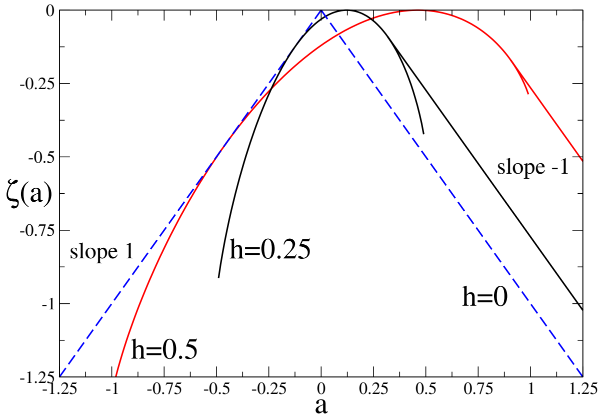

FIG. 1: An example in a stochastic model of FR. The graph gives the two functions $\widetilde{\zeta}_0(a)$ and $\widetilde{\zeta}(a)$ for $h = 0., 0.25, 0.5$. The average of a is $\langle a \rangle = \sigma_+ = 2h\tanh h$, $a_+ = 2h\tanh 3h$ and $a^* = 2h$. The function $\widetilde{\zeta}(a)$ is obtained from $\widetilde{\zeta}_0(a)$ by continuing it for $a < a_- = -\sigma_+$ and $a > a_+$ with straight lines of slope ± 1. It does not satisfy the FR for $|a| > \langle a \rangle$. As $h \to 0$, $\langle a \rangle \to 0$, which means that the interval in which the FR is verified shrinks to 0. In this limit $a_+ \to 0$, so $\widetilde{\zeta}(a)$ approaches $-|a|$ (dashed lines). Refrasing this in terms of $p = \frac{a}{\langle a \rangle}$ one obtains that FR remains always valid for $|p| < 1$, even as $h \to 0$. The three curves for $\widetilde{\zeta}_0(a)$ have the same tangent on left side. The function $\widetilde{\zeta}_0(a)$ is finite *only* in the interval $[-2h, 2h]$ and it is $-\infty$ outside it, while the function $\widetilde{\zeta}(a)$ is finite for all a'a and is a straight line outside $[a_-, a_+]$.

How to remove singularities

From the discussion above it turns out that singular terms which are proportional to total derivatives of unbounded functions (like the term $\frac{dV}{dt}$ that appears in the phase space contraction rate of thermostatted systems) can induce "undesired" (or "unphysical") modifications of the large deviations function $\zeta(p)$.

On heuristic grounds, when dealing with singular systems, one could follow the prescription that unbounded terms in $\sigma(x)$ which are proportional to total derivatives should be *subtracted* from the phase space contraction rate. If the resulting $\sigma_0(x)$ is bounded (as it is *e.g.* for the isokinetic thermostat models considered) or at least if the tails of its distribution decay faster than exponentially, then its large deviations function should verify the FR for $|p| \leq p^*$, p^* being the intrinsic dynamic quantity defined above.

Note that after the subtraction of the divergent terms the remaining contraction, in the considered cases, is bounded for isokinetic thermostats or has a Gaussian tail (*i.e.* faster than exponential) in the case of Nosé–Hoover thermostats.

If the singular terms are not subtracted, the FR *will appear to be valid only for* $|p| \leq 1$ *even if* $p^* > 1$. This seems to have generated statements that the Chaotic Hypothesis does not apply to isokinetic systems, see [17].

The heuristic prescription above can be motivated by a careful analysis of the proof of the fluctuation theorem for Anosov flows. In the following let us call again a the integral of the total phase space contraction rate $\sigma(x)$ (which includes singular terms) and a_0 the integral of the bounded variable $\sigma_0(x)$ from which singular total derivatives have been removed.

The fluctuation theorem was proved in [1, 10, 34] *for Anosov maps* and only later it has been extended in [15] to Anosov flows. Very sketchily, the extension of the fluctuation theorem to Anosov flows in [15] is proved as follows. One reduces the Anosov flow on Ω to a map via a Poincaré's section, associated with surfaces on Ω

transversal to the flow. The passage of the flow through any one of such surfaces is called a *timing event*. The map between two consecutive timing events is called a "Poincaré's map". The union Ω_P of the surfaces represents the phase space of the Poincaré's map. The surfaces in Ω_P can be suitably chosen, in such a way that the Poincaré's map is a chaotic map which although not smooth, hence not an Anosov map, has (a non trivial fact [15]), all the properties necessary to prove the fluctuation theorem (which therefore applies to systems more general than the Anosov maps, although there is not a general characterization of the systems which are not Anosov and to which it applies). So, for such a map the fluctuation theorem holds and this in turn leads to a FR for the flow by the theory in [15] *under the assumption that the variable $\sigma(x)$ is bounded*.

If, as in the case under analysis, $\sigma(x)$ is not bounded, we can interpret the chaotic hypothesis as applying to the map associated with a Poincaré's section which avoids the singularities of the potential, a very natural prescription which allows us to apply the theory in [15] and derive a FR for both the map and the flow. For instance, we can choose as timing events the instants in which either the potential energy or the Nosé's "extended Hamiltonian" exceed some fixed value $\bar{V}$. If we make this choice, the (discrete) average $\hat{a}$ of $\sigma(x)$ over a sequence of iterations of the Poincaré's map will coincide with the (discrete) average $\hat{a}_0$ of $\sigma_0(x)$ along the same sequence: this simply follows from the remark that by construction the total increment of $\sigma(x)-\sigma_0(x)$ between two timing events, given by $\beta(V_f-V_i)$, is 0 (by construction Ω_P is chosen as a subset of $\{x \in \Omega : V(x) = \bar{V}\}$ where $V_f = V_i$). Then, by the same argument in [15], the fact that the rate function of $\hat{a}_0$ satisfies a FR and that $\sigma_0(x)$ is bounded implies that the rate function of the continuous average a_0 of $\sigma_0(x)$ along a trajectory of the flow will satisfy the fluctuation theorem.

Therefore the distribution of a_0 will satisfy the FR (*by the chaotic hypothesis*) for $|a_0| < p^*\sigma_+$. By the above maximum argument, the distribution of *a will also verify, as a consequence*, the FR but only for $|a| \leq \sigma_+$ (*i.e.* in the form (5) or (6) with $|p| \leq 1$).

Then the (natural) prescription to study FR for chaotic flows is to reduce the problem to a chaotic map considering only Poincaré's sections which do not pass through a singularity of $\sigma(x)$. The integral of $\sigma(x)$ over a large number of timing events on such sections is equal to the time integral of $\sigma_0(x)$ plus a bounded term which can be neglected. Thus the prescription on the choice of Poincaré's sections is equivalent to the heuristic prescription of removing from $\sigma(x)$ all the unbounded total derivatives.

It follows that the chaotic hypothesis leads to a clear prediction on the outcome of possible numerical simulations of particle systems interacting via unbounded potentials and subject to the isokinetic or the Nosé–Hoover thermostat: the FR will hold for all $|a| \leq \sigma_+$ and, once the term $\frac{dV}{dt}$ is removed, for all $|a_0| < p^*\sigma_+$ with $p^* \geq 1$. Note that in the cases under analysis a_0 coincides with the dissipation function of Evans and Searles that was in fact predicted to satisfy FR [16, 17], even though for different reasons. We believe that the correct interpretation of the fact that FR for $\widetilde{\zeta}_0(a_0)$ holds for all $|a_0| < p^*\sigma_+$ is the one given above.

The numerical results of [35, 36] agree with the prediction that FR for the rate function of a_0 is valid even beyond $a_0 = \sigma_+$. The prediction that (at least near equilibrium) the rate function of a should satisfy FR only up to $a = \sigma_+$ and that should become linear for $a \geq a_+$ at the moment has been experimentally confirmed only in Gaussian cases [24–26]. It would be very interesting to investigate in detail the structure of $\widetilde{\zeta}(a)$ even in non Gaussian cases. Note that this is far from being an easy task (in particular the analysis in [36] was not sophisticated enough to study this problem). In fact, as discussed in detail in [35], the presence in the definition of σ of a total derivative of an unbounded function may enlarge of 2 orders of magnitudes the times needed for the probability distribution of a to reach its asymptotic shape: even in the Gaussian region (small fluctuations of a around σ_+) the convergence times for $\widetilde{\zeta}(a)$ are found to be of order 1000 decorrelation times, versus a time of order 10 decorrelation times needed for $\widetilde{\zeta}_0(a_0)$ to converge to its asymptotic shape [35]. Clearly, for times of order 1000 decorrelation times, it is very hard to observe fluctuations of a larger than $a_+ - \sigma_+$. In order to verify the prediction for the shape of $\widetilde{\zeta}(a)$ beyond $a = a_+$ an *ad hoc* experiment would be needed, together with a detailed investigation of the finite time corrections to $\widetilde{\zeta}(a)$, along the lines in [36].

Meaning of the chaotic hypothesis

It has to be stressed that the chaotic hypothesis concerns physical systems: *it is very easy to find dynamical systems for which it does not hold*. As it is easy (actually even easier) to find systems in which the ergodic hypothesis does not hold (*e.g.* harmonic lattices or black body radiation). However, if suitably interpreted, the ergodic hypothesis leads even for these systems to physically correct results (the specific heats at high temperature). Moreover the failures of the ergodic hypothesis have led to new scientific paradigms (like quantum mechanics from the specific heats at low temperature and Planck's law).

Since physical systems are almost always not Anosov systems it is very likely that probing motions in extreme regimes (*e.g.* when particles go undisturbed through infinite potential walls, as in the Week-Chandler's isokinetic models) will make visible the features that distinguish Anosov systems from non Anosov systems: for instance the fact that isokinetic or Nosé–Hoover thermostatted particle systems satisfy the FR only for $|p| \leq 1$ if the singular term $\frac{dV}{dt}$ is not removed from $\sigma(x)$. Note that these results *can be derived* from the chaotic hypothesis as discussed above: this is a quite remarkable property and a confirmation of CH rather than an argument against it.

The results in [35, 36] although not designed to probe the peculiarities of the Week–Chandler's potential show that the FR for the rate function of a_0 holds even beyond $p = 1$ and this provides further numerical evidence for the chaotic hypothesis even in situations in which the non-Gaussian nature of the large deviations function $\zeta(p)$ is manifest. As discussed above, an even stronger confirmation of CH would follow from a possible numerical observation of the scenario discussed above for the rate function of a in systems in which a_0 admits a non Gaussian distribution function; it would be very interesting to investigate explicitly this phenomenon in thermostatted driven particle systems. The situation looks much more promising in the case of stochastic systems: the simple case discussed in Fig. 1 already gives evidence in this direction.

III. REMARKS

We conclude by comparing more closely our discussion and our interpretation of apparent violations to the FR coming from singularities in the interaction potential with the corresponding discussion in [17], where a different interpretation is given (in particular it is argued that FR and CH do not hold for thermostatted systems near equilibrium).

(1) As stressed above it is dangerous (and wrong) to consider (6) without the restriction $|a| \le p^*\sigma_+$ as the prediction of fluctuation theorem. However in [17] this is exactly what the authors do, by saying, in a somewhat unusual form, that "*it is convenient to consider* [the relation Eq.(6) without the condition $|a| < p^*\sigma_+$] *as the prediction of the FT*". The attribution is not correct but it is convenient to produce arguments, in most of the rest of the paper, that show that (6) without the restriction $|a| \le p^*\sigma_+$ is incorrect (an *a priori* obvious fact).
(2) An argument in [17] is supposed to prove that the relation $\widetilde{\zeta}(-a) = \widetilde{\zeta}(a) - a$ (without the condition $|a| < p^*\sigma_+$) holds for reversible Anosov systems for all a's, also for $\sigma_+ = 0$ (in particular they say that "*the division by σ_+ does not seem to be necessary for the proof in [37]*"). Since at equilibrium as well as near equilibrium, as remarked above and as illustrated also by [23], *there are examples of Anosov systems which, although reversible, do not satisfy $\widetilde{\zeta}(-a) = \widetilde{\zeta}(a) - a$* in the sense of [17] (*i.e.* with $\widetilde{\zeta}(a)$ finite) for $a > p^*\sigma_+$, it follows that the assumption $\sigma_+ > 0$ *is, instead, essential* for the proof of fluctuation theorem. The necessity of the assumption $\sigma_+ > 0$ is stressed in the early paper [12] which the Authors of [17] quote; it is stressed also in the paper [1] which also makes clear that $\widetilde{\zeta}(-a) = \widetilde{\zeta}(a) - a$ can only hold under the assumption that $|a|$ does not exceed a maximum value.
(3) In the same reference [17] the point is made that the systems that they consider have an infinite upper bound for $\sigma(x)$ and this should explain why their numerical check of the FR fails if one looks for it in terms of the variable a (rather than in terms of p, which in their equilibrium cases would not even be defined). They attribute the failure to the fact that the system in equilibrium (*i.e.* with $\sigma_+ = 0$) is "not Anosov". While this is mathematically obvious in their case (as the system they consider is not even smooth) the apparent failure of the FR must be interpreted differently. As remarked above, at equilibrium a correct repetition of the proof of fluctuation theorem yields $\widetilde{\zeta}(a) = \widetilde{\zeta}(-a)$ instead of $\widetilde{\zeta}(a) = \widetilde{\zeta}(-a) + a$: *irrespective of whether the chaotic hypothesis holds and irrespective of whether the system is Anosov or not.* For $\sigma_+ > 0$ and small, by the arguments above, one expects to see the FR for $\widetilde{\zeta}(a)$ only for values of a with $|a| \le \sigma_+$, while, for the variable a_0 obtained by subtracting from a the singular contributions due to the the singular term $\frac{dV}{dt}$, one expects the FR to hold up to $a_0 = p^*\sigma_+$. In both cases the FR is expected to hold up to a quantity proportional to σ_+. Moreover the asymptotic shape of the probability distribution for a or a_0 will be visible at best on a time scale proportional to σ_+^{-1}. As remarked above the time scales needed to see the distributions of a or of a_0 reaching their asymptotic shapes are different: the one for a might be about 2 orders of magnitudes larger than the one for a_0, see [35].

Hence in the near to equilibrium case the FR holds for a in a tiny interval around $a = 0$ and becomes visible only on a very long time scale $\sim \sigma_+^{-1}$. The quotations of numerical simulations in [17] fail to take into account these aspects and this could explain why the FR "is not seen" in numerical experiments which look at moderate time scales and, furthermore, at values of a which deviate by several multiples of the width of the distribution of a which (by the FR) is precisely the (small) value of σ_+. For instance in [37] the fluctuations are studied over an interval of variation of a which is an order of magnitude larger than the average phase space contraction. The experiment in [38] is a very convincing evidence of the relevance of the general theory in [25] and of our analysis in Sect. 2 (which is an extension of [25] to non Gaussian cases).
(4) The analysis in Sect. 2 above shows that the probability distribution describing isokinetic systems near equilibrium *are SRB distributions* (contrary to what is claimed in [17]): this is mathematically obvious by the very definition of SRB distribution in the case of Anosov systems (*even if isokinetic*, see *e.g.* the geodesic flow discussed above) and it appears to be true also in non Anosov systems that have so far been considered.
(5) If the term $\frac{dV}{dt}$ is removed (as in Ref. [35, 36]), the resulting quantity $\sigma_0(x)$ is bounded and its distribution verifies the FR also for $|p| > 1$. This prescription, as discussed above, is equivalent to the very reasonable prescription that the Poincaré's section used for mapping the flow into a map does not pass through a singularity of $\sigma(x)$.
(6) In the case of the thermostatted particle systems con-

sidered in [17] the unbounded derivative $\frac{dV}{dt}$ is also the contraction rate of the volume in equilibrium, *i.e.* with $\mathbf{E} = 0$. Thus, for $\mathbf{E} \neq \mathbf{0}$, one can remove the total derivative from $\sigma(x)$ simply considering the contraction *with respect to the equilibrium invariant distribution* $e^{-\beta V}$, as stated in [16, 17]. However, this observation does not provide a general prescription to remove the singular part from the phase space contraction rate because it rests on the very special fact that the singularities of the function $V(x)$ (*i.e.* of the potential Φ) do not depend on $\mathbf{E}$. The prescription that the phase space contraction should be computed on non singular Poincaré's sections, instead, does not require any other assumption.
(7) The analysis in Sect. 2 above applies as well to understand how to apply the FR to systems with Gaussian (or unbounded) noise and the compatibility between the general theory of [32, 33, 39] with the works [25] and [26, 38].
(8) In other words our analysis suggests that the Chaotic Hypothesis is an appropriate characterization of thermostatted systems, even for isokinetic or Nosé–Hoover thermostatted systems, see concluding remark of [17].

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