Fluctuation relation for the temperature derivative of Lyapunov exponents

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We derive a fluctuation expression for the temperature derivative of the largest Lyapunov exponent for a large system of classical particles in contact with a heat bath. The resulting expression is strikingly similar to the statistical-mechanical fluctuation expression for the temperature derivative of thermodynamic variables in a canonical ensemble.

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In this paper we exploit a recently established analogy [1] between nonequilibrium statistical mechanics in *phase space* and the dynamics of Lyapunov instability in *tangent space* to derive a fluctuation expression for the temperature derivative of the largest Lyapunov exponent for a classical N-body system. This fluctuation expression has a close resemblance to the expression for the temperature derivative of thermodynamic functions as evaluated in the canonical ensemble. We also provide computer-simulation data which support the validity of our fluctuation relation.

We write the equations of motion for an autonomous, N-body, classical system as

$$\dot{\mathbf{\Gamma}}=\mathbf{G}(\mathbf{\Gamma}) , \tag{1}$$

where $\mathbf{\Gamma}$ is the phase-space vector consisting of the position coordinates and the momenta of all N particles in the system. We can define a separation vector $\boldsymbol{\delta}_1=\mathbf{\Gamma}_1-\mathbf{\Gamma}_0$ between two phase space vectors $\mathbf{\Gamma}_0$ and $\mathbf{\Gamma}_1$. If the length of the separation vector goes to zero it becomes a tangent vector whose equation of motion is [1]

$$\dot{\boldsymbol{\delta}}_1=\left.\frac{\partial\mathbf{G}}{\partial\mathbf{\Gamma}}\right|_{\Gamma+\Gamma_0}\cdot\boldsymbol{\delta}_1\equiv\mathbf{T}\cdot\boldsymbol{\delta}_1 , \tag{2}$$

where $\mathbf{T}=\mathbf{T}(\mathbf{\Gamma})$ is the stability or Jacobian matrix of the equations of motion. The largest, i.e., most positive, Lyapunov exponent, $\lambda_{\max}$, is obtained as the limit [2]

$$\lambda_{\max}=\lambda_1\equiv\lim_{t\to\infty}\frac{1}{2t}\ln\frac{\delta_1^2(t)}{\delta_1^2(0)} , \tag{3}$$

where $\delta_1(t)=|\boldsymbol{\delta}_1(t)|$.

For our present purposes it is convenient to compute this Lyapunov exponent using a continuous rescaling method [3,4]. Consider a constrained tangent vector $\boldsymbol{\delta}_1^c$ whose length is held constant,

$$\dot{\boldsymbol{\delta}}_1^c=\mathbf{T}\cdot\boldsymbol{\delta}_1^c-\zeta_{11}\boldsymbol{\delta}_1^c ,$$

where

$$\zeta_{11}=\zeta_{11}(\mathbf{\Gamma},\boldsymbol{\delta}_1^c)=\frac{\boldsymbol{\delta}_1^c\cdot\mathbf{T}\cdot\boldsymbol{\delta}_1^c}{\delta_1^{c2}} . \tag{4}$$

It is easy to show that there is an exact relation [1] between the largest Lyapunov exponent and the long-time average of the multipler ζ_{11} used to contain the tangent vector length,

$$\lambda_1=\lim_{t\to\infty}\frac{1}{t}\int_0^t ds\,\zeta_{11}(s) . \tag{5}$$

Suppose we consider the ensemble averaged response of $\zeta_{11}(\mathbf{\Gamma}^*)$, $\{\mathbf{\Gamma}^*\}\equiv\{\mathbf{\Gamma},\boldsymbol{\delta}_1^c\}$, to an initial tangent space distribution $f(\mathbf{\Gamma}^*,0)$,

$$f(\mathbf{\Gamma}^*,0)=\frac{e^{-\beta H_0(\mathbf{\Gamma})}\delta(\delta_1^{c2}-d^2)}{\int d\mathbf{\Gamma}^* e^{-\beta H_0(\mathbf{\Gamma})}\delta(\delta_1^{c2}-d^2)} . \tag{6}$$

This means that the initial phase of the N-body system, $\mathbf{\Gamma}=\mathbf{\Gamma}(0)$, is distributed canonically and the ensemble temperature is $T=1/k_B\beta$ where k_B is Boltzmann's constant. The initial tangent vector $\boldsymbol{\delta}_1=\boldsymbol{\delta}_1(0)$ is uniformly distributed on a $6N$-dimensional hypersphere of radius d.

Assuming the equivalence of time and ensemble averages, the standard methods of response theory [5] can be used to calculated the largest Lyapunov exponent.

$$\lambda_1=\lim_{t\to\infty}\langle\zeta_{11}(t)\rangle=\lim_{t\to\infty}\int d\mathbf{\Gamma}^*\zeta_{11}(\mathbf{\Gamma}^*)f(\mathbf{\Gamma}^*,t) . \tag{7}$$

If the system consists of N particles in three dimensions, and *if* the dynamics conserves energy H_0 and the tangent vector length $\delta_1(t)$, one can show that

$$f(\mathbf{\Gamma}^*,t)=\frac{\delta(\delta_1^{c2}-d^2)\exp\left[-\beta H_0(\mathbf{\Gamma})+\int_0^t ds\,6N\zeta_{11}(-s)\right]}{\int d\mathbf{\Gamma}^*\delta(\delta_1^{c2}-d^2)\exp\left[-\beta H_0(\mathbf{\Gamma})+\int_0^t ds\,6N\zeta_{11}(-s)\right]} . \tag{8}$$

The factor of $6N$ appearing in the exponent arises from the fact that $[\partial/\partial\mathbf{\Gamma}^*]\cdot d\mathbf{\Gamma}^*/dt=-6N\zeta_{11}$ and $df(\mathbf{\Gamma}^*,t)/dt=-[\partial/\partial\mathbf{\Gamma}^*]\cdot d\mathbf{\Gamma}^*/dt$. Substituting (8) into (7), differentiating the resulting equation for $\langle\zeta_{11}(t)\rangle$ with respect to time, and reintegrating shows that

$$\lambda_1=\lim_{t\to\infty}\langle\zeta_{11}(t)\rangle=\lim_{t\to\infty}6N\int_0^t ds\langle\zeta_{11}(0)\zeta_{11}(s)\rangle\ . \tag{9}$$

This relation, which superficially resembles the famous Green-Kubo relations for transport coefficients, has recently been tested numerically [1] and was found to be in agreement with computer-simulation results.

From Eqs. (7) and (8), one can derive expressions for the temperature derivative of the largest Lyapunov exponent. If we subject a canonical ensemble of systems to Newtonian constant-energy dynamics, the time propagator is clearly independent of the Boltzmann factor β. One can therefore easily compute the derivative with respect to β as

$$\begin{aligned}\frac{\partial}{\partial\beta}\langle\zeta_{11}(t)\rangle&=-\int d\mathbf{\Gamma}^*\zeta_{11}H_0f(t)\\&\quad+\int d\mathbf{\Gamma}^*H_0f(t)\int d\mathbf{\Gamma}^*\zeta_{11}f(t)\\&=-\langle\zeta_{11}(t)H_0(t)\rangle+\langle\zeta_{11}(t)\rangle\langle H_0(t)\rangle\\&=-\langle\Delta\zeta_{11}(t)\Delta H_0(t)\rangle\ ,\end{aligned} \tag{10}$$

where

$$\Delta B(t)\equiv B(t)-\langle B(t)\rangle\ .$$

This exact expression for the temperature derivative of the largest Lyapunov exponent bears a striking resemblance to the corresponding expression for the temperature derivative of thermodynamic quantities in the canonical ensemble [6].

We decided to test (10) numerically by using molecular-dynamics computer simulations. Evaluating the expression as written, (10) would require performing an *ensemble* of constant-energy molecular-dynamics simulations from an initial ensemble of phases and tangent vectors distributed according to (6). This would be time consuming and cumbersome.

It has been known for some time that in an ergodic system one can generate canonical ensemble averages by time averaging along a single phase-space trajectory generated by Nosé-Hoover equations of motion,

$$\begin{aligned}\dot{\mathbf{q}}_i&=\mathbf{p}_i/m\ ,\\\dot{\mathbf{p}}_i&=\mathbf{F}_i-\alpha\mathbf{p}_i\ ,\\\dot{\alpha}&=\left[\frac{\sum_i p_i^2/2m}{3Nk_BT/2}-1\right]\Big/\tau^2\ ,\end{aligned} \tag{11}$$

where α is the thermostatting multiplier and τ is the Nosé-Hoover time constant. However, if we attempt to calculate the temperature derivative $\partial/\partial\beta$ using these equations, we should in principle obtain new terms not represented in (10) because the time propagator itself is directly dependent on β.

This problem is easily avoided. If we replace the last equation in (11) by

$$\dot{\alpha}=\left[\frac{\sum_i p_i^2/2m}{\sum_i[p_i(0)]^2/2m}-1\right]\Big/\tau^2\ , \tag{12}$$

it is trivial to show that an ensemble of such trajectories has precisely the same long-time tangent-space distribution $\lim_{(t\to\infty)}f(\mathbf{\Gamma}^*,t)$ as the distribution of states along a single trajectory generated by (11). Moreover, the propagator in (12) has no explicit temperature dependence. Therefore the temperature derivative of the Lyapunov exponent can be calculated from a time average along a *single* trajectory as

$$\frac{\partial\lambda_1}{\partial\beta}=\lim_{t\to\infty}\left[\frac{1}{t}\int_0^t ds\,\zeta_{11}(s)H_0(s)-\frac{1}{t^2}\int_0^t ds_1\zeta_{11}(s_1)\int_0^t ds_2H_0(s_2)\right]\ . \tag{13}$$

We carried out equilibrium molecular-dynamics simulations of $N=56$ disks of units mass, characterized by the Weeks-Chandler-Anderson (WCA) interaction potential $\phi(r)$,

$$\phi(r)=\begin{cases}4[r^{-12}-r^{-6}] & \text{for } r<2^{1/6}\\0 & \text{for } r>2^{1/6}\end{cases} \tag{14}$$

within periodic boundary conditions. The length of the tangent vector δ_1 was fixed using Eq. (4) at 10^{-6}.

In Fig. 1 we show the variation of the largest Lyapunov exponent λ_1 with respect to β, at a density $\rho=N/V=0.8$ and a temperature $T\sim1.0$. Equation (5) was used to calculate λ_1. As can be seen, over this narrow range of β the data are rather accurately represented as a linear function of β. At $\beta=1.0$ the slope $\partial\lambda_1/\partial\beta|_1=-1.61\pm0.02$. This estimate of the temperature derivative was compared with a calculation using

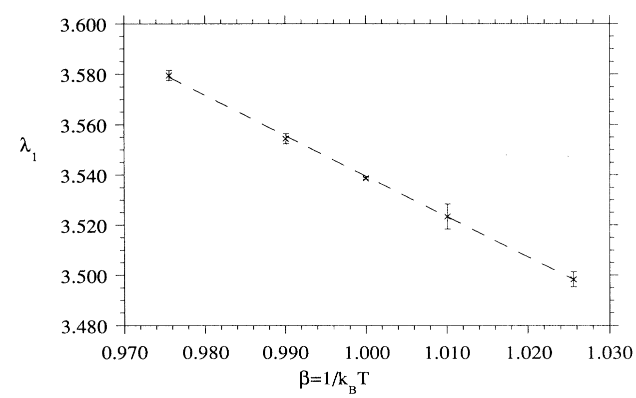

FIG. 1. We show the variation of the largest Lyapunov exponent with respect to the reciprocal of the temperature, β. The system is 56 WCA particles at a reduced density of 0.8. The results are shown in reduced units as explained in the text. As can be seen, the data are consistent with a linear variation with β and $\partial\lambda_1/\partial\beta|_1=-1.6\pm0.02$. This slope agrees with that predicted from our fluctuation formula (13), namely $\partial\lambda_1/\partial\beta|_1=-1.60\pm0.03$.

our fluctuation formula (13) from a simulation lasting 33 000 reduced time units, $\partial\lambda_1/\partial\beta|_1 = -1.60 \pm 0.03$. The results obtained therefore verify (13) within estimated statistical uncertainties.

The work described in this paper shows that Lyapunov exponents can be calculated and characterized by methods that are extensions of those employed in nonequilibrium statistical mechanics. Using the same methods as those used here one can also derive an analogous fluctuation formula for the pressure dependence of the largest Lyapunov exponent in many-body systems. These methods can be extended to the full $6N$ Lyapunov exponents but the presently known formulas are, for the smaller Lyapunov exponents, somewhat cumbersome. Similarly expressions can be derived for the temperature derivative of the largest Lyapunov exponent in thermostatted nonequilibrium systems rather than the equilibrium case treated here.

Finally, we have recently proved a direct relation between the difference of the two maximal Lyapunov exponents of nonequilibrium steady states and the transport coefficients describing the dissipation in the system, preventing its relaxation to equilibrium [7]. This latter relation shows that the variation of Lyapunov exponents with respect to thermodynamic state variables is important macroscopically since it can be related to the corresponding variation of transport coefficients to those same state variables.

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