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Chaos and linear response: Analysis of the short-, intermediate-, and long-time regime

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We study the response of a classical Hamiltonian system to a weak perturbation in the regime where the dynamics is mixing, with the purpose of critically examining both the foundation of the Kubo linear response theory (LRT) and van Kampen's well known objections to LRT [Phys. Norv. **5**, 279 (1971)]. Although the exactness of LRT for short times is not surprising, we prove that for the class of model studied here the LRT must also become accurate in the limit of long times, even for macroscopically large external perturbations. Hence, if the LRT breaks down, the breakdown occurs in the region of intermediate times. We also show that, for a given system, if *any* macroscopic linear response exists, it must coincide with Kubo LRT; thus, if a generic system responds nonlinearly to an external perturbation, this nonlinear response is observable only in an intermediate-time range. Numerical calculations carried out on some model systems with only a few degrees of freedom support these arguments.

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I. INTRODUCTION

The theoretical difficulties with the treatment of nonequilibrium processes are widely reduced by the adoption of a linear response formalism [1]. Under the assumption that the macroscopic variables of interest are only slightly moved from their equilibrium conditions, it is possible to express their mean values as a linear function of the external disturbances, thereby reducing the original problem to the determination of the static susceptibility. Let us consider, for instance, a macroscopic variable of interest, B. In general, this is a function of many microscopic variables; and we are interested, for instance, in its average (statistical) value, $\langle B\rangle$. If we assume that the average $\langle B\rangle_0$ in absence of the external field is zero, then after the time $t=0$, the time at which the constant field of intensity K is switched on, we can write for the average $\langle B\rangle_K$

$$\langle B\rangle_K = K\chi_B(t) + O(K^2), \tag{1.1}$$

where $\chi_B(t)$ is the susceptibility for the macroscopic variable B. In a recent letter, Chernov *et al.* [2] provided the first derivation of Ohm's law based on a deterministic mechanical model. They proved the validity of linear response theory (LRT) in the context of a Lorentz model. They noted that this result stands in contrast to the objections raised by van Kampen [3] against the Kubo formulation of LRT. However, in their letter the authors do not directly address the criticism of van Kampen, but more simply derive the susceptibility for their model, and do not provide a theoretical explanation of why the LRT in practice works so well. This long-standing controversy between Kubo and van Kampen [3] is to date without solution in a general context.

Let us briefly review this controversy. To derive the linear relations discussed above, it is tempting to assume that the microscopic trajectories respond linearly to an external perturbation. By virtue of this hypothesis, it is relatively easy to derive the linear behavior of macroscopic variables from a microscopic picture; this is the essence of the approach followed by Kubo [1]. Actually Kubo used a perturbative treatment of the Liouville equation

$$\frac{\partial}{\partial t}\rho(t) = (\mathcal{L}_0 + K\mathcal{L}_I)\,\rho(t), \tag{1.2}$$

where $\rho(t)$ is the distribution function of the system, $\mathcal{L}_0$ the unperturbed Liouvillian and $\mathcal{L}_I$ the perturbation; however, due to the basic equivalence between the Hamilton and the Liouville description, it seems [3] that the Kubo approach is essentially equivalent to deriving the linear macroscopic properties from the linearization of the classical trajectories.

The approach of Kubo is appealing, since it leads to expressions for the susceptibility in terms of time-integral of correlation functions:

$$\chi_B^{\text{Kubo}}(t) = \int_0^t \langle B_0(\tau)\mathcal{L}_I\rangle_0 d\tau, \tag{1.3}$$

in which

$$B_0(\tau) \equiv e^{\mathcal{L}_0^\dagger \tau} B \tag{1.4}$$

is the unperturbed evolution of B, leading to elegant theoretical predictions, which have been found to date to be always in remarkable agreement with experimental results [4]. Yet, van Kampen [3] pointed out that macroscopic linearity cannot have the same source as microscopic linearity. Due to the highly unstable character of the microscopic trajectories, the macroscopic linearity could derive from the microscopic one only in the limiting case of perturbations of virtually vanishing intensity. A rough estimate of the time t spent to observe the system of interest, the intensity K of the external perturbation field, the Lyapunov exponent of the system λ, and the energy per degree of freedom ϵ, in order to observe linear response of the individual trajectories, is given by

$$K\left[e^{\lambda t} - 1\right] \ll \omega\sqrt{\epsilon}, \tag{1.5}$$

where ω is a characteristic frequency of the system. It is clear that if t is a macroscopic time while $\bar{t} \equiv 1/\lambda$ is a microscopic time scale, then the intensity of the external perturbation K, satisfying Eq. (1.5), must become extremely small. On the basis of Eq. (1.5), we also expect linear response even to macroscopic external perturbations as long as t is in the range of microscopic times, i.e., for $t \lesssim \bar{t}$.

The reply of Kubo and coworkers to the criticism of van Kampen has more recently been that the perturbation of the single, and unstable, trajectory is not equivalent to the perturbation treatment of the classical Liouville equation. They make this explicit statement [1]: "[...] It is true that the characteristics of the Liouville equation are the phase trajectories and so the Liouville equation and the Hamilton equation of motion are equivalent in this sense. However, the limitation of the class of distribution functions on which the Liouville operator operates makes them nonequivalent. *Unfortunately it is not easy to formulate the mathematical condition for this statement.* But we realize that the instabilities of the trajectories are in fact the cause of mixing, favoring rather than unfavoring the stability of distribution functions. [...]"

Herein we show that, in spite of the van Kampen criticism, for the class of mixing hamiltonian systems with a finite volume in phase space, the susceptibility given by Eq. (1.3) is correct not only for microscopic times less than $\bar{t}$, but also for very large times *and* macroscopic intensities of the external field, despite a possible discrepancy between Kubo LRT and the true response in the region of intermediate times. In this region of intermediate times we have that in practice not only Eq. (1.3) for the susceptibility does not seem to work, but also that we can hardly define a susceptibility, given that the system does not respond linearly, not even for external fields small enough to yield linear response at small and large times. We are led to conclude that very likely van Kampen's argument about a difference between macroscopic and microscopic linear response is not correct: in our case, the system responds linearly on a macroscopic scale as long as the microscopic linear response theory (Kubo LRT) is correct. In the spirit of Ref. [5], we then prove that under the only assumption that the system responds linearly on a macroscopic scale at any time t, it follows that the susceptibility for the system is indistinguishable from the one obtained via Kubo's LRT.

The outline of the paper is as follows. In Sec. II we derive an exact expression for the stationary (asymptotically large times) susceptibility using only geometric arguments. In Sec. III we prove that in the large times limit the Kubo expression for the susceptibility coincides with the exact geometric one evaluated in Sec. II and that if a linear response exists at any times, then its predictions must coincide with those of the Kubo LRT. This leads us to predict that in the intermediate time region, if the Kubo LRT turned out to be invalid, the system would indeed respond nonlinearly, and *no* LRT would be applicable. This prediction is supported by the results of computer calculations, which are illustrated in Sec. IV. In Sec. V, we draw some conclusions.

II. HAMILTONIAN CHAOTIC SYSTEMS: EXISTENCE OF A LRT BASED ON "GEOMETRY"

Let us consider a system driven by the Hamiltonian:

$$H = H_0 + KH_I, \tag{2.1}$$

where H_0 is the unperturbed system Hamiltonian. The external perturbation has the form

$$KH_I = KAf(t), \tag{2.2}$$

where A is a given function of the phase-space coordinates. The direction of this force is defined by the unit vector $\hat{\mathbf{n}} = -\nabla A$, where ∇ is a gradient operator in the phase-space coordinate. The quantity $f(t)$ is a time-dependent function that specifies how the external field depends on time. We restrict our attention to the case where $f(t)$ is the unit step function and to a spatial displacement $A = x$ so that

$$H = \begin{cases} H_0, & t < 0 \\ H_0 + Kx, & t \geq 0 \end{cases} \tag{2.3}$$

is the Hamiltonian of the system of interest.

We consider systems for which H generates a mixing chaotic dynamics. At times $t = 0$, we assume that the phase-space distribution function of the system is the unperturbed equilibrium one $\rho_{\text{eq},0}(x, v, \mathbf{y}, \mathbf{w})$ satisfying the equation

$$\mathcal{L}_0 \rho_{\text{eq},0}(x, v, \mathbf{y}, \mathbf{w}) = 0, \tag{2.4}$$

where v is the momentum conjugate to the x coordinate and $\mathbf{y}, \mathbf{w}$ refer, respectively, to the canonically conjugate coordinates and momenta of the rest of the system. Here the Liouville operator for the unperturbed system is given by the Poisson brackets:

$$\mathcal{L}_0 \cdot \equiv \{H_0, \cdot\}_{\rm PB} \,. \tag{2.5}$$

In the chaotic regime, for fixed energy E the only solution of Eq. (2.4) is the microcanonical distribution [6]

$$\rho^E_{\rm eq,0}(x, v, \mathbf{y}, \mathbf{w}) = \frac{\delta(E - H_0)}{A_0(E)}, \tag{2.6}$$

where $A_0(E)$ is the normalization of the unperturbed microcanonical distribution on the energy manifold in phase space

$$A_0(E) \equiv \int \delta(E - H_0) dx dv d\mathbf{y} d\mathbf{w}. \tag{2.7}$$

We put an E superscript on the distribution to denote a *fixed energy E*.

At time $t = 0$ the external field is turned on and after some transient time the system reaches an equilibrium distribution $\rho_{{\rm eq},K}$ satisfying the equation

$$\mathcal{L}_0 \rho_{{\rm eq},K}(x, v, \mathbf{y}, \mathbf{w}) + K \mathcal{L}_I \rho_{{\rm eq},K}(x, v, \mathbf{y}, \mathbf{w}) = 0, \tag{2.8}$$

where

$$\mathcal{L}_I \cdot \equiv \{x, \cdot\}_{\rm PB} = \frac{\partial}{\partial v} \cdots \tag{2.9}$$

is the perturbed part of the Liouville operator. For fixed energy E we have again [6] that the solution of Eq. (2.8) is the microcanonical distribution, where now the Hamiltonian is the perturbed one:

$$\rho^E_{{\rm eq},K}(x, v, \mathbf{y}, \mathbf{w}) = \frac{\delta(E - H_0 - Kx)}{A_K(E)}, \tag{2.10}$$

with

$$A_K(E) \equiv \int \delta(E - H_0 - Kx) dx dv d\mathbf{y} d\mathbf{w}. \tag{2.11}$$

The microcanonical distribution function is by inspection a smooth function of K and can be expanded in power series

$$\rho^E_{{\rm eq},K}(x, v, \mathbf{y}, \mathbf{w}) = \rho^E_{\rm eq,0}(x, v, \mathbf{y}, \mathbf{w}) + K \rho^E_{\rm eq,1}(x, v, \mathbf{y}, \mathbf{w}) + O(K^2). \tag{2.12}$$

We obtain $\rho^E_{\rm eq,1}(x, v, \mathbf{y}, \mathbf{w})$ directly from the exact "perturbed" equilibrium distribution of Eq. (2.10), which reads

$$\begin{aligned}\rho^E_{\rm eq,1}(x, v, \mathbf{y}, \mathbf{w}) &= \left[\frac{\partial}{\partial K} \rho^E_{{\rm eq},K}(x, v, \mathbf{y}, \mathbf{w})\right]_{K=0} \\ &= -\frac{x}{A_0(E)} \frac{\partial}{\partial E} \delta(E - H_0) \\ &\quad + \frac{1}{[A_0(E)]^2} \delta(E - H_0) \frac{\partial}{\partial E} \langle x \rangle_{\rm eq,0} \,,\end{aligned} \tag{2.13}$$

where the K derivative is replaced by one with respect to E.

Up to now we have considered equilibrium distributions at fixed energy E, throughout the application of the external field, but actually the switching on of the perturbation at $t = 0$ triggers a transition from a probability distribution at a fixed value of the energy to one with a distribution of energy values. The exact change in energy of each single system of our Gibbs realization depends on the value of the coordinate x at the moment of application of the perturbation. In fact, if at times $t = 0^-$ a particular system were in the phase-space point $x_0, v_0, \mathbf{y}_0, \mathbf{w}_0$, the energy would take the value

$$E^- = H_0(x_0, v_0, \mathbf{y}_0, \mathbf{w}_0). \tag{2.14}$$

Immediately after the application of the external constant force, i.e., at times $t = 0^+$ the value of the energy becomes

$$E^+ = H_0(x_0, v_0, \mathbf{y}_0, \mathbf{w}_0) + K x_0 = E^- + K x_0. \tag{2.15}$$

Thus, the distribution after the application of the external force is no longer the microcanonical distribution of Eq. (2.10), but a collection of microcanonical distributions with different energies E each one corresponding to different x_0, i.e.,

$$\begin{aligned}\rho_{{\rm eq},K}(x, v, \mathbf{y}, \mathbf{w}) = \int & \rho^E_{\rm eq,0}(x_0, v_0, \mathbf{y}_0, \mathbf{w}_0) \\ & \times \rho^{E+Kx_0}_{{\rm eq},K}(x, v, \mathbf{y}, \mathbf{w}) dx_0 dv_0 d\mathbf{y}_0 d\mathbf{w}_0,\end{aligned} \tag{2.16}$$

where

$$\rho^{E+Kx_0}_{{\rm eq},K}(x, y, \mathbf{y}, \mathbf{w}) \equiv \frac{\delta(E + Kx_0 - H_0 - Kx)}{A_K(E + Kx_0)}. \tag{2.17}$$

Although the resulting distribution is not longer microcanonical, it is still smooth and this makes it legitimate to adopt a Taylor series expansion with respect to the strength K of the perturbation. We find the following first-order contribution

$$\begin{aligned}\rho_{\rm eq,1}(x, v, \mathbf{y}, \mathbf{w}) &= \left[\frac{\partial}{\partial K} \rho_{{\rm eq},K}(x, v, \mathbf{y}, \mathbf{w})\right]_{K=0} \\ &= \int \rho^E_{\rm eq,0}(x_0, v_0, \mathbf{y}_0, \mathbf{w}_0) \left\{\frac{\partial}{\partial E} \rho^E_{\rm eq,0}(x, v, \mathbf{y}, \mathbf{w}) x_0 + \rho^E_{\rm eq,1}(x, v, \mathbf{y}, \mathbf{w})\right\} dx_0 dv_0 d\mathbf{y}_0 d\mathbf{w}_0 \\ &= \langle x \rangle_{\rm eq,0} \frac{\partial}{\partial E} \rho^E_{\rm eq,0}(x, v, \mathbf{y}, \mathbf{w}) + \rho^E_{\rm eq,1}(x, v, \mathbf{y}, \mathbf{w}),\end{aligned} \tag{2.18}$$

where $\rho^E_{\rm eq,1}(x, v, \mathbf{y}, \mathbf{w})$ is given by Eq. (2.13) [note that in Eq. (2.13) the energy is kept constant]. Assuming that

$\langle x\rangle_{\text{eq},0} = 0$, we obtain

$$\rho_{\text{eq},1}(x,v,\mathbf{y},\mathbf{w}) = \rho^{E}_{\text{eq},1}(x,v,\mathbf{y},\mathbf{w}). \tag{2.19}$$

This means that in the simplified case where $\langle x\rangle_{\text{eq},0} = 0$, the first-order contribution to the perturbed distribution is identical to the first-order perturbation of the microcanonical distribution, a property which makes it easy to derive the "geometric" expression for the stationary susceptibility. Indeed, using Eq. (2.19) as well as Eq. (2.13), we obtain the following expression for the susceptibility

$$\begin{aligned}\chi &\equiv \int \rho_{\text{eq},1}(x,v,\mathbf{y},\mathbf{w})x dx dv d\mathbf{y} d\mathbf{w} \\ &= -\frac{1}{A_0(E)}\frac{\partial}{\partial E}\int \delta(E-H_0)x^2 dx dv d\mathbf{y} d\mathbf{w} + \int \delta(E-H_0)x dx dv d\mathbf{y} d\mathbf{w}\frac{1}{[A_0(E)]^2}\frac{\partial}{\partial E}\langle x\rangle_{\text{eq},0}, \\ &= -\frac{1}{A_0(E)}\frac{\partial}{\partial E}\left[\langle x^2\rangle_{\text{eq},0}A_0(E)\right] + \frac{\langle x\rangle_{\text{eq},0}}{A_0(E)}\frac{\partial}{\partial E}\left[\langle x\rangle_{\text{eq},0}A_0(E)\right],\end{aligned} \tag{2.20}$$

from which we have

$$\chi = -\frac{1}{A_0(E)}\frac{\partial}{\partial E}\left[\langle x^2\rangle_{\text{eq},0}A_0(E)\right]. \tag{2.21}$$

It is now easy to extend this result to the case when the unperturbed mean value of the variable of interest is not zero. The susceptibility is invariant by spatial translation; then, to obtain the analytical expression for this case, we can simply replace $\langle x^2\rangle_{\text{eq},0}$ in Eq. (2.21) with the variance $(\langle x^2\rangle_{\text{eq},0} - \langle x\rangle^2_{\text{eq},0})$, obtaining

$$\chi = -\frac{1}{A_0(E)}\frac{\partial}{\partial E}\left[(\langle x^2\rangle_{\text{eq},0} - \langle x\rangle^2_{\text{eq},0})A_0(E)\right]. \tag{2.22}$$

Equation (2.22) is an exact expression for the susceptibility, based only on geometric considerations, which circumvents the dynamical arguments of van Kampen criticism. Thus, we have demonstrated the existence of a LRT based on the geometric constraints of the system dynamics in phase space. To be able to say that the system response is linear, however, we should prove that the perturbative contributions following the first one in a power series in the external field intensity K are small compared to the first one. We can expand the exact equilibrium distribution in power series in K [see Eq. (2.12)], we have for the average value of x

$$\langle x\rangle_{\text{eq},K} = K\chi + K^3\beta + O(K^5), \tag{2.23}$$

having assumed that the unperturbed odd moments of the variable x identically vanish [consistently with Eq. (2.21)].

The susceptibility χ is the one in Eq. (2.21), and applying the same analysis which led to (2.21) to Eq. (2.23) yields

$$\beta = -\frac{1}{6A_0(E)}\frac{\partial^3}{\partial E^3}\left[\langle x^4\rangle_{\text{eq},0}A_0\right]. \tag{2.24}$$

We can roughly estimate the contribution β. Defining $\epsilon \equiv E/n$ where n is the number of degrees of freedom, assuming that $A_0 \approx \epsilon^n$ (which is certainly correct for large n), we have that the Eq. (2.23) can be kept to first order when

$$\langle x^2\rangle_{\text{eq},0}/\epsilon \gg \langle x^4\rangle_{\text{eq},0}K^2(n-1)(n-2)/6n^2\epsilon^3, \tag{2.25}$$

which leads to the condition on K

$$K^2 \ll 6\epsilon^2\langle x^2\rangle/\langle x^4\rangle. \tag{2.26}$$

Finally, note that the susceptibility of Eq. (2.21) is evaluated using the stationary equilibrium distribution and it does not explicitly involve the time taken by the observation t, which entered Eq. (1.5), but only the strength of the external field K. Furthermore, recalling that the energy per degree of freedom ϵ is a macroscopic quantity (it coincides with the macroscopic temperature, for canonically distributed systems), it follows that the condition on K given by Eq. (2.26) is a condition on fields of macroscopic intensity.

III. ONLY ONE LRT

In Sec. II we derived an expression for the susceptibility that relied only on the hypothesis of randomization due to chaos and, related to this, we made the assumption that for each value of K the system reaches a unique [6] state of equilibrium, denoted by $\rho_{\text{eq},K}(x,v,\mathbf{y},\mathbf{w})$. Thus our system, after a certain macroscopic time t^* (a time much larger than the microscopic time $\bar{t}$) must be so close to the perturbed equilibrium state as to satisfy the time independent Liouville equation

$$(\mathcal{L}_0 + K\mathcal{L}_I)\,\rho_{\text{eq},K}(x,v,\mathbf{y},\mathbf{w}) = 0. \tag{3.1}$$

If K satisfies Eq. (2.26), we can safely use a perturbation approach to derive from Eq. (3.1) the following first-order perturbation equation for the equilibrium distribution

$$\mathcal{L}_0\rho_{\text{eq},1}(x,v,\mathbf{y},\mathbf{w}) + \mathcal{L}_I\rho_{\text{eq},0}(x,v,\mathbf{y},\mathbf{w}) = 0. \tag{3.2}$$

Formally the solution of Eq. (3.2) reads

$$\rho_{\text{eq},1}(x,v,\mathbf{y},\mathbf{w}) = -\frac{1}{\mathcal{L}_0}\mathcal{L}_I\rho_{\text{eq},0}(x,v,\mathbf{y},\mathbf{w}), \tag{3.3}$$

and this first-order perturbed distribution gives the following formal expression for the stationary susceptibility, for a generic observable B of the system,

$$\chi_B = -\int B\frac{1}{\mathcal{L}_0}\mathcal{L}_I\rho_{\text{eq},0}(x,v,\mathbf{y},\mathbf{w})dx dv d\mathbf{y} d\mathbf{w}. \tag{3.4}$$

For microcanonical systems the formal expression of Eq. (3.4) must be identical to that of Eq. (2.22).

We now prove that *for times t going to infinity Eq. (1.3) (Kubo prediction for the time-dependent susceptibility) converges to the "exact" prediction of (3.4) for the stationary susceptibility.*

Inverting the phase space and time integration (Fubini-Tonelli theorem), and explicitly evaluating the time integration in Eq. (1.3), we get

$$\chi_B^{\mathrm{Kubo}}(t) = -\int B\frac{1}{\mathcal{L}_0}\mathcal{L}_I\rho_{\mathrm{eq},0}(x,v,\mathbf{y},\mathbf{w})dxdvd\mathbf{y}d\mathbf{w} + \left\langle B_0(t)\frac{1}{\mathcal{L}_0}\mathcal{L}_I\right\rangle_{\mathrm{eq},0}. \tag{3.5}$$

Due to the mixing property of the system, the second term on the right hand side of this equation vanishes (we assume that $\langle B\rangle_{\mathrm{eq},0} = 0$). In the long times limit Eq. (3.5) becomes identical to Eq. (3.4). It is ironic that chaos, which is expected to invalidate the Kubo LRT due to the phenomenon of chaos-induced trajectory instability, actually turns out to be the key ingredient to ensure the validity of Kubo prediction.

In the microcanonical case the Kubo susceptibility reads

$$\begin{aligned}\chi^{\mathrm{Kubo}}(t) &= \int dxdvd\mathbf{y}d\mathbf{w}x\int_0^t e^{\mathcal{L}_0\tau}\frac{\partial}{\partial v}\frac{\delta(E-H_0)}{A_0(E)}d\tau\\ &= -\frac{1}{A_0(E)}\int dxdvd\mathbf{y}d\mathbf{w}x\\ &\quad\times\int_0^t v(-\tau)d\tau\frac{\partial}{\partial E}\delta(E-H_0)\\ &= \frac{1}{A_0(E)}\frac{\partial}{\partial E}\left\{\left[\langle xx(t)\rangle_{\mathrm{eq},0} - \langle x^2\rangle_{\mathrm{eq},0}\right]A_0(E)\right\},\end{aligned} \tag{3.6}$$

and, since the system is mixing, in the long times limit the correlation function goes to $\langle x\rangle^2_{\mathrm{eq},0}$. As expected, in this limit it coincides with the exact "geometric" result Eq. (2.22).

We are left with the following picture. There are two time ranges, a microscopic one ($t < \bar{t}$) and a macroscopic one ($t > t^*$), for which the system responds linearly to an external perturbation of macroscopic intensities, and the Kubo LRT is applicable. An open question is what happens in the intermediate time region ($\bar{t} < t < t^*$): Kubo LRT would be, in general, violated.

We now show that *if* a linear response theory (even a "phenomenological" one) exists, *then* it must coincide with Kubo LRT. For instance, this implies that the macroscopic susceptibility derived in [5] *coincides* with Kubo LRT. For the proof, we change the perspective within which the external perturbation is treated. In particular, we assume that we are working at times larger than zero, i.e., that the external perturbation is already switched on. The corresponding correct equilibrium distribution is obviously $\rho_{\mathrm{eq},K}$, and we shall consider the relaxation to this distribution from the initial nonequilibrium distribution $\rho_{\mathrm{eq},0}$. In other words, we are here studying an Onsager process: the distribution $\rho_{\mathrm{eq},0}$ now specifies the initial condition, shifted by an amount proportional to K (for small K) with respect to the equilibrium distribution which is now given by $\rho_{\mathrm{eq},K}$. It is clear that now the dynamics is a pure free relaxation (no external perturbation), where the effect of the external field enters *only* via the definition of the initial conditions at $t = 0$.

We enforce the hypothesis that the system responds linearly. The only possible meaning of this is that the relaxation process is "macroscopically" linear in the initial ($t = 0$) perturbation of the equilibrium distribution: there is indeed no other possible meaning of linearity with respect to K. Let us introduce a macroscopic probability $P(t)$ associated in some way to the microscopic one, for example, via a coarse graining of the space phase. Irrespectively of the actual approach followed for the derivation of $P(t)$, we assume that it is this probability which leads to the macroscopically linear behavior of the system. Notice that the microscopic equilibrium distribution $\rho_{\mathrm{eq},K}$ and the initial one $\rho_{\mathrm{eq},0}$ are smooth on a macroscopic scale. Thus, we have

$$P(\infty) = P_{\mathrm{eq}} = \rho_{\mathrm{eq},K}, \tag{3.7}$$

and

$$P(0) = \rho_{\mathrm{eq},0} = P_{\mathrm{eq}} + KP_1(0) + O(K^2). \tag{3.8}$$

On the other hand, we have already shown that the two stationary distributions $\rho_{\mathrm{eq},0}$ and $\rho_{\mathrm{eq},K}$ are, for small K, close to each other, i.e.,

$$\rho_{\mathrm{eq},0} = \rho_{\mathrm{eq},K} - K\rho_{\mathrm{eq},1} + O(K^2), \tag{3.9}$$

where $\rho_{\mathrm{eq},1}$ is given by Eq. (3.3); hence, from Eqs. (3.7), (3.8), and (3.9) we obtain

$$P_1(0) = -\rho_{\mathrm{eq},1}. \tag{3.10}$$

We now introduce the generator for the time translation, $U(t)$, defined as

$$P(t) = P_{\mathrm{eq}} + KU(t)P_1(0) + O(K^2). \tag{3.11}$$

In general the operator $U(t)$ is not the microscopic Liouvillian operator but it guides the time evolution of the distribution resulting in the macroscopic probability $P(t)$. With the help of Eqs. (3.10) and (3.11) we can write

$$\langle B\rangle(t) = \langle B\rangle_{\mathrm{eq}} + Kg(t) + O(K^2), \tag{3.12}$$

where

$$g(t) \equiv -\int BU(t)\rho_{\mathrm{eq},1}dxdvd\mathbf{y}d\mathbf{w}. \tag{3.13}$$

Equation (3.12) describes the decay process of the average value of B, a function of the microscopic variables. This decay is given by the time behavior of the function $g(t)$, which is a correlation function. In fact, using the explicit form of $\rho_{\mathrm{eq},1}$ given in Eq. (3.3), we have

$$g(t) = -\int BU(t)\frac{1}{\mathcal{L}_0}\mathcal{L}_I\rho_{\text{eq},0}dxdvd\mathbf{y}d\mathbf{w}$$
$$= -\left\langle B(t)\frac{1}{\mathcal{L}_0}\mathcal{L}_I\right\rangle_{\text{eq}} + O(K). \qquad (3.14)$$

Notice that $g(t)$ must be multiplied by K to obtain the first-order contribution to the average value of B, then it must be computed at zero order in K and we can discard the contribution $O(K)$ in Eq. (3.14).

Now we go back to the original problem, i.e., the calculation of the susceptibility to an external perturbation. We express the susceptibility in terms of the relaxation of B given by (3.12), (3.13), and (3.14):

$$\chi_B(t) \equiv \lim_{K\to 0}\frac{\langle B\rangle(t) - \langle B\rangle(0)}{K} = -g(0) + g(t)$$
$$= -\int B\frac{1}{\mathcal{L}_0}\mathcal{L}_I\rho_{\text{eq},0}(x,v,\mathbf{y},\mathbf{w})dxdvd\mathbf{y}d\mathbf{w}$$
$$+\left\langle B(t)\frac{1}{\mathcal{L}_0}\mathcal{L}_I\right\rangle_{\text{eq}}. \qquad (3.15)$$

We stress that in the derivation of the susceptibility in Eq. (3.15) we did *not* linearize the microscopic Liouvillian. We simply enforced the hypothesis of the validity of some (macroscopic) LRT, which implies that Eq. (3.11) holds true, i.e., that an initial small perturbation from equilibrium, linear in K, leads, in the subsequent time evolution, to a deviation of the macroscopic probability from the equilibrium, which is still linear in K.

Comparing Eq. (3.15) with the Kubo result [Eq. (3.5)], it is clear that the difference is that in the former case the correlation function is computed in the presence of the external field, whereas in the latter case the external field is absent. However, both the susceptibility and $g(t)$ ought to be computed at order zero in K. It follows that at zeroth order in K the susceptibility given by Eq. (3.15) coincides with the one obtained following the Kubo approach. We conclude that

(i) If the system responds linearly then it must do so as per the Kubo LRT prediction.

(ii) For Hamiltonian mixing chaotic systems, a breakdown of a LRT can only take place for "intermediate" times.

(iii) If Kubo LRT breaks down, it implies that no other theory based on a linear response will be applicable.

We should appreciate that the result Eq. (3.6) has not been found previously, and it is formally different from the standard one obtained by Kubo which involves the time integral of the mixed correlation function $\langle xv(t)\rangle_{\text{eq},0}$ instead of Eq. (1.3); they are nevertheless equivalent because the only difference is the exchange of the time and phase-space integrations, an exchange which can be carried out for the case at hand. There is yet another difference, i.e., that in the standard case [1] the canonical distribution is used, whereas here we used the microcanonical distribution. However, having assumed that $\langle x\rangle_{\text{eq},0} = 0$, using a canonical equilibrium distribution and repeating the algebra leading to Eq. (3.6), we find for the susceptibility the expression

$$\chi^{\text{Kubo}}_{\text{can}}(t) = \chi^{\text{Kubo}}_{\text{can}}(\infty)\left[1-\varphi(t)\right], \qquad (3.16)$$

where

$$\chi^{\text{Kubo}}_{\text{can}}(\infty) = -\frac{\langle x^2\rangle_{\text{eq},0}}{k_BT} \qquad (3.17)$$

and

$$\varphi(t) \equiv \frac{\langle xx(t)\rangle_{\text{eq},0}}{\langle x^2\rangle_{\text{eq},0}}. \qquad (3.18)$$

If we took the limit of an infinite number of degrees of freedom for our microcanonical system we would expect the susceptibility given by Eq. (3.6) to coincide with the one given by Eq. (3.16), which was obtained working with the canonical distribution. Rewriting Eq. (3.6)

$$\chi^{\text{Kubo}}(t) = \chi^{\text{Kubo}}(\infty)\Bigg\{1-\varphi(t) - \frac{\partial}{\partial E}\varphi(t)$$
$$\times\left[\frac{\partial}{\partial E}\ln\left(A(E)\langle x^2\rangle_{\text{eq},0}\right)\right]^{-1}\Bigg\}, \qquad (3.19)$$

where

$$\chi^{\text{Kubo}}(\infty) \equiv -\frac{1}{A(E)}\frac{\partial}{\partial E}\left(A(E)\langle x^2\rangle_{\text{eq},0}\right) \qquad (3.20)$$

and noticing that

$$\lim_{n\to\infty}\frac{\partial}{\partial E}\langle x^2\rangle_{\text{eq},0}, = 0,$$
$$\lim_{n\to\infty}\frac{\partial}{\partial E}\varphi(t) = 0,$$
$$\lim_{n\to\infty}\frac{\partial}{\partial E}\ln A(E) = (k_BT)^{-1}, \qquad (3.21)$$

we find that, in the limit $n\to\infty$, Eq. (3.19) coincides with Eq. (3.16), which in turn is equivalent to the standard Kubo formula for the susceptibility. We generally term "Kubo susceptibility" the different expressions obtained for the susceptibility, starting from a perturbation of the microscopic Liouvillian: this is so because all these apparently different expressions are equivalent.

IV. NUMERICAL SIMULATIONS AND COMPARISON WITH THE THEORY

To check our conjectures, we have numerically integrated the equations of motion for two different Hamiltonian systems. The first system we have studied numerically is described by the Hamiltonian

$$H_0 = m_1\frac{v^2}{2} + m_2\frac{w^2}{2} + g(m_1x + m_2y). \qquad (4.1)$$

The two particles of masses m_1 and m_2, and coordinates x and y move along the vertical axis under the influence of gravity, and elastically collide between themselves and the ground. We have the constraint $0 \le x \le y$. Under the additional condition that $m_2 < m_1$, it was proved [7] that the system is mixing in the whole accessible phase space and then is ergodic, even when an additional term Kx is added to the Hamiltonian: the mixing character of the dynamics of this system implies that it belongs to the class that we consider in this paper and thus, using the result of the previous sections, we conclude that the LRT should give an exact value for the susceptibility in the long time limit. We have from Eq. (2.22)

$$\chi = \frac{2E}{9\,(m_1+m_2)^2\,g^2}. \tag{4.2}$$

We set $g = 9.8$, $m_1 = 2$, $m_2 = 1$, and $E = 2.0$.

The numerical simulations consisted in generating a microcanonical distribution over the unperturbed phase space, obtained following the unperturbed trajectory and sampling it at times larger than the inverse of the Lyapunov exponent of the system. Then, for each initial condition obtained in this way, we applied the perturbation and followed the subsequent evolution: finally, the average $\langle x(t)\rangle_K$ is computed averaging over the different initial conditions. For very long times we obtained the static susceptibility that is plotted as squares in Fig. 1. The agreement with the theoretical prediction (solid line) of Eq. (4.2) is very good. Because of the very simple geometry of the manifold at given energy, it is easy to analytically evaluate the exact (not first order in K) stationary response of this system ($\langle x\rangle_K - \langle x\rangle_0$), for any value of the intensity K of the external perturbation. The comparison between theory and numerical simulations, carried out at $E = 2.0$ is shown in Fig. 2. The dashed line is the Kubo LRT (first order in K). We can easily estimate that the K values allowed to be in the linear regime is $K \ll g(m_1+m_2)/2 \sim 15$. Note that the satisfactory agreement between the LRT and the numerical result shown by both figures refer to the stationary regime of the response, namely, the long-time regime, while, according to van Kampen's argument, the LRT, being a microscopic treatment of chaotic processes, should hold only in a very restricted short-time regime.

We then turned to the time-dependent case: again we set $E = 2.0$. For the theoretical susceptibility we used Eq. (3.6), with $\langle xx(t)\rangle_{\mathrm{eq},0}$ obtained from the simulations done without any external perturbation and for values of E very close to $E = 2.0$. For the numerical simulations, we followed the same procedure illustrated above, with the only obvious difference that we stored the whole time dependent quantity $\langle x(t)\rangle_K$ for comparison with the theory. The result obtained for two different values of K is plotted together with the theoretical predictions in Fig. 3. Note the very good agreement between simulations and theory in the range of small times. Then, as time increases, we observe a clear departure between the numerical $\chi(t)$ and the theoretical prediction based on Eq. (3.6). Also, the breakdown between theory and simulation happens at smaller times for the larger K value considered. In the region of intermediate times the numerical $\chi(t)$ obtained for different values of K are fairly distinct and their shape is only roughly similar to the theoretical prediction. In the region of large times, as seen in Fig. 1, we would expect the theory and the numerical simulations to agree well. The residual small discrepancy (around four percent for $K = 0.6$) can totally be accounted for using the first nonlinear correction to Kubo LRT. It is clear that, as expected, at intermediate

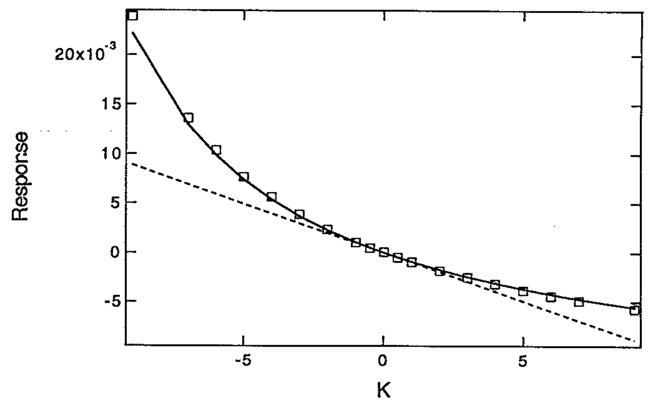

FIG. 2. The stationary response as a function of the external perturbation K for the Hamiltonian of Eq. (4.1). The squares are the result of numerical simulations, the solid line refers to the exact (nonlinear) theoretical prediction, the dashed line is Kubo LRT. See text for the value of the parameters chosen in the simulations.

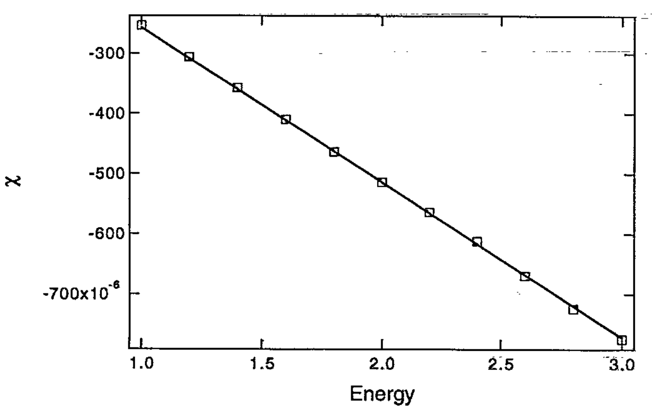

FIG. 1. The stationary susceptibility χ as a function of the energy E for the Hamiltonian of Eq. (4.1). The squares are the result of numerical simulations, the solid line refers to the theoretical prediction of Eq. (4.2). See text for the value of the parameters chosen in the simulations.

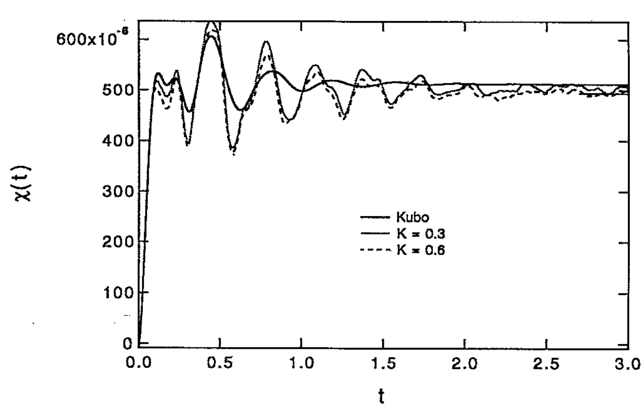

FIG. 3. Time-dependent susceptibility for the Hamiltonian of Eq. (4.1). See figure for a key to the different curves. The curves relative to different K are the result of numerical simulations.

times the discrepancy between numerical simulations and theoretical prediction is much larger than a few percents, lending weight to the conjecture that the Kubo LRT is indeed broken in this time range; in turn this implies that no LRT is applicable, and this is confirmed by the discrepancy between the two numerical simulations shown, which again differ, in the intermediate-time region, by more than a few percents.

The system we studied next is given by the Hamiltonian

$$H_0 = m_1 \frac{v^2}{2} + m_2 \frac{w^2}{2} + \frac{5}{4}\left(x^2 - 1\right)^2 + \frac{3}{2} y \left(x^2 - 1\right) + 2y^2 \tag{4.3}$$

with the choice $m_1 = m_2 = 1$. In this case, we are not aware of a proof that the system is mixing, and hence that its equilibrium distribution is microcanonical. However, studying the unperturbed dynamics we found that for energies E between 0.5 and 2 the microcanonical distribution is satisfactorily realized. A full discussion of the static susceptibility for a wide range of energies has been reported elsewhere [8]. The simulations we present here were done with $E = 1.25$, which guarantees that we are fairly reasonably in the middle of the region for which the LRT should apply. The result of the numerical simulations for three different values of K and the theoretical predictions are plotted in Fig. 4. It is clear that, as expected, the agreement between numerics and theory is extremely good in the region of small times; then a region of poor agreement follows; and finally, for larger times, we again have good agreement between simulations and theory for all values of K considered. A closer look at the region of intermediate times is particularly interesting: it should be clear that the numerical simulations do *not* approach the Kubo formula uniformly as K is decreased. The effect is striking (see the region of times between 30 and 70): looking at the susceptibility for $K = 0.10$ and 0.02 one would be led to conclude that we are indeed approaching the limit given by the Kubo expression. And yet, as K is further reduced, the numerical susceptibility "swings" to the other side of the Kubo expression, implying that no convergence has been achieved. Note that the K values used here are macroscopic, because $K\sqrt{\langle x^2 \rangle}$ is comparable to the energy per degree of freedom, and hence much larger than the K value allowed following the van Kampen argument (see Introduction). We made sure, however, that Eq. (2.26) was satisfied.

Interesting conclusions are reached by magnifying the region of small times. We plot in Fig. 5 the difference between the numerical susceptibilities obtained for different values of K and the Kubo prediction for the susceptibility. We see that the departure from the Kubo prediction follows an exponential function, and, much more remarkably, that the exponent of the best fitting exponential function is very close to the largest Lyapunov exponent for the system, strongly confirming that the breakdown of the LRT at short times is due to the chaotic dynamics, in agreement with van Kampen argument against Kubo LRT.

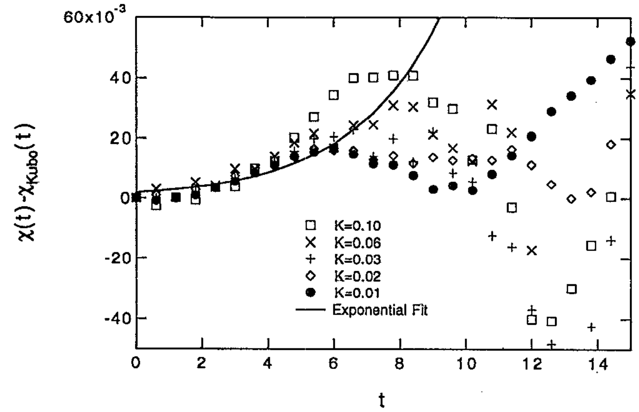

FIG. 5. A short-times region blow up from Fig. 4. Symbols are the difference between the susceptibility obtained from numerical simulations and the Kubo expression, for different values of K (see figure). The solid line is an exponential function, with exponent equal to the largest Lyapunov exponent of the system.

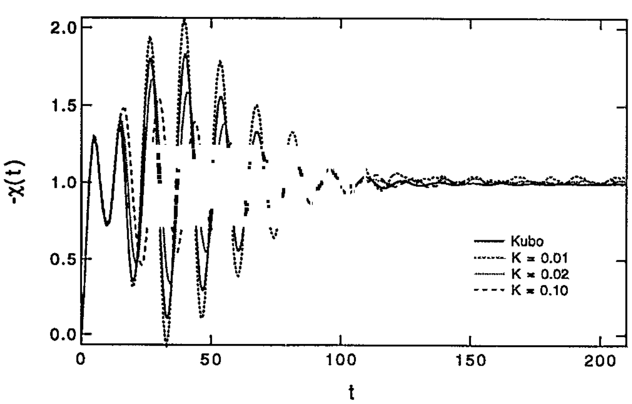

FIG. 4. Time-dependent susceptibility for the Hamiltonian of Eq. (4.3). See figure for a key to the different curves. The curves relative to different K are the result of numerical simulations.

V. CONCLUSIONS

We have studied the class of mixing systems with a finite volume in phase space. We have shown that for large times these systems respond linearly to external perturbations of macroscopic amplitudes. As expected, for large enough amplitudes of the external perturbation this linearity breaks down; we characterized analytically this breakdown, and showed that the perturbations allowed are far larger (macroscopic as opposed to microscopic) than what could be expected, for instance, using the van Kampen argument. For external perturbation amplitudes for which the system of interest responds linearly, the stationary susceptibility that we obtain formally coincides with the one derived by Kubo (LRT).

Intuitively, the reason why for long times the system responds linearly is that the Liouville distribution becomes indistinguishable from a coarse grained macroscopic probability due to the mixing dynamics in the phase space. Note that a fundamental role is played by the finiteness of the volume taken by our systems in phase space. This equivalence is not appropriate for intermediate times, because the fragmentation of the Liouville density induced by mixing dynamics is not yet "complete."

For these systems, the van Kampen scenario is still appropriate to describe the short-times region: the Kubo LRT is applicable for relatively short times (microscopic ones, for sizeable external fields), due to the linearity of the response of the individual trajectories; then the chaotic dynamics takes over, and the linear response breaks down.

In the region of intermediate times we cannot prove that the LRT of Kubo should be applicable; indeed, the numerical simulations in this region do not agree with the Kubo LRT. However, it is most interesting to note that, in this region of disagreement, the numerical susceptibility seems not to converge as the external field amplitude is reduced. We are led to the conclusion that the system does not seem to respond linearly, even for relatively small perturbations.

We have then proved that this behavior could be expected: we have shown that there is no distinction between macroscopic linearity and microscopic linearity. In other words, if a system responds linearly in a macroscopic sense, then the macroscopic susceptibility coincides with the microscopic one, and, vice versa, so that if Kubo LRT breaks down, no macroscopic linearity is to be expected.

What would be the effect of increasing the number of degrees of freedom on the size of the intermediate-time region where the Kubo LRT is invalid, and on the deviations from the Kubo results? The numerical calculations which led us to the discovery of the breakdown of the Kubo LRT are unfortunately limited to the case of a system with only two degrees of freedom. However, on the basis of the physical interpretation given above, we argue that the response of the system becomes more and more linear increasing the number of degrees of freedom of the system. If we are interested in the average of a function of few variables we have that the distribution function, projected on the subspace of these variables, becomes more and more "regular" as one increases the number of degrees of freedom. Thus, we think that the increase of the number of degrees of freedom has the desirable effect of actually making the coarse grained distribution virtually equivalent to the Liouville density, resulting in an unlimited regime of validity for the Kubo LRT.

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