Steady states, invariant measures, and response theory

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Using the method of computer simulation we test the predictions of nonlinear response theory for classical systems subject to dissipative external fields. We provide convincing numerical evidence that Kawasaki methods agree with both the transient time correlation function predictions and with a direct measurement of the nonlinear response. Furthermore, this numerical agreement is observed over a time scale which is sufficiently long for the response to have relaxed to within $\sim 1\%$ of its nonequilibrium steady-state value. This is in spite of the fact that in the steady state the N-particle distribution function ultimately becomes fractal. We discuss the normalization of the Kawasaki distribution and derive a "Lagrangian form" of the Kawasaki response function, and show that it is consistent with predictions that are obtained using a natural invariant measure for nonequilibrium steady states.

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I. INTRODUCTION

In recent years a number of numerical tests [1–3] have been performed on response theory predictions of the nonlinear thermostated response of classical systems to applied dissipative fields. In the most stringent numerical test [1] that has thus far been performed the nonlinear response computed using the so-called [1] transient time correlation function formalism (TTCF), has been shown to be in agreement with the directly observed response to an accuracy of better than 0.15%, for time scales that are sufficiently long that the response has relaxed to within 1% of its steady-state value. For this same system the difference between the observed nonlinear response and the extrapolated linear response was ~30%.

However, another formulation of nonlinear response theory, the so-called Kawasaki formalism [1,4,5] has never been subject to a convincing numerical test. In the same computer experiment as that used to test the TTCF prediction, large statistical uncertainties in the Kawasaki predictions made the test fairly meaningless [1,2]. However, of the two formalisms, the Kawasaki approach has generally proven to be the more useful in terms of predicting new steady-state fluctuation inter-relations for specific heats and compressibilities [1,6,7]. These fluctuation relations have been confirmed to reasonably high numerical accuracy. It is against this background that we present in this paper the first convincing numerical test of the predictions of the Kawasaki response. We also discuss the Kawasaki normalization.

In order to test the predictions of nonlinear response theory, we examine a model system that exhibits the salient features of a general system undergoing nonlinear response. We consider an ensemble of systems each of N particles in a volume V, interacting with an external dissipative field $\mathbf{F}_e(t)=\mathbf{F}_e\Theta(t)$, where $\Theta(t)$ is a unit Heaviside step function. The initial phases $\Gamma\equiv(x_1,y_1,z_1\ldots,p_{yN},p_{zN})$ are assumed to be distributed according to a canonical distribution $f(\Gamma,0)=\exp[-\beta_0 H_0(\Gamma)]/\int d\Gamma \exp[-\beta_0 H_0(\Gamma)]$ with absolute temperature $T_0=1/k_B\beta_0$, where k_B is Boltzmann's constant. The zero-field Hamiltonian is $H_0(\Gamma)=\sum_{i=1}^{N} p_i^2/2m+\Phi$, where m is the particle mass and Φ the total interatomic potential energy.

For $t\geq 0$ the equations of motion for the system are

$$\begin{aligned}\dot{\mathbf{q}}_i&=\frac{\mathbf{p}_i}{m}+C_i\mathbf{F}_e(t)\ ,\\ \dot{\mathbf{p}}_i&=\mathbf{F}_i+D_i\mathbf{F}_e(t)-\alpha\mathbf{p}_i\end{aligned} \tag{1}$$

where $\mathbf{q}_i$ are the particle coordinates, $\mathbf{p}_i$ are the peculiar momenta, and C_i,D_i are phase functions that describe the coupling of the field to the system. The term involving α is known as the thermostat term since it is used to maintain the internal energy H_0 or the kinetic temperature T at a fixed value. It is determined using Gauss' principle of least constraint [1]. The dissipative flux $\mathbf{J}(\Gamma)$ is defined in terms of the adiabatic (i.e., unthermostated) time derivative of the internal energy,

$$\dot{H}_0^{\rm ad}\equiv-\mathbf{J}\cdot\mathbf{F}_e=\alpha\sum\mathbf{p}_i^2/m\ . \tag{2}$$

The thermostat multiplier α that is required to fix the internal energy is

$$\alpha=\frac{-\mathbf{J}\cdot\mathbf{F}_e}{\sum\mathbf{p}_i^2/m}\ . \tag{3}$$

For an arbitrary phase function, $B(\Gamma)$, the nonlinear thermostated response that is obtained under the combined influence of the external field and the thermostat can be written as the TTCF expression [2],

$$\begin{aligned}\langle B(t)\rangle&=\langle B(0)\rangle-\mathbf{F}_e\cdot\int_0^t ds\langle\beta(0)\mathbf{J}(0)B(s)\rangle\\ &=\langle B(0)\rangle+3N\int_0^t ds\langle\alpha(0)B(s)\rangle\ .\end{aligned} \tag{4}$$

Unless otherwise indicated, the time evolution is generated by the field-dependent thermostated equations of

motion (1) and the brackets $\langle\ \rangle$ denote the ensemble average calculated at time t,

$$\langle B(t)\rangle \equiv \int d\Gamma\, B(\Gamma(t))f(\Gamma,0) = \frac{\int d\Gamma\, B(\Gamma(t))\exp[-\beta_0 H_0(\Gamma)]}{\int d\Gamma \exp[-\beta_0 H_0(\Gamma)]} . \quad (5)$$

In (4) we have used the fact that at constant internal energy $\beta(t)\mathbf{F}_e\cdot\mathbf{J}(t) = -3N\alpha(t)$, where $\beta(t)\equiv 3Nm/\sum \mathbf{p}_i^2$. We note that for strong applied fields, $\mathbf{F}_e$, $\langle\beta(t)\rangle \neq \beta_0$, since $H_0(t)=H_0(0)$.

From (4) it is trivial to see that in the zero-field limit the linear response (if it exists) can be written

$$\lim_{F_e\to 0}\langle B(t)\rangle = \langle B(0)\rangle - \beta_0\mathbf{F}_e\cdot\int_0^t ds\,\langle \mathbf{J}(0)B(s_0)\rangle , \quad (6)$$

where the zero subscript on the time argument denotes the fact that the time dependence is generated by the thermostated *field free* equations of motion. Equation (6) is a thermostated generalization of the well-known Green-Kubo relation for the linear response.

An equivalent form for the nonlinear thermostated response is the so-called "bare form," BK, of the Kawasaki response [5],

$$\langle B(t)\rangle = \left\langle B(0)\exp\left[3N\int_0^t ds\,\alpha(-s)\right]\right\rangle \quad (7)$$

while the "renormalized," RK, expression for the Kawasaki nonlinear response is

$$\langle B(t)\rangle = \frac{\left\langle B(0)\exp\left[3N\int_0^t ds\alpha(-s)\right]\right\rangle}{Z(t)} , \quad (8)$$

where the denominator, $Z(t)$, defined as

$$Z(t)\equiv\left\langle \exp\left[3N\int_0^t ds\alpha(-s)\right]\right\rangle = \int d\Gamma \exp\left[-3N\int_0^{-t} ds\,\alpha(s)\right] f(\Gamma,0) , \quad (9)$$

is the Kawasaki normalization factor. It is easy to show that in the linear regime both the Kawasaki forms [(7)–(9)] reduce to the Green-Kubo linear response expression (6).

When we say that each of these expressions [(4) and (7)–(9)] are equivalent we mean that their identity can be proven using the thermostated field-dependent Liouville equation, $\partial f(\Gamma,t)/\partial t = -\partial[f(\Gamma,t)\dot{\Gamma}]\cdot/\partial\Gamma$, where $f(\Gamma,t)$ is the N-particle distribution function at time t evaluated under the combined influence of the external field and the thermostat. The identity of Eqs. (7) and (8) follows from the fact that one can show from the Liouville equation that $Z(t)=1$ for all values of t.

Both the Kawasaki forms involve averages of exponentials of integrals of extensive quantities and hence are exceedingly difficulty to calculate. Typically averages computed using the renormalized Kawasaki expression, (8), have a smaller variance than those using the bare form, (7). From previous numerical data it was by no means obvious that $Z(t)=1$ at long times. Previous numerical data seemed to indicate that $Z(t)$ is a monotonically decreasing function of time. However, the extreme difficulty of carrying out such calculations has meant that the situation was largely unresolved.

Hoover, Morriss and co-workers have shown that in the nonequilibrium steady state, the distribution function becomes fractal [1,8-11]. Holian *et al.* [10] argued that the fractal nature of the distribution function means that although for finite times the TTCF expression is correct, both of the Kawasaki forms for the nonlinear response are incorrect. However, no numerical data were provided in support of this assertion.

In the present paper we present computer simulation data comparing the directly observed transient nonlinear response with the TTCF, Kawasaki and renormalized Kawasaki theoretical predictions. Our results show, to unprecedented accuracy ($<1\%$), that numerical agreement between theory and experiments is excellent. We argue that the concerns raised in [10] regarding the nonanalytic nature of the fractal steady-state distribution function, although correct in principle, are not relevant in practice. The reason is that nonlinear response theory predictions are for the finite time *transient* response. The distribution functions on the other hand only become truly fractal (to the arbitrarily small phase-space length scales required for nonanalyticity) in the infinite time limit. For any finite time, no matter how large, the distribution functions are analytic and only *approximate* the steady-state fractal attractors. Hence for any finite time, no matter how long, nonlinear response theory predictions are correct.

In this work we also discuss the Kawasaki normalization. This is the key to a clear understanding of nonlinear response theory. Finally we give a derivation of a long sought connection between nonlinear response theory and the dynamical systems theory description of the fractal steady-state attractors.

Recently Evans, Cohen, and Morriss [12] have derived a formula for the ratio of probabilities of observing, in a steady state, finite duration trajectory segments and their time-reversed trajectory antisegments. This ratio formula was derived from a natural invariant measure (which we shall call the Lyapunov instability measure), originally proposed by Eckmann and Procaccia [13] for the steady-state attractor. In the present paper we show that the ratio formula for observing segments and antisegments can also be derived from a new generalization of the Kawasaki expressions of nonlinear response theory. When combined with accurate numerical tests of nonlinear response theory predictions, this theoretical result provides a long sort link between the two apparently different approaches to describing nonequilibrium steady states.

II. THE KAWASAKI NORMALIZATION

If the equations of motion are integrated exactly, the Liouville equation can be used to show that the normalization factor (9), $Z(t)$, is unity. Since $Z(0)$ is a phase integral of the normalized equilibrium distribution function, $Z(0)$, is unity. The rate of change of $Z(t)$ after the external field is switched on and the thermostat applied,

is given by

$$\frac{dZ(t)}{dt}=\int d\Gamma f(\Gamma,0)\frac{\partial}{\partial t}\exp\left[3N\int_0^t ds\,\alpha(-s)\right]$$
$$=3N\int d\Gamma f(\Gamma,t)\alpha(-t)$$
$$=3N\int d\Gamma f(\Gamma,0)\alpha(0)=0\ . \tag{10}$$

In deriving the last line of this equation we have used the Schrödinger-Heisenberg equivalence [1] to show that $\int d\Gamma f(\Gamma,t)\alpha(\Gamma(-t))=\int d\Gamma f(\Gamma,0)\alpha(\Gamma)=\langle\alpha(0)\rangle=0$. This implies that the bare Kawasaki distribution is normalized for all times provided the dynamical equations of motion are solved essentially exactly and the N-particle distribution is therefore governed by the Liouville equation. Of course as time increases in any real system it will eventually become impossible for the forward and reverse propagators in line 2 of Eq. (10), $f(\Gamma,t)$ and $\alpha(\Gamma(-t))$, respectively, to annihilate each other. When this occurs, the proof of normalization given in (10) breaks down.

The time over which the forward and reverse propagators annihilate each other can be measured by the so-called lifetime of the antisteady state, $t_{1/2}$. This lifetime is defined in the following manner. In a typical nonequilibrium steady state the average entropy production is positive and the average dissipative flux is negative. Since the equations of motion are time reversible, when we apply the time-reversal mapping to any typical phase, sampled in the steady state, the trajectory will reverse itself *exactly* with negative entropy production. We call such a trajectory segment an *antisegment* and we say that the positive entropy producing trajectory segment from which it was constructed is its *conjugate*. In reversible systems with reversible thermostats every trajectory segment has a conjugate antisegment. The existence of this conjugacy is a sufficient condition for the Kawasaki normalization to be unity.

We will now consider the Kawasaki normalization in more detail. Without the loss of generality we assume the dissipative flux J is odd under time-reversal mapping M^T: $M^T(q,p,F_e)-(q,-p,F_e)$. It is then straightforward to show that $M^T iL(\Gamma,F_e)=iL(\Gamma^T,F_e)=-iL(\Gamma,F_e)$. From this it is easy to show that

$$J(-s,\Gamma,F_e)=-J(s,\Gamma^T,F_e),$$
$$\alpha(-s,\Gamma,F_e)=-\alpha(s,\Gamma^T,F_e)\ . \tag{11}$$

Applying this to the Kawasaki normalization (9), gives

$$Z(t)=\int d\Gamma\exp\left[3N\int_0^t ds\,\alpha(\Gamma(-s))\right]f(\Gamma,0)$$
$$=\int d\Gamma\exp\left[-3N\int_0^t ds\,\alpha(\Gamma^T(s))\right]f(\Gamma^T,0)$$
$$=\int d\Gamma\exp\left[-3N\int_0^t ds\,\alpha(\Gamma(s))\right]f(\Gamma,0)=Z(-t)\ , \tag{12}$$

where we have used that $|\partial\Gamma^T/\partial\Gamma|=1$, in obtaining the last line of (12).

Intuitively, Eq. (10) is somewhat unexpected since the overwhelming majority of trajectory segments will be entropy increasing, which implies for each of these segments the time-averaged thermostat multiplier will be positive and the dissipative flux negative, leading to a negative exponent in that segment's contribution to $Z(t)$, [see (12)]. Therefore, in the overwhelming majority of trajectory segments, as $t\to\infty$, $\exp(-3N\langle\alpha\rangle_t)t\to 0$, where $\langle\cdots\rangle_t$ denotes a time average calculated over a time t.

However, in those rare cases where an entropy reducing trajectory segment is observed, the exponent will be positive and because of the highly nonlinear dependence of an exponential upon its argument, such entropy reducing segments, although rare, will have a highly enhanced effect on the determination of $Z(t)$. It turns out that these two effects cancel exactly: the rarity of observing antisegments exactly cancels their exponential effect in the Kawasaki exponent leading to the normalization being unity.

To show this, it is convenient to consider a small phase-space volume $V(\Gamma(0))$, about an initial phase, $\Gamma(0)$. We can analyze the time evolution of such a volume by writing the solution of the Louiville equation,

$$\frac{\partial f(\Gamma,t)}{\partial t}\equiv -i\mathcal{L}f(\Gamma,t)=-\dot\Gamma\cdot\frac{\partial f(\Gamma,t)}{\partial\Gamma}+f(\Gamma,t)\frac{\partial}{\partial\Gamma}\cdot\dot\Gamma$$
$$=-\dot\Gamma\cdot\frac{\partial f(\Gamma,t)}{\partial\Gamma}+3N\alpha(\Gamma)f(\Gamma,t)+O(1)\ . \tag{13}$$

The formal solution of the Liouville equation can be written in terms of the distribution function propagator, $\exp[-i\mathcal{L}(\Gamma)t]$, as $f(\Gamma,t)=\exp[-i\mathcal{L}(\Gamma)t]f(\Gamma,0)$. Clearly one can write

$$\exp[i\mathcal{L}(\Gamma)t]f(\Gamma,0)=f(\Gamma,-t)\ . \tag{14}$$

However, since this equation is true for all Γ it must also be true for $\Gamma(-t)$, so that

$$\exp[i\mathcal{L}(\Gamma(-t))t]f(\Gamma(-t),0)=f(\Gamma(-t),-t)\ . \tag{15}$$

Using a Dyson decomposition of the distribution function propagator in terms of the phase function propagator, $\exp[iL(\Gamma)t]$, where $A(\Gamma(t))=\exp[iL(\Gamma)t]A(\Gamma)$, one can show [1] that

$$\exp[i\mathcal{L}(\Gamma)t]=\exp\left[-\int_0^t 3N\alpha(\Gamma(s))ds\right]\exp[iL(\Gamma)t]\ . \tag{16}$$

Substituting Eq. (16) into (15) gives

$$f(\Gamma(-t),-t)=\exp\left[-\int_0^t 3N\alpha(\Gamma(s-t))ds\right]$$
$$\times\exp[iL(\Gamma(-t))t]f(\Gamma(-t),0)$$
$$=\exp\left[-\int_0^t 3N\alpha(\Gamma(s-t))ds\right]f(\Gamma(0),0)$$
$$=\exp\left[\int_0^{-t}3N\alpha(\Gamma(s))ds\right]f(\Gamma(0),0) \tag{17}$$

and therefore,

$$f(\Gamma(t),t)=\exp\left[\int_0^t 3N\alpha(\Gamma(s))ds\right]f(\Gamma(0),0)\ . \tag{18}$$

We call this equation the Lagrangian form of the

Kawasaki distribution, LK. It should be contrasted with the usual Kawasaki expression for the nonlinear N-particle distribution function which can easily be obtained from (7):

$$f(\Gamma,t)=\exp\left[\int_0^t 3N\alpha(\Gamma(-s))ds\right]f(\Gamma,0)$$
$$=\exp\left[-\int_0^{-t}3N\alpha(\Gamma(s))ds\right]f(\Gamma,0)\ . \quad (19)$$

Equation (18) shows that from almost every initial phase, $\Gamma(0)$, the distribution function along the trajectory, $\Gamma(t)$, diverges to positive infinity. Equation (19) on the other hand says that at time t, almost everywhere in phase space, Γ, the distribution function collapses towards zero. These two statements are consistent with the conservation of total probability.

Equation (18) enables us to characterize the time-dependent evolution of a small, comoving, phase-space volume, $V(\Gamma(t))$, about a moving phase vector, $\Gamma(t)$. If this volume initially contains M ensemble members then $V(\Gamma(t))=M/f(\Gamma(t),t)$. Using (18) we can show

$$V(\Gamma(t))=V(\Gamma(0))\exp\left[-\int_0^t ds\ 3N\alpha(\Gamma(s))\right]\ . \quad (20)$$

This equation shows that for normal positive entropy producing trajectories ($\langle\alpha\rangle_t>0$), which are produced from almost all starting phases $\Gamma(0)$, the streaming phase-space volume element undergoes contraction. Substituting this equation into (12) shows

$$Z(t)=\langle V(\Gamma(t))/V(\Gamma(0))\rangle\ . \quad (21)$$

We can now analyze the time evolution of the Kawasaki normalization as a result of an individual trajectory segment i and its conjugate antisegment i^*. These segments are depicted schematically in Fig. 1. Without loss of generality the figure shows segment i as a positive entropy producing segment with a negative value for the average dissipative flux. As was discussed in detail in Ref. [14] segment i^* can be constructed from segment i by applying a time-reversal mapping M^T to the phase $\Gamma_i(t)$ of segment i at time t. Following this time-reversed phase *backwards* in time to $t=0$ is equivalent to following the original $\Gamma_i(0)$ phase forward in time from t to $2t$.

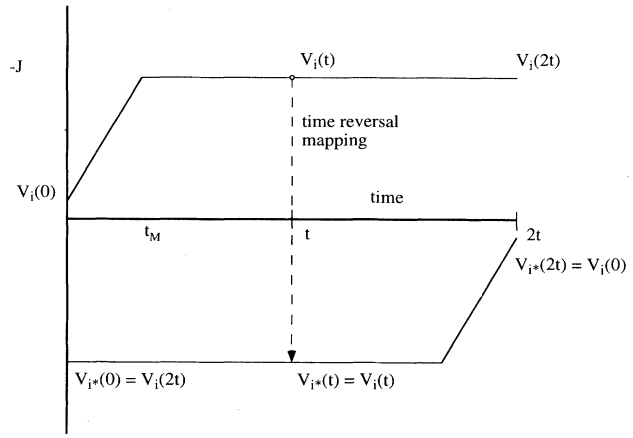

FIG. 1. A schematic diagram depicting the time evolution of the dissipative flux of a trajectory segment i and its antisegment i^*. Segment i^* is constructed from segment i by applying a time-reversal mapping to the phase, $\Gamma_i(\tau)$, of segment i at time τ. For convenience we denote $\Gamma_i(0)$ as 1 , $\Gamma_i(\tau)$ as 2, $\Gamma_i(2\tau)$ as 3, $\Gamma_{i*}(0)$ as 4, $\Gamma_{i*}(\tau)$ as 5, and $\Gamma_{i*}(2\tau)$ as 6.

Because the Jacobian of the time-reversal map is unity, $V_{i*}(t)=V_i(t)$, and from the reversibility of the equations of motion $V_{i*}(0)=V_i(2t)$ while $V_{i*}(2t)=V_i(0)$.

The normalization $Z(t)$ can be written in terms of comoving Lyapunov volumes, as

$$Z(t)\sim\sum_i V(\Gamma_i(0))\frac{V(\Gamma_i(t))}{V(\Gamma_i(0))}+V(\Gamma_{i*}(0))\frac{V(\Gamma_{i*}(t))}{V(\Gamma_{i*}(0))}\ , \quad (22)$$

where we have used the fact that in the microcanonical ensemble the probability of observing phases inside a volume is simply proportional to the magnitude of that volume (in other ensembles this becomes asymptotically true at $t\to\infty$). Using the fact that $V(\Gamma_{i*}(0))=V(\Gamma_i(0))\exp\left[-\int_0^{2t}3N\alpha(s_i)ds\right]$ (see Fig. 1 and Ref. [14]), the last relation can be written as

$$Z(t)\sim\sum_i V(\Gamma_i(0))\exp[-3N\langle\alpha\rangle_i t]+V(\Gamma_{i*}(0))\exp[-3N\langle\alpha\rangle_{i*}t]$$
$$\sim\sum_i V(\Gamma_i(0))\exp[-3N\langle\alpha\rangle_i t]+V(\Gamma_{i*}(0))\exp[+3N\langle\alpha\rangle_i t]$$
$$\sim\sum_i V(\Gamma_i(0))\exp[-3N\langle\alpha\rangle_i t]+V(\Gamma_i(0))\exp[-3N\langle\alpha\rangle_i 2t]\exp[+3N\langle\alpha\rangle_i t]$$
$$\sim\sum_i V(\Gamma_i(0))\exp[-3N\langle\alpha\rangle_i t]+V(\Gamma_i(0))\exp[-3N\langle\alpha\rangle_i t]\ . \quad (23)$$

Thus the contribution of the antisegment i^* to the normalization at time t is *exactly* the same as that made by its conjugate, namely, i. The rarity of observing antisegments is *exactly* canceled by the exponentially enhanced effect these expanding phase volumes have on the normalization.

III. THE KAWASAKI NORMALIZATION: WEAK FIELDS

It is often not realized that the problem of Kawasaki normalization also arises in the linear response regime. In this regime, we can, however, use the special results of

linear response theory to analyze the function. $Z(t)$ can be expressed as

$$\left\langle \exp\left[\beta \mathbf{F}_e \cdot \int_0^{-t} ds\, \mathbf{J}(s)\right]\right\rangle = \int_{-\infty}^{\infty} d\langle J\rangle_{t,F_e} p(\langle J\rangle_{t,F_e}) \times \exp(\beta_0 F_e \langle J\rangle_{t,F_e} t)\ , \tag{24}$$

where

$$\langle J\rangle_{t,F_e} \equiv 1/t \int_0^{-t} ds\, \mathbf{J}(s)\cdot \mathbf{F}_e / F_e \tag{25}$$

is the time average value of the dissipative flux, J, along the trajectory segment of length t, in the presence of the field F_e and the thermostat. $p(\langle J\rangle_{t,F_e})$ is the probability of observing a t segment with value $\langle J\rangle_{t,F_e}$. We note that in Eq. (25), $t=0$ refers to the initial time when the phases are distributed canonically. Thus as time increases the ensemble passes from an equilibrium state through a transient period into the steady state as $t\to\infty$. In this limit we expect that the influence of the transient states to the averages $\langle J\rangle_{t,F_e}$ will be statistically insignificant. We also expect that in the same limit, successive t averages of $\langle J\rangle_{t,F_e}$ will be uncorrelated.

The central limit theorem can be used to deduce the distribution of $\langle J\rangle_{t,F_e}$,

$$p\left[\langle J\rangle_{t,F_e}\right] = \frac{1}{\sqrt{2\pi}\sigma} \exp\left[-\frac{(\langle J\rangle_{t,F_e} - \bar{J}_{F_e})^2}{2\sigma^2}\right] \tag{26}$$

where $\bar{J}_{F_e}$ is the mean of $\langle J\rangle_{t,F_e}$, which is independent of t provided that t is sufficiently large that the contributions to averages caused by the initial transients are insignificant. The variance of $\langle J\rangle_{t,F_e}$ is

$$\sigma^2 = \langle(\langle J\rangle_t - \bar{J}_{F_e})^2\rangle = \frac{1}{t^2}\left\langle \int_0^{-t} ds_1 \int_0^{-t} ds_2\, J(s_1)J(s_2)\right\rangle - \bar{J}_{F_e}^2\ . \tag{27}$$

Using a change of variables, $\tau_1 = s_1 - s_2$ and $\tau_2 = s_1 + s_2$, and the fact that in the mixing steady state, correlation functions only depend on the time *differences*, we can see that

$$\begin{aligned}\langle J(s_1)J(s_2)\rangle &= \left\langle J\left[\frac{\tau_2}{2}+\frac{\tau_1}{2}\right] J\left[\frac{\tau_2}{2}-\frac{\tau_1}{2}\right]\right\rangle \\ &= \left\langle J\left[\frac{\tau_2}{2}+\tau_1\right] J\left[\frac{\tau_2}{2}\right]\right\rangle, \quad \forall \tau_2 \\ &= \langle J(\tau_1)J(0)\rangle, \quad \forall \tau_2\ .\end{aligned} \tag{28}$$

If we invoke the assumption of local thermodynamic equilibrium in the $F_e\to 0$ limit, the variance [15] is given by

$$\begin{aligned}\lim_{t\to\infty}\lim_{F_e\to 0} t\sigma^2 &= \lim_{t\to\infty}\lim_{F_e\to 0} t\left[\frac{1}{t^2}\int_0^t d\tau_2 \int_{-\tau_2}^{\tau_2} d\tau_1 [\langle J(\tau_1)J(0)\rangle - \bar{J}_0^2]\right] = \lim_{t\to\infty}\lim_{F_e\to 0}\left[\frac{2}{t}\int_0^t d\tau_2 \int_0^{\tau_2} d\tau_1 \langle \Delta J(\tau_1)\Delta J(0)\rangle\right] \\ &= \lim_{t\to\infty}\lim_{F_e\to 0}\left[\frac{2}{t}\int_0^t d\tau_1 \int_{\tau_1}^{t} d\tau_2 \langle \Delta J(\tau_1)\Delta J(0)\rangle\right] = \lim_{t\to\infty}\left[\frac{2}{t}\int_0^t d\tau (t-\tau)\langle \Delta J(\tau_0)\Delta J(0)\rangle\right] \\ &= \frac{2\tilde{L}(0)}{\beta} + \left[\frac{2\tilde{L}'(0)}{\lim \beta_0 t}\right] = \lim_{F_e\to 0}\frac{2\bar{J}_{F_e}}{\beta_0 F_e}\ ,\end{aligned} \tag{29}$$

where $\tilde{L}(s) \equiv \int_0^\infty dt\, e^{-st}\langle \Delta J(t_0)\Delta J(0)\rangle$ denotes a Laplace transform of an *equilibrium* time correlation function and we have assumed that both the Laplace transform and its derivative with respect to s, exist.

The Green-Kubo expression for the transport coefficient $L(F_e=0)$ where $\langle J\rangle = -L(F_e)F_e$, is obtained from (6) by setting $B \equiv J : L(0) = -\beta_0 \int_0^\infty dt \langle J(t_0)J(0)\rangle$.

Substituting (26) and (29) into (24) shows that, in the limits where $\tau \gg \tau_M$ and $F_e \to 0$,

$$\begin{aligned}\lim_{F_e\to 0} Z(t) &= \lim_{F_e\to 0}\int_{-\infty}^{\infty} d\langle J\rangle_{t,F_e} \frac{1}{\sqrt{2\pi}\sigma}\exp\left[-\frac{(\langle J\rangle_{t,F_e} - \bar{J}_{F_e})^2}{2\sigma^2} + \beta_0 F_e \langle J\rangle_{t,F_e} t\right] \\ &= \lim_{F_e\to 0}\int_{-\infty}^{\infty} d\langle J\rangle_{t,F_e} \frac{1}{\sqrt{2\pi}\sigma}\exp\left[-\frac{(\langle J\rangle_{t,F_e} + \bar{J}_{F_e})^2}{2\sigma^2}\right] = 1\ .\end{aligned} \tag{30}$$

Thus in the linear regime close to equilibrium, we see that the Kawasaki normalization, $Z(t_0)$, can be expected to be unity provided that the fluctuations in the dissipative flux satisfy the Green-Kubo relaxation. Sufficient conditions for this to occur are that the derivative of the zero-frequency Laplace transform exists and that the equations of motion are accurate over the time scale required for the system to relax to the steady state, which is of the order of the Maxwell relaxation time τ_M. This is much weaker than the requirement that the equations be accurate for the times over which $Z(t_0)$ is actually calculated.

IV. NONEQUILIBRIUM MOLECULAR-DYNAMICS SIMULATIONS

The validity of the TTCF and the Kawasaki expressions for nonlinear response was examined using nonequilibrium molecular-dynamics simulations of a system that was subject to a color field. A two-dimensional system of two disks with periodic boundary conditions was studied using the self-diffusion algorithm [16]. The response of the color current was observed when a constant color field is applied to the equilibrium ensemble. The disks were characterized by a simple short-ranged pair potential, $\phi(r)$, which is, in reduced units,

$$\phi(r)=\begin{cases}4[r^{-12}-r^{-6}]+1 & \text{for } r<2^{1/6}\\ 0 & \text{for } r>2^{1/6}\end{cases}. \tag{31}$$

This potential is known as the WCA (Weeks-Chandler-Andersen) potential [17]. Reduced units, where the mass is unity, will be used throughout this section.

The self-diffusion algorithm assumes the system consists of two species that differ only by a color label. The equations of motion for an N-body system of color labeled species subject to a constant field, F_e, in the x direction can be derived from the color Hamiltonian [13] and are given by

$$\begin{aligned}\dot{\mathbf{q}}_i&=\mathbf{p}_i/m\ ,\\ \dot{\mathbf{p}}_i&=\mathbf{F}_i+\mathbf{i}c_iF_e-\alpha(\mathbf{p}_i-m\mathbf{u}_{s_i})\ ,\end{aligned} \tag{32}$$

where $\mathbf{i}$ is the unit vector in the x direction, $c_i=(-1)^i$ is the color label of particle i, ρ is the particle density, $\mathbf{u}_{s_i}$ is the streaming velocity of atom i and the term $\alpha(\mathbf{p}_i-m\mathbf{u}_{s_i})$ is the Gaussian thermostat, which is used to constrain the temperature. In the definition of the temperature, the peculiar particle velocity relative to the streaming velocity of each species is used. That is, the constraint equation is given by

$$\sum_{i=1}^{N}(\mathbf{p}_i-m\mathbf{u}_{s_i})^2/m=(2N-3)k_BT\ . \tag{33}$$

Hence, the thermostat multiplier is

$$\alpha=\frac{\sum_{i=1}^{N}\mathbf{F}_i\cdot(\mathbf{p}_i-m\mathbf{u}_{s_i})}{\sum_{i=1}^{N}\mathbf{p}_i\cdot(\mathbf{p}_i/m-\mathbf{u}_{s_i})}\ . \tag{34}$$

In general, the streaming velocity, $\mathbf{u}_{s_i}$, is given by $\mathbf{i}c_iJ_x/\rho$, however, in the two-particle system it is not possible to define an instantaneous streaming velocity, therefore it is assumed to be zero.

A periodic system of WCA disks was studied and the response of the dissipative flux was monitored, which in this case is the color current density J_x,

$$J_x=\frac{1}{V}\sum_{i=1}^{N}c_i\dot{q}_{x_i}\ . \tag{35}$$

The nonequilibrium molecular-dynamics simulations used the fourth-order Runge-Kutta method to integrate the equations of motion with a time step $\Delta t=0.005$. The Runge-Kutta method is self-starting, which is necessary for an examination of the transient response. A density of $\rho=0.396\,850$ (which is sufficiently low that the box length is greater than twice the range of the WCA potential) and a temperature of $T=1.0$ was used. The color labels for the two species were $c_1=+1$ and $c_2=-1$. After equilibration a constant color field was applied.

The response of the color current density for the two-particle periodic system calculated using the TTCF, the bare Kawasaki, and the renormalized Kawasaki expressions was compared with the direct ensemble average. Color fields of $F_e=0.1$ and $F_e=1.0$ were used. In each case, the average was carried out over a set of 2×10^6 starting states from the isothermal equilibrium ensemble. In the simulations these were generated as phase-space points of a single trajectory, which are 4800 time steps apart, which is sufficiently long that they are not correlated. For efficiency, phase-space maps were carried out at each of the generated phases to produce additional starting points from the isothermal equilibrium ensemble. For each starting phase, $\Gamma=(q_x,q_y,p_x,p_y)$ a time-reversal mapping $M^T(\Gamma)=(q_x,q_y,-p_x,-p_y)$ was used.

Figure 2(a) shows the response of the color current

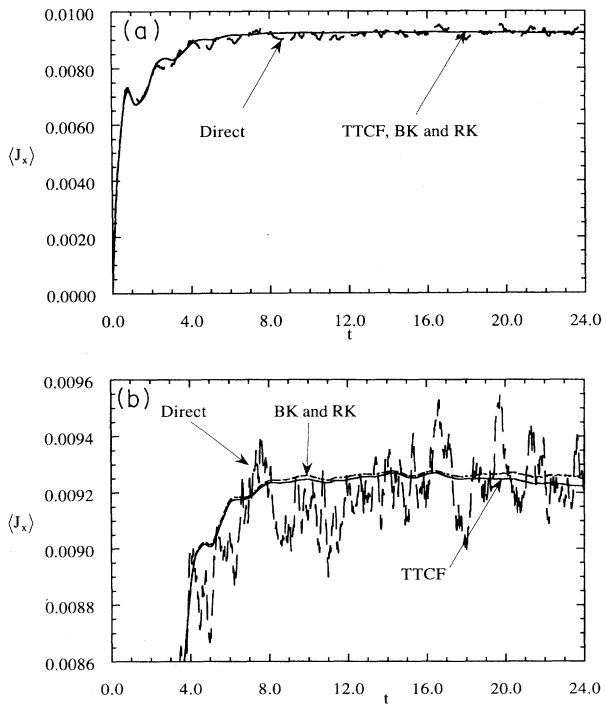

FIG. 2. (a) The color current density response to a constant field of 0.1 determined using nonequilibrium molecular-dynamics simulation. The response calculated using a direct ensemble average (— — —) is compared with that calculated using the TTCF (——), bare Kawasaki (BK) (- - - - - -) and renormalized Kawasaki (RK) (—-—-) expressions. The color field was applied to a microcanonical equilibrium ensemble of 2×10^6 initial configurations. The system consisted of two WCA disks in a periodic box at $T=1.0$ and $\rho=0.396\,850$. (b) The scale in (a) is expanded for part of the data.

density to a color field of $F_e = 0.1$. The field was applied at $t = 0$ and averaged over 2×10^6 starting states. In Fig. 2(b), a section of the same data is magnified. The figures show that for a color field of 0.1 the bare Kawasaki (BK) and renormalized Kawasaki (RK) expressions are in excellent agreement with each other ($\pm 0.05\%$) and with the comparatively low accuracy ($\pm 5.0\%$) direct results. The BK and TTCF results disagree with each other by no more than 0.3%. The ensemble averaged current is highly structured and is not a monotonic increasing function of time. This is a reflection of the growing underlying complexity of the phase-space distribution function.

The half life of the antisteady state, which is a function of our integration accuracy, for this system is ~ 8. Therefore the accuracy of the Kawasaki expressions and the TTCF expression is maintained for times that are much longer than the decay time of the antisteady state. This decay time is a measure of the time over which microscopic reversibility holds. Throughout the time scale shown on Fig. 2(a), the Kawasaki renormalization factor is unity, within estimated statistical uncertainties ($\pm 0.04\%$). Thus microscopic reversibility is apparently *not* a necessary condition for the Kawasaki normalization to be unity, at least for relatively weak fields. This is in agreement with our theoretical discussion in Sec. III.

In Fig. 3(a) the response of the color current density of the same system to a color field of 1.0 is shown. Averages are carried out using 2×10^6 initial configurations. For $F_e = 1.0$, the results determined using the direct method are more precise than for $F_e = 0.1$ and these results are in agreement to within 0.7% with those obtained using the TTCF expression. For $t < 6.5$, both the bare Kawasaki and renormalized Kawasaki expressions agree with the direct response to within 1.0% and 0.7%, respectively. After this time, larger fluctuations occur with the renormalized Kawasaki expression generally in better agreement than the bare expression. In Fig. 3(b) the deviation of the results obtained using the TTCF, renormalized Kawasaki, and bare Kawasaki response expression from the direct response results are plotted. The deviation of the TTCF expression has been magnified by 100 for clarity and is just 0.2% at $t = 12$. The agreement of the TTCF expression and the directly calculated results indicates that the Green-Kubo-type expressions are accurate far from the thermodynamic limit—in this case for just 2 particles.

In Fig. 4 the time evolution of a selected region of the coordinate space pair distribution function is shown for this system with a field of $F_e = 1.0$ applied at $t = 0$. Clearly a developing structure is observed soon after the field is applied and this structure becomes more distinct as time progresses. The structure that is observed here in the pair distribution function is a projection of the developing fractal characteristics of the full phase-space distribution. Importantly, the departure of the Kawasaki-based averages from their directly computed counterpart does not appear to correspond to the emergence of the fractal characteristics in the projected phase-space distribution function [10]. These results indicate that although the phase-space distribution function approaches a fractal in the long time limit, this is not directly related to the apparent inaccuracy of the Kawasaki expressions for phase-space averages.

We have shown in Sec. II that the Kawasaki normalization factor, $Z(t)$, is unity for all times $t < t_{1/2}$ (i.e., while they are microscopically reversible). However, examination of molecular-dynamics simulation results would appear to contradict this result. Figure 5 gives the value of $Z(t)$ obtained using a molecular-dynamics simulation as described above with $F_e = 1.0$. Evidently, $Z(t)$ is unity until $t \approx 6$, which is similar to the time for which the Kawasaki response was observed to be accurate for $F_e = 1.0$ in molecular-dynamics simulations. For this simulation $t_{1/2} \sim 8.0$. This behavior is still observed when the time step is halved, indicating that the effect is not due to the microscopic reversibility of the simulation.

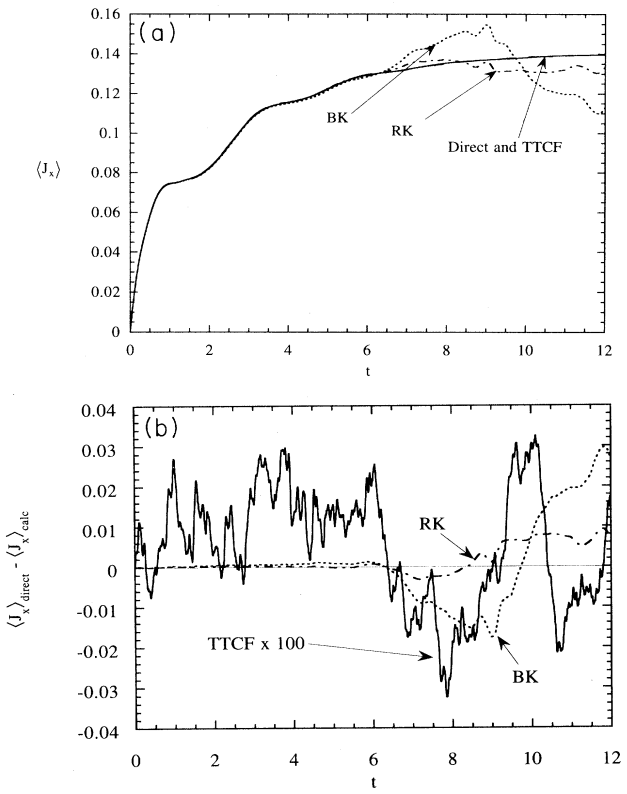

FIG. 3. (a) The color current density response to a constant field of 1.0 determined using nonequilibrium molecular-dynamics simulation. The response calculated using a direct ensemble average (— — —) is compared with that calculated using the TTCF (——), bare Kawasaki (BK) (- - - - - -) and renormalized Kawasaki (RK) (—·—·—) expressions. The color field was applied to a microcanonical equilibrium ensemble of 2×10^6 initial configurations. The system consisted of two WCA disks in a periodic box at $T = 1.0$ and $\rho = 0.396\,850$. (b) Deviation from the direct ensemble average of the color current density response calculated using the TTCF, bare Kawasaki and renormalized Kawasaki expressions for the system examined in (a). The deviation of the TTCF results is magnified by 100 for clarity.

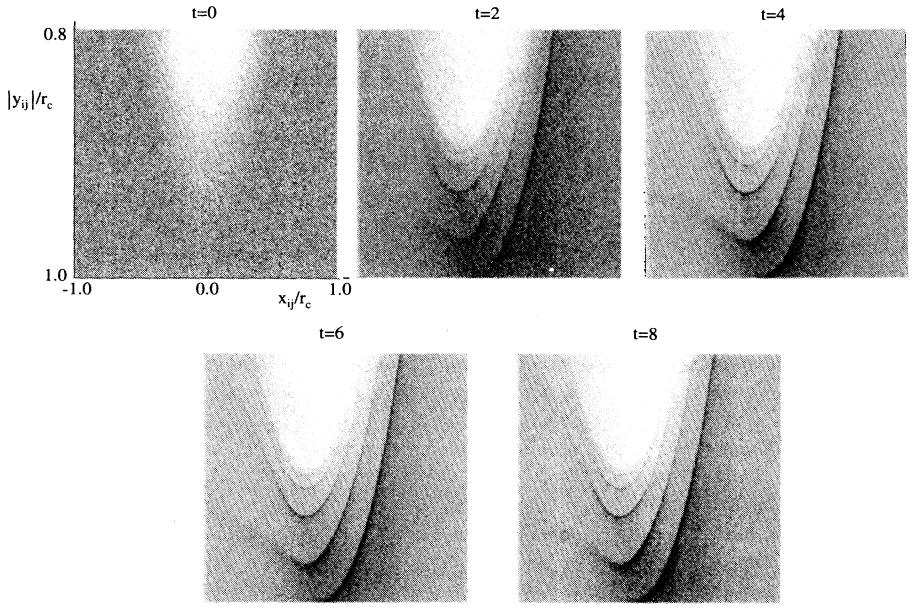

FIG. 4. The time evolution of the coordinate space distribution function for a system to which a constant color field of 1.0 is applied at $t=0$. The data was obtained from a nonequilibrium molecular-dynamics simulation of two WCA disks in a periodic box at $T=1.0$ and $\rho=0.396\,850$.

As predicted in the discussion of Sec. II, rarely observed antisegments have an exponentially enhanced effect on the value of $Z(t)$, hence the departure from unity is likely to be due to the improper sampling of statistically rare events in the system.

The effect of incomplete sampling of phase space (as obtained in molecular-dynamics simulations) can be demonstrated using Eq. (30) and Monte Carlo simulations. Random τ-averaged currents $\langle J \rangle_\tau$ were sampled from a Gaussian distribution with $\langle J \rangle_0 = 0.141$ (which corresponds to that obtained in the molecular-dynamics simulation) and variance given by Eq. (29) (with the parameters set to those used in the molecular-dynamics simulations). Equation (30) was then used to determine the value of $Z(t)$ for $0 < t < 12$, using sets of 2×10^8, 2×10^6, 2×10^4, and 2×10^2 samples. The results are shown in Fig. 5 for comparison with the molecular-dynamics results obtained using 2×10^6 initial configurations. As the number of samples increases (which corresponds to a more thorough sampling of phase space), $Z(t)$ approaches unity for progressively longer times. That the absence of rare *antisegments* is usually responsible for the apparent departure of $Z(t)$ from unity is evidenced by the *nonuniform* approach of $Z(t)$ to unity with increasing numbers of samples. At sufficiently long times $Z(t)$ almost invariably approaches unity from below. (At times longer than those shown in the figure the 2×10^8) sample Monte Carlo run also decays towards zero.) Therefore, the reason molecular-dynamics simulations have shown a departure from unity of $Z(t)$ at long times is due to insufficient sampling of phase space and in particular of rare antisegments.

The explicit normalization of the Kawasaki distribution function improves the agreement of the RK results because it introduces a "compensating error" to the denominator of Eq. (8) which somewhat cancels with the error in the numerator as it is subject to the same inadequate sampling of phase space. It thus provides a form of variance reduction.

To confirm the validity of Eq. (29) in the linear limit ($F_e \to 0$), Fig. 6 shows the reciprocal of the normalized variance of the current distribution, $2\bar{J}_{F_e}/\beta t\sigma^2$, as a function of F_e for $t=1.0$, 2.5, 5.0, and 10.0. A straight line of unit slope is approached in the small field limit, indicating Eq. (29) is valid. Given that Eq. (29) is valid in the $t \to \infty$ and $F_e \to 0$ regime, it then follows that the Kawasaki normalization must be unity in the same regime. This again demonstrates that at least in the linear response regime, microscopic reversibility is not a necessary condition for the Kawasaki normalization to be unity.

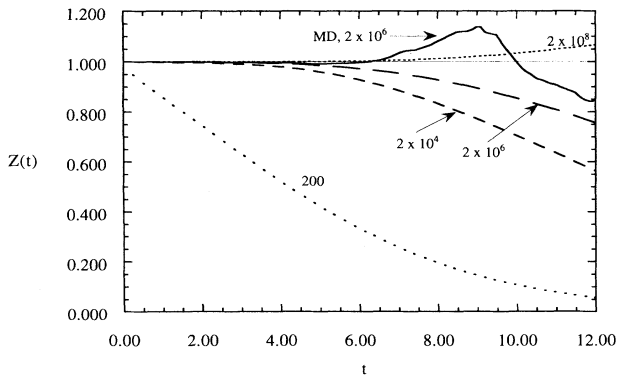

FIG. 5. The Kawasaki normalization factor determined using a nonequilibrium molecular-dynamics simulation with 2×10^6 initial configurations (——) and Monte Carlo simulations with 2×10^2 (- - - - - - -), 2×10^4 (— — —), 2×10^6 (—— ——), and 2×10^8 (- - - - - - - - - -) configurations. The system consisted of two WCA disks in a periodic box at $T=1.0$ and $\rho=0.396\,850$ subject to a constant color field of 1.0. In the molecular-dynamics simulation, the field is applied at $t=0$.

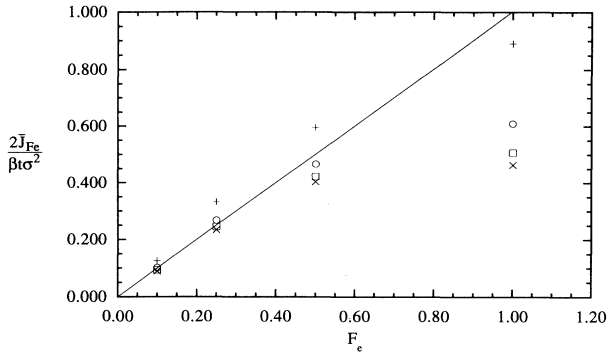

FIG. 6. $(2\bar{J}_{F_e}/\beta t\sigma^2)^*$ as a function of F_e determined using a nonequilibrium molecular-dynamics simulation of an eight-particle systems subject to a constant color field. The results for time segments of $t=1.0(+),2.5$ (○)5.0, (□), and 10.0 (×) are shown. The straight line of unit slope shows the results expected when Eq. (29) is valid. In the simulation $T=1.0$ and $\rho=0.396\,850$.

V. A CONNECTION BETWEEN DYNAMICAL SYSTEMS THEORY AND NONLINEAR RESPONSE THEORY

Evans, Cohen, and Morris (ECM) [12] recently derived an expression for the probability ratio of observing phase-space trajectory segments that satisfy the second law of thermodynamics and their time-reversed antisegments that violate the second law. This ratio formula was based on a natural invariant measure (which we shall call the Lyapunov instability measure), which was originally proposed by Eckmann and Procaccia [13] for the steady-state attractor. We now show that the Lagrangian form of the Kawasaki response distribution (18) can be used to deduce the same expression for this ratio.

In the steady state ECM predicted that the probability, μ_i, of observing a steady-state trajectory segment, i, should, for long segments, be proportional to exponential of the negative sum of segment i's local Lyapunov exponents, λ_{n_i}, multiplied by the observation time τ,

$$\lim_{\tau\to\infty}\mu_i=\frac{\exp\left[-\sum_{n|\lambda_{n_i}>0}\lambda_{n_i}\tau\right]}{\sum_j\exp\left[-\sum_{n|\lambda_{n_j}>0}\lambda_{n_j}\tau\right]}\ . \tag{36}$$

We call this the τ-segment probability formula. The observation time τ must be shorter than the lifetime of the antisteady state, that is $\tau<t_{1/2}$ (i.e., trajectories must be accurate for a time τ), however, the equation is supposed to be valid arbitrarily long *after* the system was at equilibrium.

Consider a microcanonical ensemble of initial, $t=0$, phases, which is used to generate an ensemble of subsequent nonequilibrium trajectory segments. The ratio of probabilities of observing a segment of duration 2τ $(t-\tau,t+\tau)$, namely, segment i, and its conjugate antisegment, i^*, is

$$\begin{aligned}\lim_{\tau\to\infty}\frac{\mu_{i^*}}{\mu_i}&=\frac{\exp\left[-\sum_{n|\lambda_{i^*n}>0}\lambda_{i^*n}2\tau\right]}{\exp\left[-\sum_{m|\lambda_{i_m}>0}\lambda_{i_m}2\tau\right]}\\&=\frac{\exp\left[\sum_{n|\lambda_{i_n}<0}\lambda_{i_n}2\tau\right]}{\exp\left[-\sum_{m|\lambda_{i_m}>0}\lambda_{i_m}2\tau\right]}\\&=\exp\left[\sum_n\lambda_{i_n}2\tau\right]=\exp[-3N\langle\alpha\rangle_{2\tau,i}2\tau]\ ,\end{aligned} \tag{37}$$

where $\sum_n\lambda_{i_n}$ is the sum of all Lyapunov exponents for segment i and $\langle\alpha\rangle_{2\tau,i}$ is the time average value of the thermostat multiplier over the segment i. This result was first accurately tested against simulation results in ECM [12].

As τ increases to the value of t, the starting time for the segments approaches zero. We have shown [14] that when $\tau=t$, and the segments begin from microcanonically distributed equilibrium phases, the probability ratio given above is exact for all $\tau(=t)$ not just in the long $\tau(=t)$ limit.

Following the arguments given in [14] for shear flow, if we select an initial, $t=0$, phase, $\Gamma_{(1)}$, and advance time from 0 to τ using the equations of motion (1) we obtain $\Gamma_{(2)}=\Gamma(\tau;\Gamma_{(1)})=\exp[iL(\Gamma_{(1)},F_e)\tau]\Gamma_{(1)}$. Continuing on to 2τ gives $\Gamma_{(3)}=\exp[iL(\Gamma_{(2)},F_e)\tau]\Gamma_{(2)}=\exp[iL(\Gamma_{(1)},F_e)2\tau]\Gamma_{(1)}$ (see Fig. 1).

At the *midpoint* of the trajectory segment $\Gamma_{(1,3)}$ (i.e., at $t=\tau$) we apply the time-reversal map to $\Gamma_{(2)}$ generating $M^{(T)}\Gamma_{(2)}\equiv\Gamma_{(5)}$. (Note: we denote the trajectory τ segment $\Gamma_{(i)}\to\Gamma_{(j)}$, segment $\Gamma_{(i,j)}$.) If we now reverse time, keeping F_e fixed, we obtain $\Gamma_{(4)}=\exp[-iL(\Gamma_{(5)},F_e)\tau]\Gamma_{(5)}$. $\Gamma_{(4)}$ is the initial $t=0$ phase from which a segment $\Gamma_{(4,6)}$ can be generated with $\Gamma_{(6)}=\exp[iL(\Gamma_{(4)},F_e)2\tau]\Gamma_{(4)}$. (See Fig. 1 for details). By construction, the segments $\Gamma_{(1,3)}$ and $\Gamma_{(6,4)}$ are conjugate.

We now discuss the ratio of probabilities of finding the initial phases $\Gamma_{(1)},\Gamma_{(4)}$ which generate these conjugate segments. The probabilities of observing the segments $\Gamma_{(1,3)},\Gamma_{(4,6)}$ are of course proportional to the probabilities of observing the initial phases that generate those segments. It is convenient to consider a small phase-space volume, $V(\Gamma_{(i)}(0))$ about an initial phase, $\Gamma_{(i)}(0)$. As time evolves the number of ensemble members inside $V(\Gamma_{(i)}(t))$ is fixed.

Because the segment $\Gamma_{(4,6)}$ is related to $\Gamma_{(1,3)}$ by a time-reversal mapping that is applied at $t=\tau$, and the Jacobian of that mapping is unity, $V_2=V_5$, $V_3=V_4$, and $V_1(0)=V_6$. However, since $V_1(0)$ and V_4 are volumes at $t=0$ and since the distribution of initial phases is assumed to be microcanonical, we can compute the ratio of probabilities of observing $t=0$ phases within $V_1(0)$ and V_4. This ratio is just the volume ratio,

$$\mu_{1*}/\mu_1 = V_4(0)/V_1(0) = V_1(2\tau)/V_1(0)$$
$$= f(\Gamma_1(0),0)/f(\Gamma_1(2\tau),2\tau), \quad \forall\tau \ . \tag{38}$$

Combining (37) and (38) we obtain an equation which is identical to (18), except that this proof is valid only in the $\tau \to \infty$ limit. This limit is unnecessary for τ segments that extend back to, and include, the equilibrium phase.

This result shows that there is a deep connection between nonlinear response theory and recent work which has used the mathematical machinery of dynamic systems theory to characterize the natural invariant measure (i.e., the N-particle steady-state distribution function) of nonequilibrium steady states [12,13].

VI. CONCLUSION

For the first time we have provided an accurate confirmation of the validity of the Kawasaki expression for nonlinear response. We have shown that the TTCF and Kawasaki expressions for the nonlinear response in autonomous systems are accurate for systems with as few as two particles. For autonomous systems taking the thermodynamic limit is not required. Although the bare and renormalized Kawasaki expressions are formally exact, it has been demonstrated that they are subject to large (systematic) statistical errors and they are therefore usually not of much direct computational use. This remark must be qualified, however, for systems subject to weak external fields. For example in Figs. 2(a) and 2(b) we see a situation where the response computed using both the Kawasaki forms (7) and (8) are in fact more accurate than direct simulation.

We have shown that previous attempts to verify the Kawasaki expressions have been unsuccessful due to insufficient sampling of phase space, rather than the fractal nature of phase space, as has been previously presumed [10].

We have shown that microscopic reversibility is a *sufficient* condition for the Kawasaki normalization factor to be unity. This theoretical result is supported by the present nonequilibrium molecular-dynamics simulation results. However, these results suggest that microscopic reversibility is not a *necessary* condition either for the Kawasaki normalization to be unity or for the ability of either the renormalized or of the bare Kawasaki response to correctly predict the nonlinear response. This empirical result mirrors that obtained many years ago for the linear response [18]. In the linear regime we have proved that microscopic reversibility is a sufficient but not a necessary condition for the Kawasaki normalization to be unity. A weaker sufficient condition is microscopic reversibility over the convergence time of the Green-Kubo integrand for the dissipative flux.

Finally we have shown that there is a deep connection between the Lagrangian form of the Kawasaki distribution function and a recently proposed natural invariant measure for the nonequilibrium steady state. Far from being incompatible with the fractal nature of the nonequilibrium steady-state distribution, nonlinear response theory is consistent with it and leads to a better understanding of at least one of the invariant measures that have been proposed for nonequilibrium steady states.

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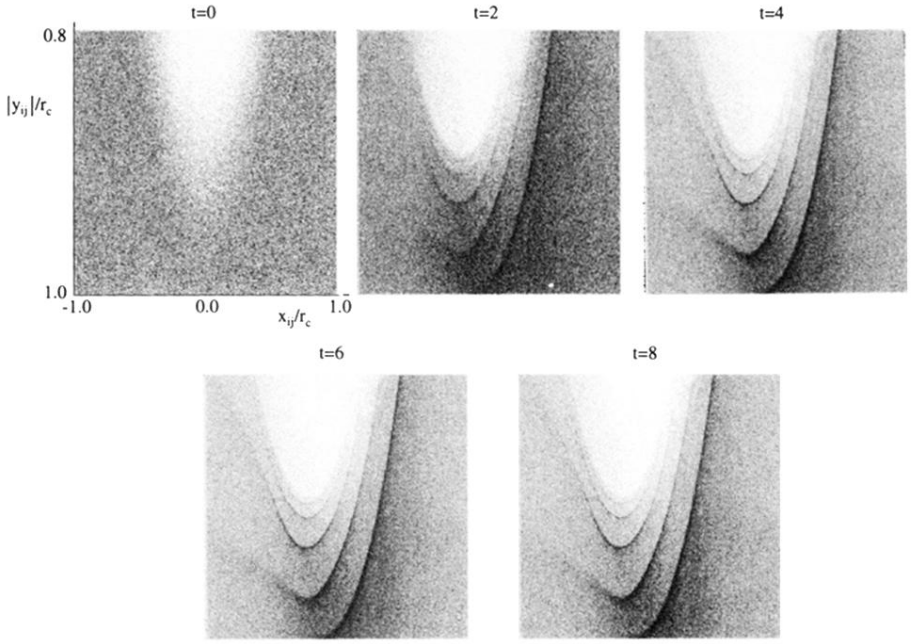

FIG. 4. The time evolution of the coordinate space distribution function for a system to which a constant color field of 1.0 is applied at $t=0$. The data was obtained from a nonequilibrium molecular-dynamics simulation of two WCA disks in a periodic box at $T=1.0$ and $\rho=0.396\,850$.