Numerical study of the steady state fluctuation relations far from equilibrium

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A thermostatted dynamical model with five degrees of freedom is used to test the fluctuation relation of Evans and Searles (Ω-FR) and that of Gallavotti and Cohen (Λ-FR). In the absence of an external driving field, the model generates a time-independent ergodic equilibrium state with two conjugate pairs of Lyapunov exponents. Each conjugate pair sums to zero. The fluctuation relations are tested numerically both near and far from equilibrium. As expected from previous work, near equilibrium the Ω-FR is verified by the simulation data while the Λ-FR is not confirmed by the data. Far from equilibrium where a positive exponent in one of these conjugate pairs becomes negative, we test a conjecture regarding the Λ-FR [Bonetto et al., Physica D 105, 226 (1997); Giuliani et al., J. Stat. Phys. 119, 909 (2005)]. It was conjectured that when the number of nontrivial Lyapunov exponents that are positive becomes less than the number of such negative exponents, then the form of the Λ-FR needs to be corrected. We show that there is no evidence for this conjecture in the empirical data. In fact, when the correction factor differs from unity, the corrected form of Λ-FR is less accurate than the uncorrected Λ-FR. Also as the field increases the uncorrected Λ-FR appears to be satisfied with increasing accuracy. The reason for this observation is likely to be that as the field increases, the argument of the Λ-FR more and more accurately approximates the argument of the Ω-FR. Since the Ω-FR works for arbitrary field strengths, the uncorrected Λ-FR appears to become ever more accurate as the field increases. The final piece of evidence against the conjecture is that when the smallest positive exponent changes sign, the conjecture predicts a discontinuous change in the “correction factor” for Λ-FR. We see no evidence for a discontinuity at this field strength.


INTRODUCTION

Steady state fluctuation relations (SSFRs) describe the statistical fluctuations in time-averaged properties of non-equilibrium steady state dynamical systems. They show how thermodynamic irreversibility emerges from the time-reversible dynamics of the particles, and thus are of fundamental importance. The relationships also make quantitative predictions about these fluctuations, and these have been tested in computer simulations (for example, see Refs. 1–20) and in laboratory experiments.21–24

A number of different classes of fluctuation relations (FRs) have been proposed for deterministic, reversible dynamics. Transient fluctuation relations (TFRs) describe the statistics of time-averaged properties along a set of trajectory segments all initiated from a known distribution function at \( t=0 \). For systems with the time-averaging commencing at \( t=0 \), they can be written as

\[
\frac{1}{t} \ln \frac{\Pr(\tilde{\Omega}_t=A)}{\Pr(\tilde{\Omega}_t=-A)} = A,
\]

where \( \tilde{\Omega}_t = \int_0^t \Omega(s)ds \), and \( \Omega(t) \), the dissipation function, is a generalized entropy production that is uniquely defined for a specified dynamical system \( \Gamma(t) \), and initial distribution of states \( f(\Gamma,0) \).25 A precise definition of the dissipation function is given later; see Eq. (10). The notation \( \Pr(\tilde{\Omega}_t=A)dA \) is used to represent the probability that \( \tilde{\Omega}_t \) takes on a value \( (A - dA/2, A + dA/2) \). These relations have been derived for reversible deterministic systems that satisfy the ergodic consistency condition.25 They are valid at all times, and do not explicitly require the dynamics to be chaotic. Transient fluctuation relations are derived using the time reversal symmetry of the dynamics and hence apply to systems that are arbitrarily far from equilibrium. Of course in such systems

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the probability of negative values for the time-averaged dissipation function becomes rather small, necessitating either short observation times or small system sizes. As written, (1), these TFRs are formally ensemble $f(\Gamma, 0)$ and dynamics $\Gamma(t)$, independent, although the precise expression for the dissipation function will change with different ensembles and dynamics.

Historically the first FRs that were proposed\textsuperscript{26} concerned fluctuations in time-averaged entropy production in nonequilibrium steady states, where trajectory segments were sampled from the single, unique steady state trajectory. The first SSFR was proposed for isoenergetic steady state systems and can be expressed as

$$\lim_{t \to \infty} \frac{1}{t} \ln \frac{\Pr(\hat{\Lambda}_t = A)}{\Pr(\hat{\Lambda}_t = -A)} = A,$$

where $t$ is the averaging time, $\hat{\Lambda}_t = \frac{1}{t} \int_0^t \Lambda(s) ds$, and $\Lambda$ is the phase space contraction rate $\Lambda = (\partial/\partial \Gamma) \cdot \Gamma$.\textsuperscript{25} We will refer to the fluctuation relation (2) by the acronym $\Lambda$-FR. Following the early work of Evans \textit{et al.}\textsuperscript{26} a formal derivation of this SSFR was given by Gallavotti and Cohen\textsuperscript{27-29} under the condition that $A$ is bounded by a value $\Lambda^*: A \in (-\Lambda^*, \Lambda^*)$.\textsuperscript{30} Evans \textit{et al.}\textsuperscript{26} considered only isoenergetic dynamics but the work of Gallavotti and Cohen allows the application of the SSFR to a much wider class of dynamics (e.g., constant temperature dynamics). Evans and Searles\textsuperscript{4,6,25} addressed the issue of SSFRs for steady states that are not maintained at constant energy. They gave an heuristic proof backed up by extensive numerical data that in steady states that are unique (i.e., except for a set of measure zero, steady state thermophysical properties are independent of the initial phase) the dissipation function satisfies the SSFR,

$$\lim_{t \to \infty} \frac{1}{t} \ln \frac{\Pr(\hat{\Omega}_t = A)}{\Pr(\hat{\Omega}_t = -A)} = A.$$  

This expression is derived from the corresponding TFR (1). The definition of the dissipation function [defined later, see Eq. (10)] depends precisely on the initial ensemble and the details of the time-reversible equations of motion, but the form of Eq. (1) does not. In contrast to the Gallavotti-Cohen proof of (2) the proof of (3) requires no bounds on the values of $A$. Equation (3) is expected to be valid for any suitable dynamics (constant energy, constant temperature, and constant pressure). In the particular case of constant energy dynamics (3) is identical to (2) since $\Omega(t) = -\Lambda(t)$ but for other dynamics (such as thermostatted dynamics), $\Omega(t) \neq -\Lambda(t)$ instantaneously therefore (2) and (3) are not equivalent. We will refer to the fluctuation relation (3) by the acronym $\Omega$-FR. For a dissipative system of interest in contact with a thermostat, that satisfies the condition of adiabatic incompressibility of phase space,\textsuperscript{25,31} the dissipation function is given by

$$\Omega(t) = -\beta J(t)/V F_e,$$

(4)

where $J$ is the dissipative flux of the system of interest, $V$ is the volume of the system of interest, $F_e$ is the (constant) dissipative field which is applied to the system of interest, $\beta = 1/(k_B T)$, $k_B$ is Boltzmann’s constant, and $T$ is the temperature of the thermostat which may in general be different from the temperature of the system of interest. Indeed the thermodynamic temperature of the (nonequilibrium) system of interest is usually undefined. Equation (3) is expected to apply for all observable values of $A$. Equation (4) shows that in the linear regime close to equilibrium the average dissipation function is indeed the spontaneous entropy production discussed in linear irreversible thermodynamics.

The derivations of both $\Lambda$-FR (2) and $\Omega$-FR (3) exploit the time reversal symmetry in the equations of motion to obtain the ratio of the likelihood of observing fluctuations of equal magnitude but opposite sign in the time average of the appropriate quantity $\bar{\Lambda}(t)$ or $\bar{\Omega}(t)$, respectively. While the argument of the $\Omega$-FR may be related to entropy production under the precise conditions given above, the physical significance of $\Lambda(t)$ (the argument of $\Lambda$-FR) is not always so clear. For well-behaved synthetic thermostats $\Lambda(t)$ may be identified with the rate at which the synthetic thermostat exchanges heat with the system. There are special circumstances where this can be of experimental relevance.\textsuperscript{32} For a given system of interest the quantity $\Lambda(t)$ depends on how far the synthetically thermostatted particles are separated from the system of interest (see Ref. 33). As the thermostatted particles are moved further and further from the system of interest the convergence time for Eq. (2) to hold grows without limit. By way of contrast, after a certain separation distance, the convergence time for Eq. (3) is independent of that distance. For all systems that come to a steady state, $\lim_{t \to \infty} (\hat{\Omega}_t + \hat{\Lambda}_t) = O(1/t) \rightarrow 0$. These FRs have been tested on various systems, for example.\textsuperscript{1-20,24} Equation (2) has been shown to apply in both the linear and nonlinear regimes to isoenergetic systems, and Eq. (3) has been shown to apply in both the linear and nonlinear regimes to a range of systems including isoenergetic, isokinetic, and Nosé-Hoover thermostatted systems,\textsuperscript{1-10,17,19,20,34} and has recently been verified experimentally.\textsuperscript{24} More recently Searles \textit{et al.} have presented a detailed mathematical proof of (3) for chaotic systems.\textsuperscript{35}

The $\Lambda$-FR, (2), has only been validated numerically for constant energy dynamics. For constant temperature dynamics it has proved impossible to confirm (2) numerically, particularly for weak fields.\textsuperscript{6,19,20} However, because (2) is an asymptotic relation it is always possible that the empirical data have not been considered at sufficiently long times for convergence to occur. The status of (2) for nonisoenergetic dynamics has recently been considered in detail by Evans \textit{et al.}\textsuperscript{36} To further complicate the issue, the formal derivation of (2) (Refs. 28 and 29), puts a limit on the magnitude of the external field. To derive (2) using the approach in Refs. 28 and 29 it is assumed that the dynamics is transitive (i.e., the dynamics has an attractor which covers all of phase space, the attractor may be fractal but any point in phase space is arbitrarily close to a point on the attractor), and in determination of (3) (Refs. 25 and 35) it is assumed that only a single steady state exists. Due to these requirements, it has been proposed that Eq. (2) might break down at large fields,\textsuperscript{1,32} particularly when the transitive property is lost, as
it is when there are unequal numbers of positive and negative exponents. In Ref. 11 Bonetto et al. propose a modified version of (2), with a factor introduced to account for the reduction in dimensionality of the system as the dissipative field increases. This proposal results in a modification of (3),

$$\lim_{t \to \infty} \frac{1}{t} \ln \frac{\Pr(-\Lambda, -\Lambda) = A}{\Pr(\Lambda, \Lambda) = A} = XA,$$

(5)

where $X$ is equal to the ratio of the number of conjugate pairs of exponents where one exponent is positive and one is negative, divided by the number of conjugate pairs of nonzero exponents. We shall refer to (5) as $A$-FRX. The “correction factor” $X$ is only expected to differ from unity at large fields. This in turn means that the $X$ factor cannot help the problems previously noted in confirming (2) for nonisoenergetic dynamics at weak fields.

In Ref. 34 Giuliani et al. propose the analogous modification to (3), namely,

$$\lim_{t \to \infty} \frac{1}{t} \ln \frac{\Pr(\bar{\Omega}, \bar{\Omega} = A)}{\Pr(\bar{\Omega}, \bar{\Omega} = -A)} = XA.$$

(6)

We refer to Eq. (6) as $\Omega$-FRX. We note that Evans and Searles have argued that their relations (1) and (3) are correct as written, without requiring any correction factors.

Here we carry out numerical tests to determine the value of $X$ at large fields, and therefore determine whether or not (3) is valid (in which case $X=1$). Although the original arguments$^{11}$ that $X \neq 1$ are based on the behavior of the phase space contraction $\Lambda$, in Ref. 34 it was proposed for $\Omega$ as well. Furthermore it seems reasonable to carry out this test since Eqs. (2) and (3) become equivalent for isoenergetic systems. In this work we also determine the Lyapunov exponents for the system at each state point to identify if we are in a region where this factor would be expected to significantly alter the FR.$^{11}$

Testing $\Omega$-FRX (6) has been attempted in the past (e.g., Ref. 34). However, it is not easy to find systems where the Lyapunov exponents are “soft” so that they can change sign at fields that are sufficiently weak that negative fluctuations in the dissipation can still be observed. In the present paper we test $A$-FRX (5) and $\Omega$-FRX (6) for thermostatted dynamics. For constant energy dynamics there is ample data showing that (2) and (3) are both valid at low to moderate field strengths when $X=1$.

We test $\Omega$-FR (3) on systems close to equilibrium and also very far from equilibrium where the number of positive and negative exponents is not equal. We consider a dynamical system which is a variation on systems developed by Hoover and co-workers$^{37,38}$ to model thermal conduction. For the equations and system parameters we choose, numerical results indicate it has a single steady state: the system is ergodic and strongly mixing so that the steady state is invariant to the initial configuration. We show that for this system, $\Omega$-FR (3) can be verified, even far from equilibrium. We also test equation $A$-FR (2) for this system, although it is not expected to hold at small fields.$^{19,50,36}$

The dynamics for this system are not symplectic or $\mu$-symplectic when the system is out of equilibrium,$^{39}$ so we do not expect conjugate pairing of Lyapunov exponents; however, we find that it is possible to drive the system so that the numbers of positive and negative exponents are unequal but negative fluctuations in the dissipation can still be observed. The system has five degrees of freedom, and therefore five Lyapunov exponents. One of these always has a value of zero since the dynamics is invariant with respect to time translation.$^{39}$ This system is suitable for directly testing the proposed modification of the FR (Ref. 11) since the value of $X$ proposed requires pairs of expanding and contracting directions close to equilibrium. In our case when close to equilibrium there will be two positive and two negative exponents corresponding to two pairs of expanding/contracting directions in phase space. When the system is driven sufficiently far from equilibrium, one of the positive exponents will become negative, reducing the number of expanding/contracting pairs from 2 to 1 with the factor $X$ being reduced from unity to $X=1/2$. If this system is described by the chaotic hypothesis, and if the postulated modification to Eq. (3) is correct, then this would be clearly evident in a test of the FR.

MODEL

Hoover and Hoover$^{37}$ give a simple oscillator model for the nonequilibrium dynamics of heat flow. The model is chaotic and mixing such that it obeys the standard canonical distribution function, with each degree of freedom being Gaussian distributed, at equilibrium. To drive the system away from equilibrium the system is subject to a temperature gradient as a function of the position $q$ through the Nosé Hoover thermal reservoir. Away from equilibrium the distribution function is fractal and when driven strongly enough the oscillator follows a limit cycle.$^{37}$ We have studied this model and accurately reproduced the results of Hoover and Hoover,$^{37}$ we have also found it to obey $\Omega$-FR (3). The Hoover oscillator has only four degrees of freedom resulting in 3 nonzero Lyapunov exponents. The odd number of nonzero exponents makes it unsuitable for investigating $A$-FRX (5) or $\Omega$-FRX (6) so we will not present these results here or discuss this model further. The model we use, given below, has a dissipation function that is different to the phase space compression factor. While the steady state average of the dissipation function (entropy production) and the steady state average of the phase space compression are equal, their distribution functions, which determine the fluctuations in these quantities, may well be different. We note that in general, although it has been falsely assumed in the past, it is not possible to derive (3) from (2) unless the dynamics is isoenergetic wherein $\Lambda(t) = -\Omega(t), \forall t$. We have chosen thermostatted dynamics where the dissipation function and phase space compression are different, in order to illustrate this elementary, yet unfortunately common error. Hopefully this will help demonstrate the importance of recognizing the difference between the dissipation function and phase space compression factor and resolve the resulting confusion.

The system we consider has three thermostating terms
and four nontrivial Lyapunov exponents allowing $\Lambda$-FRX (5) and $\Omega$-FRX (6) to be tested. The equations of motion are:

\[
\begin{align*}
\dot{q} &= p, \\
p &= -q - \alpha_1 p - \alpha_3 p^3 - \alpha_5 p^5, \\
\dot{\alpha}_1 &= (p^2 - T(q))/\tau_1^2, \\
\dot{\alpha}_3 &= (p^4 - 3p^2T(q))/\tau_3^2, \\
\dot{\alpha}_5 &= (p^6 - 5p^4T(q))/\tau_5^2,
\end{align*}
\]

\[T(q) = 1 + \varepsilon \tanh(q),\]

where $q$ is the oscillator coordinate, $p$ is the momentum, $\alpha_1$, $\alpha_3$, and $\alpha_5$ are the multipliers which control the second, fourth, and sixth moments of the momentum distribution, and $\tau_1$, $\tau_3$, and $\tau_5$ are the thermostat relaxation times. By setting $\varepsilon = 0$ we obtain the equilibrium equations of motion. Setting $0 < \varepsilon < 1$ results in a $q$-dependent temperature and the system is driven into a nonequilibrium steady state. The phase space compression factor, $\Lambda(p, \alpha_1, \alpha_3, \alpha_5)$, is given by

\[\Lambda(p, \alpha_1, \alpha_3, \alpha_5) = \frac{\partial q}{\partial \dot{q}} + \frac{\partial p}{\partial \dot{p}} + \frac{\partial \dot{\alpha}_1}{\partial \dot{p}} + \frac{\partial \dot{\alpha}_3}{\partial \dot{p}} + \frac{\partial \dot{\alpha}_5}{\partial \dot{p}},\]

\[= -\alpha_1 - 3\alpha_3 p^2 - 5\alpha_5 p^4.\]

Defining $H = H_0 + \frac{1}{2}(\tau_1^2 \alpha_1^2 + \tau_3^2 \alpha_3^2 + \tau_5^2 \alpha_5^2)$ (where $H_0$ is the Hamiltonian of the unthermostatted oscillator) we obtain $H = T(q) \Lambda(p, \alpha_1, \alpha_3, \alpha_5)$. At equilibrium we observe that $H = \Lambda(p, \alpha_1, \alpha_3, \alpha_5)$ and use the Liouville equation to obtain the equilibrium distribution function for the system,\(^{31,37}\)

\[f(q,p,\alpha_1,\alpha_3,\alpha_5) = \frac{\tau_1 \tau_3 \tau_5}{(2\pi)^{3/2}} \exp(-H(q,p,\alpha_1,\alpha_3,\alpha_5)).\]

We may now obtain the dissipation function from its definition,\(^{6,25}\)

\[\bar{\Omega}_t = \int_0^t ds \Omega(\Gamma(s)) = \ln\left[\frac{f(\Gamma(0),0)}{f(\Gamma(t),0)}\right] - \int_0^t \Lambda(\Gamma(s))ds,\]

where $\Gamma(t)$ is the point in phase space at time $t$ for the trajectory that was integrated from $\Gamma(0)$ at time 0, $f(\Gamma(t),0)$ is the probability density of observing an ensemble member at the phase $\Gamma(t)$ at time 0, and $f(\Gamma(0),0)$ is the probability density of observing an ensemble member at the phase $\Gamma(0)$ at time 0. For the $\Omega$-FRX (3) the same dissipation function as the transient case is used and thus from (8)–(10) we obtain

\[\Omega(q,p,\alpha_1,\alpha_3,\alpha_5) = (1 - T(q))(\alpha_1 + 3p^2\alpha_3 + 5p^4\alpha_5).\]

This system was chosen because it is of low dimensionality, which means that the number of exponents is small and the relative imbalance in the number of positive and negative exponents is significant, even when there is only one additional negative exponent. The low dimensionality of the system also allows the phase space distribution to be visualized, and the precise determination of the Lyapunov exponents. Furthermore, the work of Hoover and Hoover\(^{37}\) shows that their model (which is similar to ours) can be driven to a region where an imbalance in the number of exponents is obtained, and they have shown how the phase space distribution is altered.\(^{37}\)

The model may be envisaged by viewing the two dimensional projections of the distribution functions (Fig. 1). The equilibrium distribution functions [Figs. 1(a) and 1(b)] are Gaussians consistent with (9). When the oscillator is driven, a fractal structure is expected, which is most easily discerned in Fig. 1(e).

SIMULATIONS AND CALCULATION OF LYAPUNOV SPECTRA

Following Hoover and Hoover\(^{37}\) the equations of motion of the systems were solved using a fourth order Runge-Kutta algorithm. The time constants were set to $\tau_1 = 1$, $\tau_3 = 10$, and $\tau_5 = 100$. A series of nonequilibrium systems was then studied with $\varepsilon = 0.01, 0.1, 0.2, 0.3, 0.4, 0.43, 0.45$. Steady state simulations were performed, and the single long trajectory was divided into a large number of segments to form time averages and then produce histograms of $\bar{\Omega}_t$, and $\bar{\Lambda}_t$ both close to equilibrium $\varepsilon = 0.1$ and far from equilibrium $\varepsilon = 0.43$. These distributions were then used to test Eqs. (2) and (3).

The method used to calculate the Lyapunov spectra closely resembles that described in detail by Dellago et al.\(^{41}\) in their study of hard disk systems, and also used in Ref. 39. To reduce numerical error, this method was modified to ensure that the zero exponent in the direction of the flow is identically zero as expected theoretically,\(^{40}\) i.e., no displacement of the tangent vectors in the direction of the flow were allowed.

RESULTS AND DISCUSSION

The Lyapunov spectra for various values of $\varepsilon$ are presented in Table I. The exponent of the vector in the direction of flow in phase space is always zero and not included, leaving four nontrivial exponents. With $\varepsilon = 0$ we have two conjugate pairs (i.e., pairs that sum to zero) and an exponent that is identically zero. This is characteristic of an equilibrium state where the properties of the system are time reversal invariant.

With $\varepsilon = 0.1$ the negative exponents are slightly larger in magnitude than their corresponding positive exponents and the system now evolves forward in time with an increasing probability of observing positive dissipation. As the time for which the trajectory segment is observed increases, the probability of observing positive dissipation increases as quantified by the fluctuation relation of Eq. (3), the $\Omega$-FR. Under these weakly driven conditions it can be seen that the exponents conjugate pair around a nonzero value in a similar way to what would be expected if the dynamics were $\mu$ symplec-
tic. For $\epsilon=0.43$ we have three negative exponents and a single positive one. The exponents under these strongly driven conditions no longer obey the conjugate pairing rule; this is expected as the system is not symplectic. When the system is driven much harder it approaches stability where it will eventually follow a limit cycle in the steady state. A stable system is characterized by the absence of positive Lyapunov exponents.

FIG. 1. Projections of equilibrium and steady state distributions onto the $qp$ and the $q_\alpha s$ planes. $\epsilon=0$ for (a) and (b), $\epsilon=0.1$ for (c) and (d), and $\epsilon=0.43$ for (e) and (f). Each projection has $5 \times 10^4$ points plotted.
The fluctuation relations of Eqs. (2) and (3) may be partially summed to obtain what we will refer to as the integrated fluctuation relation\(^2\) (IFR)

\[
\lim_{t \to \infty} \left( \frac{1}{t} \ln \frac{p(\bar{X}, t) > 0}{p(\bar{X}, t) < 0} \right) = \frac{1}{t} \ln \langle \exp(\bar{X} t) \rangle_{\bar{X} < 0}
\]

(12)

and

\[
\lim_{t \to \infty} \left( \frac{1}{t} \ln \frac{p(\bar{\Omega}, t) > 0}{p(\bar{\Omega}, t) < 0} \right) = \frac{1}{t} \ln \langle \exp(\bar{\Omega} t) \rangle_{\bar{\Omega} > 0},
\]

(13)

where the notations \(\langle \ldots \rangle_{\bar{X} < 0}\) and \(\langle \ldots \rangle_{\bar{\Omega} > 0}\) are used to denote conditional ensemble averages. We note that in obtaining (12) it is assumed that Eq. (2) is valid for all observable values of \(A\). In case this is not true in all systems,\(^3\) we also test (2) directly.\(^4\) In Fig. 2(a) a direct test of Eqs. (12) and (13) (\(\Lambda\)-IFR and \(\Omega\)-IFR, respectively) is plotted using data from the steady state simulations with \(\varepsilon=0.1\). We observe that Eq. (13) (\(\Omega\)-IFR) converges for \(t \approx 8000\) while Eq. (12) (\(\Lambda\)-IFR) does not converge at the longest time shown in the graph. This is shown in more detail at the longest averaging time in Fig. 2(b) where (2) is tested for \(\langle \bar{X} \rangle / 2 \leq A \leq -\langle \bar{X} \rangle / 2\) (in the notation of Ref. 30: \(-1/2 \leq p \leq 1/2\), where \(p = -A / \langle \bar{X} \rangle\); see Ref. 30). At \(t = 10000\), it can be seen that Eq. (3) (\(\Omega\)-IFR) has largely converged while Eq. (2) (\(\Lambda\)-IFR) has not. A more rapid decay for the large field case of \(\varepsilon=0.43\) may be seen in Fig. 3(a). For this strongly driven system there is one pair of contracting/expanding exponents, and the other pair which are both contracting (see Table I). The postulate of Ref. 11 requires that \(X = 0.5\) under these conditions and the resulting prediction of Eqs. (5) (\(\Lambda\)-FRX) and (6) (\(\Omega\)-FRX) may be seen in Fig. 3(a). Clearly \(\Lambda\)-FRX and \(\Omega\)-FRX are both in disagreement with the data. A more detailed comparison to Eqs. (2) (\(\Lambda\)-FR), (3) (\(\Omega\)-FR), (5) (\(\Lambda\)-FRX), and (6) (\(\Omega\)-FRX) at the longest averaging time (\(t = 10000\)) is shown in Fig. 3(b). Here \(0.156 < A < -0.156\) (or \(-0.156 < p < 0.156\); see Ref. 30). Data at \(t = 600\) are also shown. Clearly good numerical agreement with the predictions of (3) (\(\Omega\)-FR) is observed at \(t = 1000\) and fair agreement at \(t = 600\). Again \(\Lambda\)-FRX and \(\Omega\)-FRX are in disagreement with the data. At this high field strength Eq. (2) (\(\Lambda\)-FR) exhibits a small but systematic disagreement with the data both at \(t = 600\) and 1000.

In Fig. 4 we show a comparison of Eqs. (12) (\(\Lambda\)-IFR) and (13) (\(\Omega\)-IFR) for the very low field strength of \(\varepsilon=0.01\). At times \(t > 1500\), convergence is observed for Eq. (13) (\(\Omega\)-IFR) but in the case of Eq. (12) (\(\Lambda\)-IFR) there is no evidence of convergence on the longest simulation time scale that we have computed. This longest time is approximately 100 times the characteristic microscopic relaxation time (i.e., Maxwell time) for the system.

Some equilibrium and steady state probability distributions for a related but different system are shown in Ref. 37. This suggests that the distributions for weakly driven systems are similar to the equilibrium distributions and span the full phase space, but this is not the case for strongly driven systems. We present similar data for our system. Figures 1(a) and 1(b) represent the equilibrium distributions projected onto the \(q_p\) and \(q_{q_3}\) planes, respectively. These projections for steady state systems with \(\varepsilon=0.1\) and \(\varepsilon=0.43\) are shown in Figs. 1(c)–1(f). At \(\varepsilon=0.43\) the phase space is no longer filled, and the system should not be considered to be transitive, yet as shown above the FR does not change. In Fig. 5, we show two sets of initial phase points. These phase points are the origins of trajectory segments of duration 1000. These origins were generated from a single very long phase space trajectory. All origins which generated a negative values of \(\bar{\Omega}\), averaged over \(t=1000\), are shown on the plot. Among the origins which generated positive values of \(\bar{\Omega}\), a random selection process was used to display a number of positive origins which was equal to the number of negative origins. Figure 5 demonstrates that the attractor is chaotic with nearby points generating very different values of \(\bar{\Omega}\). Despite the dominantly negative Lyapunov spectrum the distribution of positive and negative dissipation points is very similar.

From these results, it can be concluded that there is no sudden change in the applicability of the fluctuation relations when moving from a steady state regime with two positive and two negative exponents to a regime with one positive and three negative exponents.

The derivation of (3) (\(\Omega\)-FR) assumes that the statistics of the time-averaged properties sampled from an initial distribution that has then evolved towards an attractor will match those of segments sampled from the steady state attractor in the infinite time limit. This will be true if only a single steady state can be identified, so we have restricted ourselves to this case here.

### Table I. Lyapunov spectra with the trivial exponent omitted.

<table>
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<th>(\varepsilon)</th>
<th>(\lambda_1)</th>
<th>(\lambda_2)</th>
<th>(\lambda_3)</th>
<th>(\lambda_4)</th>
<th>Error in exponents ((\sim 2\ SE)</th>
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<td>0.000 1</td>
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<td>−0.000 9</td>
<td>−0.008 8</td>
<td>−0.022 2</td>
<td>0.000 1</td>
</tr>
<tr>
<td>0.45</td>
<td>0.001 30</td>
<td>−0.004 00</td>
<td>−0.013 30</td>
<td>−0.023 10</td>
<td>0.000 03</td>
</tr>
</tbody>
</table>
For temperature controlled dynamics Eqs. (2) (A-FR) and (3) (Ω-FR) are different, and Eq. (2) (A-FR) may be used to derive results in contradiction to well established Green-Kubo formula.\textsuperscript{36} The derivation of Eq. (2) (A-FR) leads to the expectation that it should break down when the transitive property is lost but we fail to observe this. There is no known system where Eq. (2) (A-FR) can be numerically observed to converge faster than Eq. (3) (Ω-FR) for steady state temperature controlled dynamics. This suggests that the observed convergence of Eq. (2) (A-FR) at high field strengths is a result of the fluctuations in the phase space compression factor (8) being strongly correlated to the fluctuations in the dissipation function\textsuperscript{36} (11) and is not a result of the temperature controlled dynamics being Anosov like.

CONCLUSIONS

We have performed a number of new tests of both the fluctuation relation of Evans and Searles (Ω-FR) and those of Gallavotti and co-workers (A-FR, Λ-FRX, and Ω-FRX). The system involves a triply thermostatted model of heat flow. The model is unusual in that the Lyapunov exponents are “soft”: we can observe a change in sign of one of the four nontrivial Lyapunov exponents at fields strengths (i.e., temperature gradients) that are still small enough to observe...
fluctuations which were they to continue for a very long time, would be in violation of the second law of thermodynamics. It is these “second law violating” fluctuations that are the subject of the various fluctuation relations.

Near equilibrium the $\Omega$-FR (3) is verified by the simulation data while the $\Lambda$-FR (2) is not confirmed by the data. The data shown for the integrated version of the steady state fluctuation relations are really very stark. For the weak fields studied in Fig. 4 the steady state $\Omega$-FR (3) converges but even at 100 times the Maxwell relation time the $\Lambda$-FR (2) has still failed to converge. In fact, the data in the graph could imply that the $\Lambda$-FR never converges regardless of how large the averaging time.

Far from equilibrium where a positive exponent in one of these conjugate pairs becomes negative, we test a conjecture by Gallavotti and co-workers. They conjectured that where the number of nontrivial Lyapunov exponents that are positive becomes less than the number of such negative exponents, then the form of the $\Lambda$-FR needs to be corrected. We show that there is no evidence for this conjecture in our numerical data. In fact, as the field increases, the uncorrected form of the $\Lambda$-FR appears to become more accurate. The reason for this observation is likely to be that as the field increases, the argument of the $\Lambda$-FR more and more accurately approximates the argument of the $\Omega$-FR. Since the $\Omega$-FR works for arbitrary field strengths, the uncorrected $\Lambda$-FR appears to become even more accurate as the field increases. The final point of evidence against the conjecture is that when the smallest positive exponent changes sign the conjecture predicts a discontinuous change in the “correction factor” for the $\Lambda$-FR. We see no evidence for a discontinuity at this field strength in either the $\Lambda$-FR or in the $\Omega$-FR. We only see a gradual improvement of degree of agreement as the field increases.

We note that recently Tempatarachoke43 has also verified (3) for a system that is far from equilibrium and shown (numerically) that transitivity is unnecessary.

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30 Determination of the value of $\bar{A}$ is usually very difficult; however, statements of the $\Lambda$-FR often specify that $1 \leq p^* \leq \infty$, where $p^* = -A^*/(\bar{A}_t)$, and $\bar{A}_t$ is the mean value of $\bar{A}$ (see Ref. 33), which has a negative value. Furthermore, a new treatment of systems with Nosé-Hoover thermostats suggests that Eq. (2) might only be valid in the range $|p| \leq 1$ for these systems (F. Bonetto, G. Gallavotti, A. Giuliani, and F. Zamponi, e-print cond-mat/0507672). Therefore, if it applies, the $\Lambda$-FR should be valid for at least $|p| \leq 1$, or equivalently $|A| \leq |\bar{A}_t|$.
42 It is also possible to obtain modified versions of (12) where only $A \in (-A^*, A^*)$ is considered (i.e., $\lim_{t \to \infty}(1/t)2006 \leq \bar{A}_t < 0)$ can be expected to be small at large $t$.