DIRECT MEASUREMENT OF EQUILIBRIUM AND NONEQUILIBRIUM LYAPUNOV SPECTRA

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Received 15 January 1987; revised manuscript received 29 April 1987; accepted for publication 1 June 1987
Communicated by A.R. Bishop

Lyapunov spectra are measured for a three-dimensional many-body dense fluid, not only at equilibrium, but also in the presence of an isoenergetic nonequilibrium field generating a pair of equal and opposite currents. The Lyapunov spectra bear a strong resemblance to the Debye spectrum of solid-state physics.

The spectrum of Lyapunov exponents $\{\lambda_i\}$ describes the comoving deformation of a phase-space hypersphere made up of neighboring phase-space trajectories. For a recent comprehensive review ref. [1] should be consulted. The largest Lyapunov exponent λ_1 describes the exponential growth rate of a one-dimensional line joining two neighboring trajectories. Adding the next-largest exponent λ_2 describes the exponential growth rate, $\lambda_1+\lambda_2$, of an area defined by joining three neighboring trajectories. The sum of the first n exponents likewise describes the growth rate of the corresponding n-dimensional volume. The largest of the Lyapunov exponents was characterized numerically by Benettin, Galgani and Strelcyn [2] and the general approach to the Lyapunov spectrum was described by Shimada and Nagashima [3]. Alternative methods, based on the analysis of time series, have recently been used to calculate the three largest Lyapunov exponents [4].

We have developed an approach most closely related to that of ref. [3]. We use Lagrange-multiplier constraints to measure the Lyapunov spectrum. Because the equations governing the growth of infinitesimal phase-space hypervolumes can be linearized, the exponential growth can likewise be prevented by using linear Lagrange multipliers [5,6]. Denoting the unperturbed newtonian motion of the ith phase-space basis vector δ_i (where i runs from 1 to $6N-6$) by the linearized equation of motion

$$\dot{\delta}_i(\text{newtonian}) = D\cdot\delta_i ,$$

where D is the $6N\times 6N$ dynamical matrix, the constrained motion, with the basis vectors forced to remain orthonormal, becomes

$$\dot{\delta}_i = D\cdot\delta_i - \sum \lambda_{i\geqslant j}\delta_j ,$$

$$\lambda_{i\neq j} = \delta_i^{\dagger}\cdot D\cdot\delta_j + \delta_j^{\dagger}\cdot D\cdot\delta_i ,$$

$$\lambda_{ii} = \delta_i^{\dagger}\cdot D\cdot\delta_i .$$

The ith Lyapunov exponent λ_i is given by the time-averaged value of the ith diagonal element of the lower-triangular array $\lambda_{i\geqslant j}$.

For realistic many-body systems nothing is known about the form of the Lyapunov spectrum. In the present work we apply the Lagrange-multiplier numerical method to the study of Lyapunov spectra

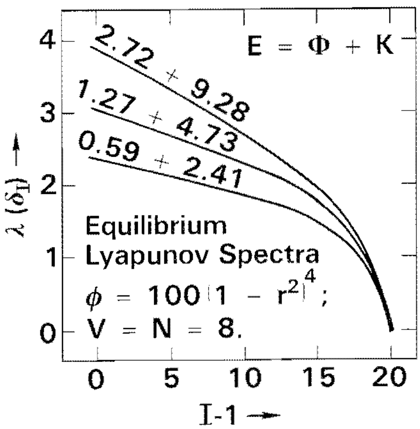

Fig. 1. Equilibrium spectra of Lyapunov exponents for three eight-particle dense-fluid states using the potential function described in the text. Only the positive half of the spectrum is shown. The total energy for each calculation is indicated as a sum of potential and kinetic parts. The runs covered dimensionless times of 100.00, using a fourth-order Runge–Kutta time step of 0.001 or 0.002. The total energy remained constant to 7 or 8 figures throughout the calculations. Exponents in power-law fits to the data shown range from 1/2 at an energy of 12 to 1/3 at an energy of 3.

for both equilibrium and nonequilibrium many-body systems.

Farmer, Ott and Yorke [7] emphasized that the primary difficulties in spectrum characterization are numerical rather than conceptual. The required computer time varies roughly as N^4 for N particles. The Lyapunov spectrum of a three-dimensional N-body system requires that $6N-5$ trajectories be followed in $6N$-dimensional phase space. For hamiltonian systems the symmetry of the spectrum [1] allows a reduction to $3N-2$ trajectories, through which the non-negative half of the exponent spectrum can be determined.

By applying this Lagrange-multiplier approach, as outlined earlier [5], we discovered [6] that the *form* of the spectrum is amazingly simple for many-body systems. It bears a generic resemblance to the solid-state Debye frequency distribution, with the number of exponents lying in a range dI varying as $(21-I)^{\alpha}$, but the exponent α, 1/3 for the Debye spectrum, varies slowly with energy, as shown in fig. 1. The curves shown there fit the 21 largest exponents $(0 \leqslant I-1 \leqslant 20)$ within the 1–2% fluctuations remaining after computer simulations of 10^5 time steps. The data refer to a dense periodic fluid, at roughly half the freezing density. The lowest-energy results are fitted quite well by the Debye distribution. The highest-energy results are described better by a square-root relationship. Thus, at equilibrium, the spectrum of coefficients has much less structure than the vibrational frequency spectrum of a simple solid.

Nosé discovered equations applicable to isothermal and isobaric systems and applied these to equilibrium many-body systems [8,9]. This approach has been extended to a variety of nonequilibrium flow problems involving mass, momentum, and energy currents [10–12]. We began by studying a three-atom chain, with a "hot" atom at one and a "cold" atom at the other, with the temperatures maintained by two independent Nosé thermostats. The phase space for such a three-atom system is eight-dimensional: three space coordinates, three momenta, and two Nosé friction coefficients. Because in the hamiltonian formulation of Nosé mechanics the friction coefficients correspond to momenta, the equations of motion,

$$\dot{q}=p/m, \quad \dot{p}=F(\{q\})-\zeta p, \quad \dot{\zeta}=(K-K_0)/K_0\tau^2 ,$$

are time-reversible. In the reversed motion the momenta p and the friction coefficients ζ change sign. Unfortunately, the three-body energy current varies in an irregular way with the temperature gradient and the initial conditions. Therefore, despite considerable historical interest in such chain problems for longer chains [13], we sought a more physically relevant three-dimensional problem.

An arbitrary limit of 40 CRAY hours per problem limited us to a three-dimensional eight-body problem (12 particles in two dimensions or 24 in one dimension would be nearly as time-consuming). We chose to minimize numerical errors by using a finite-range potential,

$$\phi=\varepsilon[1-(r/\sigma)^2]^4 ,$$

with three vanishing derivatives at the cutoff, $r=\sigma$.

Despite the formal reversibility of the nonequilibrium equations of motion, such systems invariably possess positive transport coefficients, as required by the second law of thermodynamics [14]. We studied *diffusion* by using an external field driving force $\pm F_d$ to accelerate four particles to the right and the remaining four to the left. A gaussian constraint force $F_c = -\zeta p$ was used to keep the internal energy $E = \phi + K$ constant. The results were surprising. The form of the spectrum appeared little changed from the equilibrium Debye-like case.

Unfortunately it is not possible to carry the non-equilibrium simulation out for very long times, so that we cannot rule out small changes in the shape of the spectrum at times very long compared to a lattice vibrational period. Nevertheless the results from runs of over 1000 time steps suggest that the spectrum is insensitive to the presence of nonequilibrium currents with velocities of the same order as thermal velocities.

At the highest of the energies shown in fig. 1 and with a strong field, $F_d = \pm 1$, in units where ε is 100, and the particle mass m and collision diameter σ are both unity, a drift current of order 1 results, corresponding to a dissipative power loss of the same order. This power loss corresponds to the rate at which the field does work, or alternatively, to the rate at which the constraint force F_c extracts heat from the system [12]. On the other hand the Kolmogorov entropy [15], that is the rate at which information is generated by the Lyapunov-unstable dynamics, is about 50 for the states in question. This is just the sum of the positive Lyapunov exponents shown in fig. 1. Thus the "far-from-equilibrium" current provides a perturbation to the equilibrium phase-space motion which is of order 1%. This strongly suggests that perturbation calculations will be useful far from equilibrium, just as they have been for equilibrium fluids and solids [16].

The Kaplan–Yorke conjecture [1,15] relates the strange-attractor fractal dimensionality to the Lyapunov exponents. Our exploratory calculations for the one-dimensional chain indicated a typical attractor dimensionality of about 5 in the eight-dimensional phase space. The three-dimensional fluid calculations suggest also a reduction of no more than a few in the dimensionality of an attractor far from equilibrium.

In response to a helpful referee's comment we add three remarks:

(i) For many-body systems the numerical approach [2,3] is faster than the analytic method developed here. But the analytic equations for the Lyapunov exponents are useful. They show directly that the spectrum changes sign if the motion is reversed.

(ii) The reversibility property just discussed shows that the sum of the Lyapunov exponents is negative on the dynamically-stable (contracting) zero-volume strange attractor which characterizes the steady states obeying the second law of thermodynamics. On the corresponding zero-volume repellor the exponent sum is positive, corresponding to dynamical instability (expansion). Because the sum corresponds to the irreversible entropy production $\dot{S}/k$, we expect that small-field exponents react quadratically, rather than linearly, to the external field. This is in harmony with the small spectral shifts seen "far from equilibrium".

(iii) The reduction in phase-space dimensionality demonstrated here for small steady-state systems is generic and extensive for dissipating many-body systems driven by Nosé or Nosé–Gauss reservoirs. Thus, for large systems, the reduction approaches the bulk contribution, independent of the type of driving reservoirs. Thus the sam loss applies to real systems. If we use a typical collision frequency ν as an estimate for the maximum Lyapunov exponent the nonequilibrium dimensionality loss becomes $\dot{S}/k\nu$. Applied to a simple substance like water the restriction that the loss be somewhat less than the full dimensionality restricts gradients of ln T (where T is the temperature) to be less than about 10^8/cm and the strain rate to be less than about 10^{12} Hz, in agreement with simple physical reasoning.

We thank Bill Moran for computational support, Brad Holian for being helpful, and the National Science Foundation for a generous travel grant. The work carried out at the Livermore National Laboratory was performed under auspices of the Department of Energy through University of California Contract W-7405-Eng-48.

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