Dense-fluid Lyapunov spectra via constrained molecular dynamics

W. G. Hoover,[a] H. A. Posch, and S. Bestiale[b]
Institute for Experimental Physics, University of Vienna, Boltzmanngasse 5, Vienna A-1090, Austria

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A Lagrange-multiplier method for finding the complete spectrum of Lyapunov exponents, which describe the spreading and mixing of many-body phase-space trajectories, is developed and applied here to simple two- and three-dimensional equilibrium fluids with short-range repulsive forces. The numerical values of the Lyapunov exponents converge well, in computer simulations of 10^3 to 10^5 time steps, and are insensitive both to the initial conditions and to the numerical accuracy of the trajectory integration.

I. INTRODUCTION

The microscopic dynamics of systems described by the second law of thermodynamics includes both the Lyapunov spreading of phase-space trajectories and, simultaneously, the condensation of these trajectories onto "strange attractors" with a dimensionality less than that of the full phase space. Now it is possible to study the microscopic time development of such nonequilibrium systems in steady states, where the spreading and condensation are stationary processes. In these nonequilibrium steady-state systems there is a continuous conversion of work into heat. The details have been elaborated for two relatively simple scattering problems: the two-dimensional Galton–Board[1] and the one-dimensional Frenkel–Kontorova conductor.[2] In both cases a single moving particle is accelerated by an external field, scattered by a fixed potential, and simultaneously maintained at constant (time averaged) temperature by thermostatting forces.

Here we address the simpler equilibrium trajectory spreading by analyzing the motion of Newtonian many-body systems. To simplify the analysis we omit nonequilibrium driving and constraint forces[3] as well as long-range forces. In the equilibrium case there is *no* condensation onto a strange attractor because phase-space volume is conserved. This Liouville theorem conservation corresponds to the symmetric distribution of Lyapunov exponents around zero. We follow the usual practice in equilibrium simulations of dense fluids by using periodic boundary conditions. These simplifications with respect to (1) the *form* of the equations of motion, (2) the *range* of the forces, and (3) the *type of boundaries* can all be avoided at the expense of some additional notational and computational complexity.

Consider the neighborhood of a phase-space trajectory where that trajectory is generated by autonomous time-independent deterministic equations of motion. We analyze the co-moving Lagrangian spreading of nearby phase-space trajectories by measuring the Lagrange-multiplier constraint forces required to keep these nearby trajectories in a fixed orthonormal relation to one another as time goes on. We describe the calculation first for the simplest possible case, two two-dimensional particles, and then for the general case, illustrating the general case with results for fluid-density 4-, 8-, and 32-particle systems in three space dimensions.

The same method can be applied to more complicated situations. The results suggest an interesting parallel between the equilibrium spectrum of Lyapunov exponents of *fluids* and the spectrum of lattice-dynamical vibrations for *solids*. Both the dynamics of phase transformations and the mechanism of phase-space mixing in nonequilibrium flows induced by the contraint and driving forces can be investigated in this way. In the next two sections we illustrate the method with several example problems.

II. SIMPLEST CASE: TWO PARTICLES IN TWO DIMENSIONS

Consider two particles interacting with the repulsive pair potential

$$\phi = 100(1 - r^2)^4; \quad r < 1;$$

truncated at the cutoff radius, $r = 1$, at which the first three derivatives vanish. The short-range anharmonicity of this potential was chosen to resemble the rare-gas-model Lennard-Jones potential. One-dimensional nonlinear oscillation calculations using this potential, with energies and mean free paths typical of two- and three-dimensional dense fluids indicated that this truncated, but specially smooth, potential minimizes the errors associated with numerical integration using the classic fourth-order Runge–Kutta method.[4] The sidelength of the periodic box, shown in Fig. 1, was chosen equal to 2 in order to maximize the collision rate. The total energy, a constant of the motion, was also arbitrarily taken equal to 2, the number of particles, corresponding to choosing the product of Boltzmann's constant and the temperature equal to 1.

The dynamics of this two-body system could be most economically described by giving the time history of the three independent variables, x_1, y_1, and θ, with θ giving the direction of relative velocity. Conservation of momentum and energy then provide sufficient information for a complete description. But it is far more convenient to ignore this illusory "simplification" and to use instead the full phase-space description: $(x_1, x_2, y_1, y_2, p_{x1}, p_{x2}, p_{y1}, p_{y2})$. In this full eight-dimensional phase space, Liouville's theorem is valid. But most molecular dynamics simulations, including those described here, use periodic boundaries, so that center-

[a] Permanent address: University of California at Davis-Livermore and the Lawrence Livermore National Laboratory, Livermore, California 94550.
[b] Permanent address: Institute for Advanced Studies, Palermo, Sicily, Italy.

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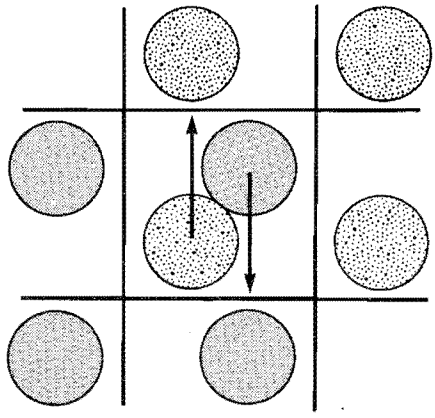

FIG. 1. Initial condition for the two-body problem discussed in the text. Periodic boundaries are used with a box length equal to twice the range of the interparticle forces.

of-mass motion is excluded. Such a motion would undergo no acceleration and is therefore irrelevant. Accordingly, for two particles we consider the phase-space *subspace* in which the center-of-mass velocity is exactly zero. This space is spanned by four orthonormal basis vectors, $\delta_1, \delta_2, \delta_3$, and δ_4. It would be possible to consider instead the energy surface spanned by three such vectors, but Liouville's theorem holds only in a time-averaged sense on that hypersurface. This slightly more complicated energy-surface treatment has no obvious advantages.

Consider now the detailed motion of the basis vectors $\{\delta_i\}$. We follow the natural convention that these are *column* vectors. The transposed *row* vectors are denoted $\{\delta_i^t\}$. Provided that the $\{\delta_i\}$ are infinitesimal in length their motion can be calculated by linearizing the equations of motion about the reference trajectory, as in Ref. 5:

$$\dot{\delta}_i = D\cdot\delta_i .$$

The square matrix D gives the coupling between the coordinate offsets δq and the momentum offsets δp:

$$\delta\dot{q} = \frac{\delta p}{m}; \quad \delta\dot{p} = \left(\frac{\partial F}{\partial q}\right)\delta q .$$

Thus the upper right quadrant of D is I/m, where I is the identity matrix, and the lower left quadrant couples together all of the coordinates of all pairs of interacting particles. This lower left matrix is *exactly the same* matrix of potential-function second derivatives used in classical solid-state lattice dynamics. But the expansion is carried out about the time-varying dynamical state of a *fluid* system rather than about a solid's potential minimum. Thus the eigenvalues are not necessarily positive. The symmetry of the matrix *does* make it possible to show that the eigenvalues occur in pairs. The matrix elements outside the lower left and upper right quadrants all vanish. In nonequilibrium systems, with momentum-dependent forces and friction coefficients[3] there are additional nonvanishing elements.

It has been abundantly demonstrated, and is by now well known, that in chaotic nonlinear systems unconstrained vectors δ will soon rotate into the direction of maximum phase-space spreading and will then diverge to infinite (that is, no longer infinitesimal) length. To prevent this relative motion we constrain[6] the first basis vector to remain at a fixed distance to the reference trajectory, using a Lagrange multiplier λ_{11}:

$$\dot{\delta}_1 = D\cdot\delta_1 - \lambda_{11}\delta_1 .$$

Next, the second basis vector is forced also to lie at a fixed infinitesimal displacement from the reference trajectory and simultaneously to remain orthogonal to δ_1. These two constraints require two more Lagrange multipliers, λ_{22} and λ_{21}:

$$\dot{\delta}_2 = D\cdot\delta_2 - \lambda_{22}\delta_2 - \lambda_{21}\delta_1 .$$

In the general case, the ith basis vector is constrained by i Lagrange multipliers to remain orthonormal with respect to each of its predecessors $j<i$:

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

where the sum is over all $j \leqslant i$.

The initial choice of the orthonormal phase-space basis vectors is largely arbitrary, but they must be chosen to match the center-of-mass velocity, which is conserved by the equations of motion, not just for the reference trajectory r, but also for the trajectory displacements δ. A convenient choice, for two particles in two dimensions is the following (the transposed row vectors are denoted δ_i^t):

$$\delta_1^t = (+\gamma, -\gamma, 0,0,0,0,0,0);$$
$$\delta_2^t = (0,0, +\gamma, -\gamma, 0,0,0,0);$$
$$\delta_3^t = (0,0,0,0, +\gamma, -\gamma, 0,0);$$
$$\delta_4^t = (0,0,0,0,0,0, +\gamma, -\gamma),$$

where $\gamma = 0.5^{1/2} = 0.7071$.

In the general case, the vectors can be chosen as the $N-1$ real one-dimensional sine or cosine normal coordinates of a periodic N-body chain (see Sec. III for a four-body example) giving $6(N-1)$ vectors δ, the components of which are $3(N-1)$ linear combinations of the x, y, or z space coordinates and a further $3(N-1)$ vectors δ, the components of which are $3(N-1)$ linear combinations of the p_x, p_y, or p_z momenta of the particles.

It is convenient to reorthonormalize the vectors occasionally, as is described in Ref. 5. The computation time associated with this step is negligible relative to that involved in calculating the Lagrange multipliers. The values of the Lagrange multipliers can be obtained by calculating the dot products of the equation of motion for each δ_i with the complete set of basis vectors. The constraint maintaining orthonormality can be written

$$\delta_i^t\cdot\dot{\delta}_j + \delta_j^t\cdot\dot{\delta}_i = 0;$$
$$\lambda_{ii} = \delta_i^t\cdot D\cdot\delta_i ;$$
$$\lambda_{i>j} = \delta_i^t\cdot D\cdot\delta_j + \delta_j^t\cdot D\cdot\delta_i .$$

We emphasize that the entire system of differential equations, not just those describing the Newtonian reference trajectory, is *time reversible*. This means that any trajectory, including those described by the orthonormal co-moving basis vectors, can be run backward in time while still satisfying the same equations. In the time-reversed motion, the mo-

mentum variables p and the Lagrange multipliers $\{\lambda_{ij}\}$ change sign. The time-averaged values of the diagonal Lagrange multipliers, the Lyapunov exponents, are thus distributed in pair symmetrically about zero, with positive and negative members of each pair.

An outline of the numerical calculation appears in the Appendix. A typical FORTRAN program, emphasizing clarity at the expense of efficiency, is about 500 to 1000 lines long and requires a few days to write and debug. We document the computation in more detail in the new journal, Molecular Simulation.[2]

The calculations carried out in the present work show that the characterization of the Lyapunov exponents within about 1% requires a time on the order of hundreds of thousands of time steps. In the two-particle, two-dimensional case, with ten Lagrange multipliers λ_{ij}, the Lyapunov exponents are the time-averaged values of λ_{11}, λ_{22}, λ_{33}, and λ_{44}. Table I indicates the range of values observed in three separate calculations with different time steps. Varying the initial conditions confirmed our expectation that the Lyapunov spectrum for this mixing system is independent of these conditions.

The reversibility of the equations of motion was tested for this same two-body problem by running forward in time for 1, 2, 4, 8, 16,... time steps, then reversing the momenta and proceeding backward in time for the same number of steps again. If the numerical algorithm were perfect, the initial condition should be recovered and the Lyapunov exponents for the reversed run should be the negatives of the corresponding exponents in the forward direction. The energy of the system still was constant to eight significant figures when, at a time near 1.0, the Lyapunov exponents showed deviations of order 1%, emphasizing the sensitivity of Lyapunov instability. Thus strict numerical reversibility of the system of equations required for the Lyapunov spectrum holds for a considerably shorter time than the reversibility time for a single many-body system.

III. FOUR THREE-DIMENSIONAL PARTICLES. 18 LYAPUNOV EXPONENTS

The number of Lagrange multipliers grows as the square of the number of particles so that the problem of determining these is one in which fast parallel computers should eventually prove to be useful. The procedure follows the lead of our two-dimensional example. The transposed phase-space basis vectors, three each in the x, y, z, p_x, p_y, and p_z subspace follow the normal vibrational modes for a periodic four-body chain:

$$(+0.50, -0.50, +0.50, -0.50),$$
$$(+0.7071,0, -0.7071,0),$$
$$(+0.50, +0.50, -0.50, -0.50).$$

Four particles, with 24 phase-space coordinates and momenta, together with 18 orthonormal basis vectors $\{\delta_i\}$, correspond to a total of 19×24 ordinary differential equations. In addition to these it is convenient to add the Lagrange multipliers $\{\lambda_{ii}\}$ and the kinetic and potential energy to the list of functions being integrated.

Initial trials, with a total four-body energy of 3.0, showed that with the same time steps used in the two-body problem, $dt = 0.001$, 0.002, and 0.004, the simulation failed after a time of order 1000 steps, with the precise failure time varying roughly as the square root of $1/dt$. The Lagrange multipliers associated with the last, most-restricted basis vector, with the most-negative Lyapunov exponent, diverged (nearly 10^{300} on the CDC 7600). There was no indication of difficulty prior to divergence. The mean values of the positive and negative exponents matched nicely—see Table II. Changing the frequency with which the orthonormalizations was carried out had no effect on the time to failure. The divergence could be delayed, but not eliminated, by leaving out more negative exponents, with the calculation finally becoming stable at long times when only positive Lyapunov exponents were retained. See Table II for details. The early results in the table satisfy the requirement that the coefficients add to zero, in accord with Liouville's theorem.

An eight-body, three-dimensional system was studied next, starting with a face-centered-cubic structure with a kinetic energy of 6.0. The $3N - 3 = 21$ largest Lyapunov exponents are shown in Table III. This calculation took about 20 h on a Cray computer.

The same program, augmented to deal with a three-dimensional 32-body system, requires on the order of 1 min per time step on a Cray computer. The most time consuming part of the calculation involves calculating the matrix products $\delta_i^t \cdot D \cdot \delta_j$. The number of multiplications involved varies as the fourth power of the number of particles. By restricting the set of basis vectors to a subset of the full $6N - 6$, it is possible to obtain a few Lyapunov exponents even for large systems. To demonstrate this, the largest three exponents for 32 particles are tabulated, for a run of 20 000 time steps, in Table IV.

TABLE I. Lyapunov exponents for two two-dimensional particles with total energy $E = 2.00$. The potential part of the energy Φ is 0.223, 0.228, and 0.230 in calculations with a fourth-order Runge–Kutta time step of 0.001, 0.002, and 0.004, respectively. The cumulative time-averaged Lyapunov exponents are given at six different times during each run for the same three time steps: (1) $dt = 0.001$, (2) $dt = 0.002$, (3) $dt = 0.004$. The initial conditions, shown in Fig. 1, were $x_1 = 0.7$, $y_1 = 0.598$, $x_2 = -0.7$, and $y_2 = -0.598$. The initial velocities were $p_{x1} = 0$; $p_{y1} = \sqrt{2}$; $p_{x2} = 0$; $p_{y2} = -\sqrt{2}$.

Time	λ_{11}	λ_{22}	λ_{33}	λ_{44}
10	+(3.6,3.4,3.6)	+(0.5,0.5,0.5)	−(0.2,0.2,0.2)	−(4.0,3.7,4.0)
20	+(3.8,3.5,3.3)	+(0.3,0.3,0.5)	−(0.1,0.1,0.3)	−(4.0,3.7,3.5)
40	+(3.8,3.6,3.5)	+(0.2,0.1,0.2)	−(0.1,0.1,0.2)	−(3.9,3.7,3.7)
100	+(3.7,3.7,3.7)	+(0.1,0.1,0.1)	−(0.0,0.0,0.1)	−(3.7,3.7,3.8)
200	+(3.6,3.7,3.7)	+(0.0,0.0,0.1)	−(0.0,0.0,0.1)	−(3.6,3.7,3.8)
400	+(3.6,3.7,3.7)	+(0.0,0.0,0.0)	−(0.0,0.0,0.1)	−(3.6,3.7,3.8)

IV. THE LYAPUNOV SPECTRUM

These investigations of the Lyapunov spectrum are the first to be carried out for realistic three-dimensional, many-body systems. It seems safe to follow Uhlenbeck in stating that "nothing is known for sure" about this subject. Accordingly it is necessary to begin with intuitive reasoning in an effect to "understand" the results of the computations.

TABLE II. Lyapunov exponents for four three-dimensional particles with total energies $E = 3.0$, 6.0, and 12.0 in a periodic cubic box with sidelength 2. At time 150.0, the average potential and kinetic energies were 0.28 and 2.72 for the lowest-energy case. Preliminary calculations at $E = 3.0$ failed at times of 0.9 for $dt = 0.004$, 1.4 for $dt = 0.002$, and 1.7 for $dt = 0.001$. The symmetry of the coefficients about zero follows from the Hamiltonian nature of the equations of motion. The more negative coefficients, indicated in the table by (*) could not be determined for long times. For times greater than 2 only the positive half of the Lyapunov spectrum was determined. Time steps of 0.004, 0.002, and 0.001 were used for energies of 3, 6, and 12 to determine the long-time data. The detailed time development of the spectrum is described for the lowest-energy case, followed by the final values of the Lyapunov exponents in the higher-energy cases.

Four-body Lyapunov spectra for energy $E = 3.00 = \Phi + K = 0.28 + 2.72$

Time =	1:	(+ 3.2, + 3.1, + 3.0, + 2.3, + 2.3, + 1.9, + 1.4, + 1.0, + 0.3; − 4.3, − 3.0, − 2.9, − 2.3, − 1.8, − 1.4, − 1.3, − 1.0, − 0.6)
	2:	(+ 3.0, + 2.6, + 2.5, + 2.3, + 2.2, + 1.8, + 1.0, + 0.7, − 0.1; − (*), − (*), − (*), − 2.4, − 2.2, − 1.4, − 1.4, − 0.7, − 0.6)
	4:	(+ 2.71,2.64,2.62,2.17,1.83,1.78,1.19,0.66,0.16)
	10:	(+ 2.73,2.64,2.57,2.37,2.17,1.99,1.61,1.30,0.17)
	50:	(+ 2.63,2.56,2.52,2.26,2.09,1.98,1.61,1.35,0.02)
	150:	(+ 2.63,2.48,2.33,2.16,2.01,1.87,1.58,1.32,0.04)

Four-body Lyapunov spectra with energies $E = \Phi + K$ of 6.00 and 12.00

$E = 0.61 + 5.39$; time = 150: (+ 3.44,3.27,3.12,2.92,2.64,2.33,2.12,1.51,0.02)
$E = 1.28 + 10.72$; time = 150: (+ 4.46,4.15,3.94,3.69,3.38,2.94,2.48,1.83,0.00)

To an excellent approximation, the *fluid-phase* Lyapunov exponents, with strongly anharmonic forces, have the three-dimensional Debye distribution familiar from *solid*-state physics. This Debye model for solid vibrations predicts that the number of frequencies $g(\nu)d\nu$ lying between ν and $\nu + d\nu$ is proportional to ν^2. The nth frequency is thus proportional to $n^{1/3}$. This relationship provides an excellent description of the Lyapunov exponents determined here. See Fig. 2. In retrospect, this is consistent with the presence of the lattice dynamical matrix in the *fluid* equations of motion for the Lagrange multipliers. If the second derivative of the pair potential at an energy kT is used to estimate the Debye frequency, the result is very close to the maximum Lyapunov exponent found in the 4-, 8-, and 32-particle data:

$$2\pi\nu_{\max} = (\phi''/m)^{1/2}_{kT}.$$

The success of the Debye model in accounting quite well for the details of Lyapunov instability suggests that the simpler one-particle Einstein model might be useful in estimating an average Lyapunov exponent. Accordingly, we again considered the 32-particle three-dimensional system, but allowing only one of the particles to move, with a total energy of 2.0. See Fig. 3. In this case, six Lyapunov exponents result. Four are statistically indistinguishable from zero. The nonzero pair has a magnitude about one-third that of the maximum shown in Table IV.

TABLE III. Three-dimensional, eight-body Lyapunov spectrum with energy $= 3.00 = \Phi + K = 0.59 + 2.41$ for a time 100.00, discarding the initial 20.00.

{2.36,2.27,2.25,2.23,2.17,2.10,2.04,
1.99,1.94,1.91,1.81,1.78,1.73,1.67,
1.53,1.37,1.32,1.15,0.94,0.54, − 0.01}

Three-dimensional, eight-body Lyapunov spectrum with energy $= 6.00 = \Phi + K = 1.27 + 4.73$.

{ + 3.02,2.92,2.85,2.81,2.72,2.65,2.60,
2.48,2.38,2.32,2.29,2.19,2.05,1.99,
1.89,1.75,1.52,1.33,0.96,0.58, − 0.03}

The data are averages of a Cray and a 7600 calculation. The Cray-1 is three times faster. The two runs were of lengths 87 and 80 and the Lyapunov spectra agreed within about 2%. The initial 20.00 were discarded from both runs.

Three-dimensional, eight-body Lyapunov spectrum with energy $= 12.00 = \Phi + K = 2.72 + 9.28$ for a time 100.00, discarding the initial 10.00.

{3.85,3.80,3.67,3.53,3.33,3.28,3.27
3.07,2.93,2.85,2.73,2.54,2.50,2.23,
2.13,1.97,1.71,1.49,1.01,0.68, − 0.01}

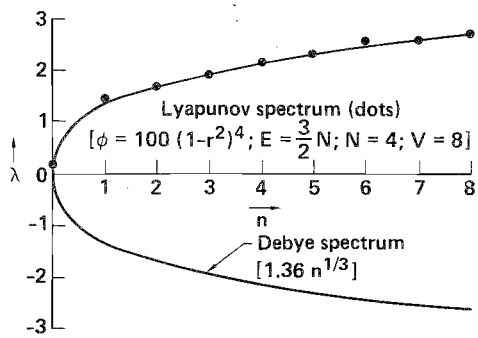

FIG. 2. Sample Lyapunov spectrum ($3N - 3 = 9$ exponents indicated by points) for a four-particle fluid compared with the Debye spectrum (full curve). Data taken from Table II at time 10. The negative exponents match the positive exponents in absolute value for Hamiltonian systems, such as those studied here.

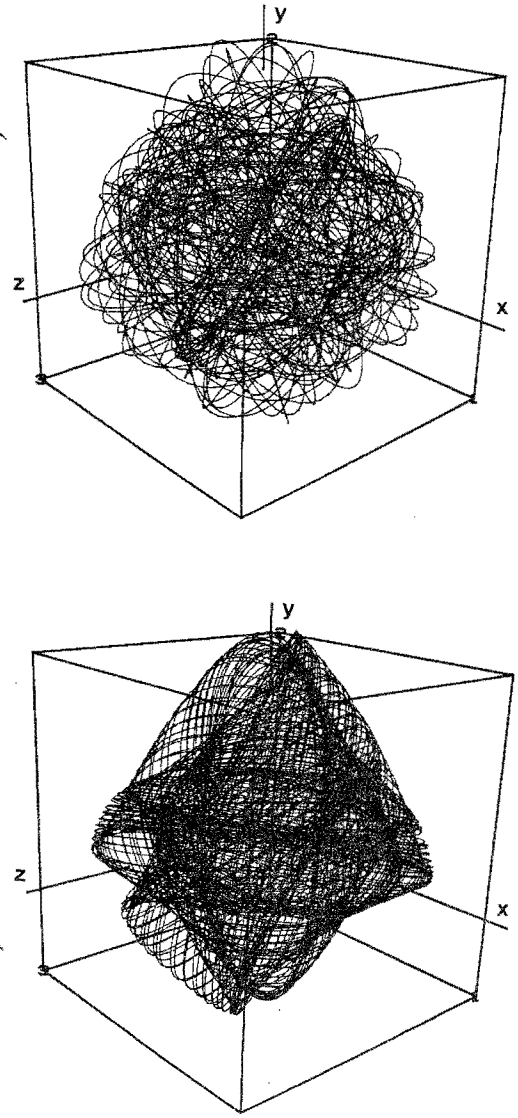

FIG. 3. Two sample one-particle, Einstein-model, coordinate-space trajectories for a single moving particle in a 32-particle face-centered-cubic lattice with nearest-neighbor spacing of 1.00. Both trajectories have the same energy, 3.00, but slightly different initial conditions. The less regular trajectory has two clearly nonzero Lyapunov exponents. The more regular trajectory has all of its exponents statistically indistinguishable from zero.

In *nonequilibrium* systems, even in the presence of Gauss or Nosé constraint and driving forces, the equations of motion still are time reversible. But experience has shown (a microscopic form of the second law of thermodynamics[7]) that the actual motion develops in such a way as to condense the phase-space distribution onto a stable dissipative strange attractor, with Lyapunov exponents whose sum is negative.

At a phase transition the pace of change slows so that a movement of Lyapunov exponents toward zero can be anticipated. It should be possible to see this effect even in the simplest possible system which undergoes a first-order phase transition, two particles with a first-order melting transition.[8]

TABLE IV. The three largest Lyapunov exponents for a 32-particle fluid in three dimensions. Initial configuration was a face-centered-cubic lattice with nearest-neighbor spacing equal to 1.0 and total energy 48.0.

Time = 10: 3.1,3.1,3.0; 20: 3.2,3.2,3.2; 40: 3.3,3.3,3.2.

V. CONCLUSIONS

We have developed a practical scheme for determining Lyapunov spectra for dense atomistic Newtonian systems. The scheme can be applied to the Newtonian equations of motion treated here, or extended to the Gauss–Nosé or Nosé–Hoover equations of motion, either at or away from equilibrium.[7] The method is complementary to that developed earlier by Shimada and Nagashima.[5]

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APPENDIX

American and Austrian versions of the 1000-line FORTRAN codes used to carry out the simulations, written independently, can be obtained from the authors. Here we outline the calculation as a series of six steps. We then append comments pertinent to each step. The steps are as follows:

(1) Choose initial values of coordinates and momenta r, and the offset vectors $\{\delta_i\}$.

(2) Compute the pair potential and its first two derivatives, providing values for r and the dynamical matrix D.

(3) Compute the current Lagrange multipliers $\{\lambda_{ij}\}$ (instantaneous Lyapunov exponents) from the products of the basis vectors $\{\delta_i\}$ with the dynamical matrix D.

(4) Compute the time derivatives of the offset vectors.

(5) Use an integrator, such as the classic fourth-order Runge–Kutta method used here, to advance r and the $\{\delta_i\}$ in time using the derivatives found in steps (3) and (4).

(6) Orthonormalize the offset vectors, if desired, before returning to step (2) for the next time step.

Any configuration of particles can be used initially. For debugging, it is convenient to start out with two particles just beginning a collision. If initially *no* particles are interacting then the total energy is the initial kinetic energy. In choosing the momenta, the *total* momentum should be zero. The insensitivity of the results to the timestep suggests that less smooth potentials would cause no particular difficulties. We have verified this supposition using a truncated Lennard-Jones potential.

In calculating the Lagrange multipliers $\{\lambda_i\}$ a potentially time-consuming sum varying as N^4 occurs. *Pairs* of basis vectors ($3N \times 3N$) are multiplied by the square ($6N \times 6N$) matrix D. But the matrix D is *sparse*. The upper right quadrant, the identity matrix, is treated first. Then only the non-

zero elements of the lower left quadrant (36 for a single interacting pair of particles) need be summed over. Most of the $9N^2$ elements of the dynamical matrix vanish if the forces are short range.

Any convenient intergrating scheme can be used, but if frequent Gram–Schmidt orthogonalization is used, a self-starting scheme, like Runge–Kutta, is best. The orthonormalization (subtracting the dot product of each vector δ_j with the preceding vectors $\{\delta_i\}$ and then rescaling) is discussed in detail in Ref. 5, in which the Lyapunov spectrum is estimated by an alternative, but related, method. For large systems, this Shimada–Nagashima approach is faster. We have used that method to obtain full equilibrium and non-equilibrium Lyapunov spectra for 32 three-dimensional particles (186 exponents).[9]

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