PHYSICAL REVIEW E VOLUME 58, NUMBER 6 DECEMBER 1998

Lyapunov instability of rigid diatomic molecules via diatomic potential molecular dynamics

Oyeon Kum
Agency for Defense Development, P. O. Box 35, Yuseong, Taejon, Korea

Young Han Shin and Eok Kyun Lee
Korea Advanced Institute of Science and Technology 373-1 Gusongdong, Yuseongku, Taejon, Korea

(Received 15 July 1998)

We develop a molecular dynamic method to evaluate the full Lyapunov spectrum for two-dimensonal fluids composed of rigid diatomic molecules. The Lyapunov spectra are obtained for 18 rigid diatomic molecules for various bond lengths $d(10^{-3} \leqslant d \leqslant 1.0)$ in two dimensions with periodic boundary conditions, and interacting with Hoover and Weeks-Chandler-Anderson short-range repulsive forces. The general trends and characteristic features of the Lyapunov spectra are examined for both potentials. Our results are compared with those obtained from I. Borzsák *et al.* [Phys. Rev. E **53**, 3694 (1996)], whose model uses the Lagrange multiplier method. [S1063-651X(98)06612-4]

PACS number(s): 05.45.+b

I. INTRODUCTION

The spectrum of Lyapunov exponents describes the mean exponential rates of divergence and convergence of neighboring trajectories in phase space, and thus provides useful information characterizing the degree of chaos present in a dynamical system. During the past decade, Lyapunov spectra for simple fluids have been investigated thoroughly through numerical simulations and theoretical studies [1–6]. Progress has been made to the point that the second law of thermodynamics and paradoxical macroscopic irreversibility can be explained with these quantities [7,8]. Furthermore, their relation to transport coefficients offers a new basis for describing irreversible processes [4]. They are characterized in terms of a set of exponents $\{\lambda_i\}, i=1, \ldots, 6N$, ordered from the largest to the smallest. The largest exponent λ_1 describes the rate of growth of trajectory separation in phase space. The sum of the largest two exponents, $\lambda_1+\lambda_2$, gives the rate of growth of area defined by three moving trajectories. Finally, the sum of the first n exponents, $\Sigma_{i=1}^{n}\lambda_i$, indicates the rate of expansion or contraction in phase-space objects spanned by n degrees of freedom. The Kolmogorov entropy, $h_k=\Sigma_{\lambda>0}\lambda$, given by the sum of the non-negative exponents [9], describes the rate of divergence of the coarse-grained phase volume. The Lyapunov dimension, following the conjecture of Kaplan and York [8], is a lower bound on the fractal dimension of the associated strange attractor of the chaotic system.

Rigid diatomic molecules are often described by hard dumbbells in shape, interacting with the so-called diatomic potential [10]. Anisotropic fluids reflect the physical transition from uncoupled translational and rotational motions to fully coupled rototranstional states [11]. Compared with the exhaustive studies on the Lyapunov instability for monoatomic molecular dynamics, those for the diatomic molecular model have been rare. Dellago and Posch studied the Lyapunov instability associated with the pure orientational order-disorder transition for the extended XY model both in equilibrium and nonequilibrium steady states [12]. For the hard dumbbells of two-dimensional hard disk fluids, Milanović, Posch, and Hoover [11] studied the physical transition from uncoupled motions between translation and rotation to coupled states. Borzsák, Posch, and Baranyai [13] evaluated the spectra of Lyapunov exponents for rigid diatomic molecules with Weeks-Chandler-Anderson repulsive potential. They performed simulations for rigid homonuclear diatomic molecules with two interaction sites separated by a distance d along the molecular axis, and interacting with the repulsive Weeks-Chandler-Anderson potential. Their molecular dynamics simulation model for diatomic molecules was a straightforward extension of the monoatomic simulations augmented with constraint forces keeping the bond length d for each molecule fixed. However, due to unavoidable computer-precision errors, each bond length d deviates from the ‘‘fixed’’ value as the numerical integration progresses. The propagation of a computational error could be reduced by making the time step Δt sufficiently small. But this procedure makes the computation inefficient. Although their model was able to explain many interesting properties of diatomic molecular systems, the development of a more general model for diatomic systems is required to study the systems with both repulsive and attractive potentials and to extend the systems of nonequilibrium diatomic fluids.

In the present study, we calculate the Lyapunov spectra for Hoover and Weeks-Chandler-Anderson repulsive diatomic potentials. As suggested in the term ‘‘rigid diatomic,’’ the position, rotational angle between the separation vector of molecular axis with an arbitrary fixed direction of the plane, and their conjugate momenta $\{R,\Theta,P,P_\Theta\}$ provide the full representation of the states of the system. We use the Lagrangian formalism to derive the classical motion equations; $L=K-\Phi$, where K is the kinetic energy and Φ the potential energy. For a two-dimensional rigid diatomic system, the kinetic energy K depends on each molecule's center-of-mass velocity and its angular velocity. The total intermolecular potential energy Φ is taken to be pairwise additive of interactions between the sites on different molecules separated by a distance r. In Sec. II we describe our diatomic potential model and derive the motion equations

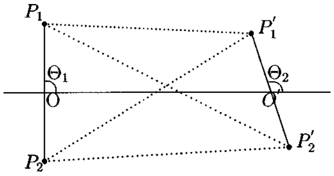

FIG. 1. Diatomic potential model used to simulate homonuclear diatomic fluids. The four pair interactions between neighboring molecules are indicated by dotted lines. For the first molecule, O represents the position of the center of mass, P_1 and P_2 the atomic positions and their interaction centers for the point mass atoms. The same with primes, for the second molecule. The molecular orientation angles are defined by Θ_1 and Θ_2, respectively, measured relative to the vector connecting the centers of mass with some arbitrary direction.

and their linearized motions for the evaluation of the Lyapunov exponents. Our results are presented and discussed in Sec. III. The conclusion follows in Sec. IV.

II. DESCRIPTION OF THE MODEL

Simulations were performed for two-dimensional classical systems consisting of N rigid homonuclear diatomic molecules in a volume V and with periodic boundary conditions. Each molecule consists of two homonuclear atoms, each mass m, separated by a rigid distance d along the molecular axis. The interaction between any given pair of molecules i and j is characterized by the diatomic potential ψ_{ij} [10]:

$$\psi_{ij}=\sum_{n=1}^{4}\phi(r_n), \tag{1}$$

where $\phi(r)$ is a pairwise potential. The r_n are the distances between the center-of-force associated with the nonbonded atoms of any two molecules. Here we consider the following short-range purely repulsive soft potentials ϕ: (a) the Weeks-Chandler-Anderson potential to resemble the hard sphere model but no second derivative at the cutoff distance [2],

$$\phi(r)=\begin{cases}4\epsilon\left[\left(\dfrac{\sigma}{r}\right)^{12}-\left(\dfrac{\sigma}{r}\right)^{6}\right]+\epsilon, & r<2^{1/6}\sigma\\ 0, & r\geq 2^{1/6}\sigma,\end{cases} \tag{2}$$

where $r_{min}=2^{1/6}\sigma$ is the location of the minimum of the Lenard-Jones potential; (b) the Hoover potential with the cutoff radius, $r=\sigma$, at which the first three derivatives vanish [3],

$$\phi(r)=\begin{cases}100\epsilon\left[1-\left(\dfrac{r}{\sigma}\right)^{2}\right]^{4}, & r<\sigma\\ 0, & r\geq\sigma.\end{cases} \tag{3}$$

Without loss of generality, reduced units are used in this paper for simple expression, for which ϵ, σ, and the atomic mass m (molecular mass $2m$) are unity.

Figure 1 shows a schematic representation of the diatomic potential model in two dimensions. $\mathbf{R}_i$ and Θ_i denote the position vector of the ith molecular center of mass and angular coordinate, respectively. Then, the positions of the two atoms belonging to the ith molecule are

$$r_{i,k}=\mathbf{R}_i-(-1)^k\mathbf{IS}, \tag{4}$$

where $i=1,2,\ldots,N$ and $k=1,2$. $\mathbf{r}$ is the Cartesian coordinates $(x,y)^t$ of each site and $\mathbf{R}$ is the corresponding molecule's center of mass vector $(X,Y)^t$. $\mathbf{S}$ is a column vector written as $[\cos(\Theta_i),\sin(\Theta_i)]^t$. Superscript t means the matrix transposition. $\mathbf{I}$ is the $(d/2)$ times 2×2 identity matrix. Then, the bond length d for each molecule, $d^2=(\mathbf{r}_{i,1}-\mathbf{r}_{i,2})^2$, is naturally fixed without any constraint.

The kinetic energy of the two-dimensional system is the sum of a translational part and a rotational part, and is written as

$$K=\sum_{i=1}^{N}[\dot R_i^2+(d/2)^2\dot\Theta_i^2]. \tag{5}$$

The potential energy is

$$\Phi=\sum_{i=1}^{N}\sum_{j\neq i}\psi_{ij}. \tag{6}$$

We denote by F_i and N_i the total force and the total torque, respectively, acting on the molecule i due to all the other interacting molecules. The equations of motion are

$$\{\dot R_i=P_i/2;\quad \dot\Theta_i=P_{\Theta_i}/I;\quad \dot P_i=F_i;\quad \dot P_{\Theta_i}=N_i\}, \tag{7}$$

where I is the moment of inertia. We note that, extending our method, a flexible diatomic molecular system between two sites can be described by merely adding a restoring potential in Eq. (1). We also note that nonequilibrium steady states can be obtained by treating temperature and heat reservoirs explicitly in the equations of motion (7).

For the evaluation of the Lyapunov exponents it is useful to represent the state of the system by the $6N$-dimensional state vector $\Gamma=\{X_i,Y_i,\Theta_i,\dot X_i,\dot Y_i,\dot\Theta_i\}, i=1,\ldots,N$. The equation of motion for the state vector $\Gamma(t)$ is conveniently written as an autonomous system of the first-order differential equations,

$$\dot\Gamma(t)=G(\Gamma(t)), \tag{8}$$

where $\Gamma(t)$ refers to a point in phase space. Its solution defines a flow $\Gamma(t)=\Phi_t(\Gamma(0))$ in phase space. We consider two trajectories separated initially by $\delta\Gamma(0)$ having a vector of norm s:

$$\Gamma'(0)=\Gamma(0)+\delta\Gamma(0). \tag{9}$$

From this equation we can define a finite-length tangent vector at $t=0$,

$$\delta(0)=\lim_{s\to 0}\frac{\Gamma'(0)-\Gamma(0)}{s}, \tag{10}$$

associated with an initial perturbation $\Gamma'(0)-\Gamma(0)$ of the reference trajectory in phase space. As time goes on, this perturbation develops into $\Gamma'(t)-\Gamma(t)$ and the associated tangent vector becomes

TABLE I. The smallest positive Lyapunov exponent λ_{51} and three vanishing (nonnegative) Lyapunov exponents ($\lambda_{52}, \lambda_{53}, \lambda_{54}$) for the seven bond lengths, d/σ. Weeks-Chandler-Anderson results are placed on the left sides and Hoover potential results on the right sides in the parenthesis. All quantities are given in reduced units. The thermodynamic information of the corresponding systems appears in Table II and Table III, respectively.

Exponents number	λ_{51}	λ_{52}	λ_{53}	λ_{54}
$d=0.001$	(0.327, 0.208)	(0.014, 0.001)	(0.000, 0.000)	(0.000, 0.000)
$d=0.2$	(0.477, 0.493)	(0.009, 0.002)	(0.000, 0.000)	(0.000, 0.000)
$d=0.5$	(0.252, 0.449)	(0.005, 0.002)	(0.000, 0.000)	(0.000, 0.000)
$d=0.6$	(0.180, 0.382)	(0.012, 0.001)	(0.000, 0.000)	(0.000, 0.000)
$d=0.7$	(0.113, 0.312)	(0.004, 0.001)	(0.000, 0.000)	(0.008, 0.000)
$d=0.8$	(0.078, 0.243)	(0.013, 0.002)	(0.000, 0.000)	(0.000, 0.000)
$d=1.0$	(0.040, 0.170)	(0.000, 0.000)	(0.000, 0.000)	(0.005, 0.000)

$$\delta(t)=\lim_{s\to 0}\frac{\Gamma'(t)-\Gamma(t)}{s}. \tag{11}$$

The stability of the reference trajectory due to the initial infinitesmal perturbation is determined by this change of length of the vector $\delta(t)$ at time *t*. Consequently, $\delta(t)$ may be viewed as a vector comoving and corotating with the phase flow in the immediate neighborhood of the phase point. The equations of motion for $\delta(t)$ are obtained by linearizing the original motion equations (7),

$$\dot{\delta}(t)=M(\Gamma(t))\delta(t)+O([\delta(t)]^2), \tag{12}$$

where $M(\Gamma)=[\partial G(\Gamma)/\partial\Gamma]$ is the stability matrix. It is a local matrix, depending on the phase point Γ. With the time ordering operator *T*, the formal solution $\delta(t)$ can be expressed as

$$\delta(t)=T\exp\left(\int_0^t M(t')dt'\right)\delta(0). \tag{13}$$

Then, the Lyapunov exponent is calculated from

$$\lambda=\lim_{\hat{t}\to\infty}\frac{1}{\hat{t}}\ln[|\delta(t)|/|\delta(0)|]. \tag{14}$$

In actual computation, $\hat{t}$ is the computing time over which the averages are taken. Usually the data for the first one-third of total running time are discarded. The Lyapunov exponents can be ordered $\lambda_1 \geqslant \lambda_2 \geqslant \cdots \geqslant \lambda_{6N}$ and if the system is ergodic, the exponents are independent of the initial phase point $\Gamma(0)$ and the initial phase-space separation $\delta(0)$.

We consider here a system of $N=18$ interacting diatomic molecules moving in a square periodic box. The side length of the periodic square box is 6σ. The initial arrangement of the molecules are 3×6 with the molecular axes parallel to the *x* axis so that the initial rotational angles are all zero. The reduced molecular number density $n^*=N\sigma^2/V$ is 0.5 for both model potentials. Since the molecular shape depends on the anisotropy parameter d/σ, we define the anisotropy-dependent density parameter as $n^d=N\sigma(\sigma+d)/V$, which is, roughly speaking, the ratio of the occupied volume to the total. n^d becomes equal to n^* for isotropic particles. We varied the anisotropy parameter from 10^{-3} to 1.0 and examined the corresponding variation of the Lyapunov spectrum. A fourth-order Runge-Kutta method with a reduced time step $\Delta t=0.001$ was used for the numerical integration. At each time step, the atomic coordinates on a molecule are obtained from the center-of-mass coordinates and the rotational angle according to the coordinate transformation equation (4). They are used to evaluate the potential energy and hence forces. The well-known algorithms for calculation of the Lyapunov exponents include the classical method proposed by Wolf *et al.* [14], and its conceptual refinement developed by Hoover, Posch, and Bestiale [1] and independently by Goldhirsch, Sulem, and Orszag [15]. Though the constrained orthonormal-vector method developed by Hoover *et al.* is conceptually useful for the systems having rotational degrees of freedom, we used here the classical Benettin method, which requires continuous reorthonomalization to avoid the extensive vector-matrix operations. Gram-Schmidt reorthonormalization was carried out after each time step.

III. RESULTS AND DISCUSSION

In order to compare our results with those calculated from the Lagrange multiplier method [13], identical thermodynamic systems, having the same thermodynamic numerical values within a few percent of the numerical fluctuation, were generated independently by us and Borzsák [16]. The agreement between the two approaches is within a few percent error for the maximum Lyapunov exponent λ_1 and the Kolmogorov entropy h_K for large bond lengths. However, they failed to agree for the relatively short bond lengths. The discrepancy seems to be due to the numerical errors in the calculation of the Lagrange multipliers for small bond length due to the limited precision of a computer [16].

The Lagrange multiplier method requires a severe reduction of the integration time step Δt to obtain the same accuracy for small bond lengths. Otherwise, the system drifts as the numerical integration progresses. The time step of the Lagrange multiplier method was reduced to 0.0003 to get four digits of overall accuracy in the total energy for the bond length $d=0.2$. However, our method achieved the same level of accuracy with $\Delta t=0.001$. Accordingly, our method decreased the computer time by a factor of three to calculate the Lyapunov exponents for $d=0.2$. Moreover, our method is easier to program and debug because of its simple structure.

TABLE II. Thermodynamic parameters characterizing the microcanonical 18 body Weeks-Chandler-Anderson potential system studied in Fig. 2. All quantities are given in reduced units. n^d is the anisotropy-dependent density, K is the kinetic energy, Φ is the potential energy, E is the total energy, T is the temperature, and T_α is the temperature of the α component. $\hat{t}$ is the time for which the trajectory was followed after the decay of transients. λ_{max} is the maximum Lyapunov exponent. $h_k = \Sigma_{\lambda>0}\lambda$ is the sum over all positive exponents.

Bond length	$d=0.001$	$d=0.2$	$d=0.5$	$d=0.6$	$d=0.7$	$d=0.8$	$d=1.0$
n^d	0.5005	0.6	0.75	0.8	0.85	0.9	1.0
$\langle K\rangle$[a]	22.87	34.68	33.00	35.47	35.35	33.65	35.56
$\langle\Phi\rangle$	2.69	4.81	10.27	14.19	19.74	25.68	42.22
E	25.56	39.48	43.27	49.67	55.09	59.33	77.78
k_BT	1.27	1.93	1.83	1.97	1.96	1.87	1.98
k_BT_x	0.63	0.63	0.60	0.62	0.64	0.63	0.71
k_BT_y	0.64	0.63	0.60	0.69	0.65	0.61	0.61
k_BT_Ω	0.00	0.66	0.64	0.66	0.67	0.63	0.66
$\hat{t}$	1000	1000	1000	1000	1000	1000	1000
λ_{max}	3.21	4.71	4.18	4.14	4.12	3.84	3.16
h_k	84.12	129.88	128.19	122.88	113.89	96.29	72.31

[a] Center of mass velocity is subtracted.

Table I shows the smallest positive exponent (λ_{51}) and three vanishing (nonnegative) exponents ($\lambda_{52},\lambda_{53},\lambda_{54}$) for the seven bond lengths, d/σ, equal to 0.001, 0.2, 0.5, 0.6, 0.7, 0.8, and 1.0. The Weeks-Chandler-Anderson results are placed on the left side and Hoover potential results on the right side in the parenthesis. All quantities are given in reduced units. The thermodynamic information of the corresponding systems are given in Table II and Table III, respectively. These tables show that the numerical values of the Lyapunov exponents converge well in the simulation. As can be seen from Table I, our method produces stable numerical results even for extremely short bond lengths.

Figure 2 summarizes the positive branches of the full Lyapunov spectra for the Weeks-Chandler-Anderson potential. Due to the Smale pairing symmetry for symplectic systems, the negative branch is obtained by reversing the sign of the positive branch. The index l numbers the exponents. Throughout the simulations we use a microcanonical system that conserves the total energy E. During the first 200 or 250 time units, the velocities are rescaled to keep the kinetic energy per molecule equal to 2, and 2/3 for each translational and rotational degree of freedom. After that, the system is relaxed and the data for the first 500 time units are discarded. The presented data are ones averaged over the next 1000 time units. During the relaxation the equipartition and total kinetic energy are deviated from the targeted value slightly.

In order to show the numerical values together with relevant thermodynamic information for the states considered, the results of the maximum Lyapunov exponents λ_1 and the Kolmogorov entropy h_k are summarized in Table II. As can be seen in Table II the maximum Lyapunov exponent and the Kolmogorov entropy depend on the anisotropy-dependent density parameter n^d. The maximum exponent λ_1 and the Kolmogorov entropy h_k decrease as the density n^d

TABLE III. Thermodynamic parameters characterizing the microcanonical 18 body Hoover potential system studied in Fig. 3. All quantities are given in reduced units. n_d is the density, K is the kinetic energy, Φ is the potential energy, E is the total energy, T is the temperature, and T_α is the temperature of the α component. $\hat{t}$ is the time for which the trajectory was followed after the decay of transients. λ_{max} is the maximum Lyapunov exponent. $h_k = \Sigma_{\lambda>0}\lambda$ is the sum over all positive exponents.

Bond length	$d=0.001$	$d=0.2$	$d=0.5$	$d=0.6$	$d=0.7$	$d=0.8$	$d=1.0$
n^d	0.5005	0.6	0.75	0.8	0.85	0.9	1.0
$\langle K\rangle$[a]	24.57	35.80	36.69	35.48	38.12	33.30	35.65
$\langle\Phi\rangle$	1.94	3.14	6.07	7.22	9.52	10.09	15.38
E	26.51	38.94	42.77	42.70	47.64	43.39	51.03
k_BT	1.37	1.99	2.04	1.97	2.12	1.85	1.98
k_BT_x	0.68	0.65	0.67	0.64	0.69	0.61	0.65
k_BT_y	0.69	0.65	0.66	0.65	0.69	0.61	0.67
k_BT_Ω	0.00	0.69	0.70	0.68	0.74	0.64	0.66
$\hat{t}$	1000	1000	1000	1000	1000	1000	1000
λ_{max}	2.74	3.64	3.54	3.46	3.56	3.40	3.45
h_k	73.16	101.35	114.38	113.66	116.13	110.48	104.83

[a] Center of mass velocity is subtracted.

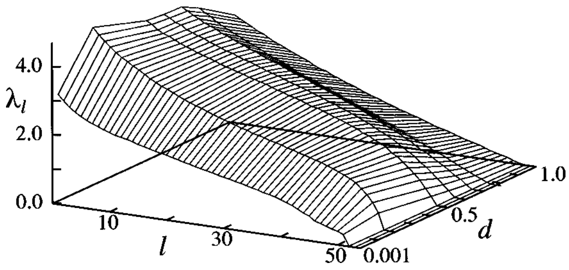

FIG. 2. Anisotropy dependence for the positive branch of the Lyapunov spectrum for the 18 body Weeks-Chandler-Anderson potential system at number density $n^*=0.5$. d is given in the unit of diameter σ, and λ in units of $(m\sigma^2)^{1/2}$. The exponents are defined at integer indexes l only.

increases. However, the Kolmogorov entropy reduces to relatively small value at solid region just as in the case of simple fluid-solid transition [17].

The positive branches of the full Lyapunov spectra for the Hoover potential are shown in Fig. 3. The simulation procedure to calculate these data is the same as that used for the Weeks-Chandler-Anderson potential. The numerical values of the maximum Lyapunov exponents λ_1 and the Kolmogorov entropy h_k are summarized in Table III together with relevant thermodynamic information on the states.

Due to the same repulsive nature of the potential, the overall trends of the numerical values look very similar. For both potentials, the Lyapunov spectra according to bond length show local maxima at $d=0.7$, which corresponds to the anisotropy-dependent density $n^d=0.85$, around the phase transition region for the case of simple fluids [17], but they show different patterns beyond the point. At $d=1.0$, the Hoover potential, which has a relatively shorter interaction range than Weeks-Chandler-Anderson potential, still shows a fluidlike Lyapunov spectrum, whereas Weeks-Chandler-Anderson potential shows typical pattern of solidlike one. An inspection of the orientational correlation function, defined as $C(t)=\langle\cos[\Theta(t)-\Theta(0)]\rangle$, also confirms that the system with Hoover potential is fluidlike at $d=1.0$. This is attributed to relatively larger effective free volume per molecule allowed in the case of Hoover potential at the same density. The interaction range seems to affect also the system potential energy, the maximum Lyapunov exponent, and the Kolmogorov entropy for all bond lengths. It is interesting that the Kolmogorov entropy, a global measure of the rate with which information is generated by the dynamics, stays at relatively high values for the dense region, as shown in Table III. For both potential systems, the number density $n^*=0.5$ is still large enough to keep the rototranslational coupling as shown in the case of smallest bond length $d=0.001$ in both figures.

IV. CONCLUSIONS

We have developed a stable and effective method for the study of Lyapunov instabilities in two-dimensional rigid diatomic molecular systems with soft potentials. We first compared our results with ones calculated by the Lagrange multiplier method. The two results are consistent with each other for large d. But they fail to agree as d decreases, which is due to the numerical instability associated with the computation of Lagrange multiplier, while our method shows reliable results even for the extremely small d. The numerical instability associated with the Lagrangian multiplier method can be reduced by decreasing the time step Δt to be sufficiently small, which may cause the computing time to increase by an order of magnitude larger.

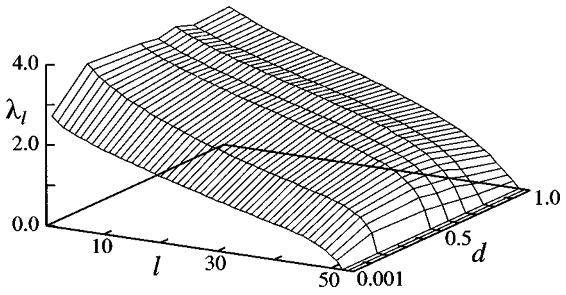

FIG. 3. Anisotropy dependence for the positive branch of the Lyapunov spectrum for the 18 body Hoover potential system at number density $n^*=0.5$. d is given in the unit of diameter σ, and λ in units of $(m\sigma^2)^{1/2}$. The exponents are defined for integer indexes l only.

We find that the system with Weeks-Chandler-Anderson potential approaches, solidlike state as the bond length increase as large as $d=1.0$. On the other hand, that with Hoover potential still remains at fluidlike state for the same bond length. This is due to the difference between the interaction ranges of the two potentials. Hoover potential has shorter interaction range than Weeks-Chandler-Anderson potential, so it allows larger free volume per molecule for the same density.

Physical transition to uncoupled states between translational and rotational motions can not be observed at such a high number density, $n^*=0.5$, even though d is reduced to 0.001. Therefore, molecular dynamic simulation at low density is needed to study the mixing between these qualitively different degrees of freedom and its influence on the full Lyapunov spectrum. Work investigating the Lyapunov exponents and their physical meaning for many different types of soft interacting potentials having attractive part, are now in progress.

ACKNOWLEDGMENTS

We thank Professor Wm. G. Hoover for many stimulating discussions on this and related subjects and for his reading the manuscript with many helpful comments. We are also grateful to Professor H. A. Posch and Dr. I. Borzsák for their helpful provision of the detailed thermodynamic data and useful communication. This work was supported in part by ADD, in part by Com^{2}Mac-KOSEF, and in part by the MOST under the natural R&D Project.

[1] W. G. Hoover, H. A. Posch, and S. Bestiale, J. Chem. Phys. **87**, 6665 (1987).
[2] H. A. Posch and W. G. Hoover, Phys. Rev. A **38**, 473 (1988).
[3] H. A. Posch and W. G. Hoover, Phys. Rev. A **39**, 2175 (1989).
[4] D. J. Evans, E. G. D. Cohen, and G. P. Morriss, Phys. Rev. E **42**, 5990 (1990).
[5] W. G. Hoover, H. A. Posch, and C. G. Hoover, Chaos **2**, 245 (1992).
[6] H. v. Beijeren, J. R. Dorfman, E. G. D. Cohen, H. A. Posch, and C. Dellago, Phys. Rev. Lett. **77**, 1974 (1996).
[7] W. G. Hoover and H. A. Posch, Phys. Rev. E **49**, 1913 (1994).
[8] W. G. Hoover, O. Kum, and H. A. Posch, Phys. Rev. E **53**, 2123 (1996).
[9] G. Benettin, L. Galgani, and J. M. Strelcyn, Phys. Rev. A **14**, 2338 (1976).
[10] H. Sweet and W. A. Steele, J. Chem. Phys. **47**, 3022 (1967).
[11] L. Milanović, H. A. Posch, and W. G. Hoover, Chaos **8**, 455 (1998).
[12] Ch. Dellago and H. A. Posch, Physica A **237**, 95 (1997).
[13] I. Borzsák, H. A. Posch, and A. Baranyai, Phys. Rev. E **53**, 3694 (1996).
[14] A. Wolf, J. B. Swift, H. L. Swinney, and A. Vastano, Physica D **16**, 285 (1985).
[15] I. Goldhirsch, P-L. Sulem, and S. A. Orszag, Physica D **27**, 311 (1987).
[16] Dr. I. Borzsák (private communication).
[17] Ch. Dellago, H. A. Posch, Physica A **230**, 364 (1996).