Local (in time) maximal Lyapunov exponents of fragmenting drops

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We analyze the dynamics of fragment formation in simulations of exploding three-dimensional Lennard-Jones hot drops, using the maximum local (in time) Lyapunov exponent (MLLE). The dependence of this exponent on the excitation energy of the system displays two different behaviors according to the stage of the dynamical evolution: one related to the highly collisional stage of the evolution, at early times, and the other related to the asymptotic state. We show that in the early, highly collisional, stage of the evolution the MLLE is an increasing function of the energy, as in an infinite-size system. On the other hand, at long times, the MLLE displays a maximum, depending mainly on the size of the resulting biggest fragment. We compare the time scale at which the MLLE's reach their asymptotic values with the characteristic time of fragment formation in phase space. Moreover, upon calculation of the maximum Lyapunov exponent (MLE) of the resulting fragments, we show that their dependence with the mass can be traced to bulk effects plus surface corrections. Using this information the asymptotic behavior of the MLLE can be understood and the fluctuations of the MLE of the whole system can be easily calculated. These fluctuations display a sudden increase for that excitation energy which produces a power-law-like asymptotic distribution of fragments.

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I. INTRODUCTION

The process by which a highly excited finite system develops a collective radial flow and disassembles into a set of clusters is called fragmentation.

This kind of phenomenon appears in different areas of physics. Just to mention a few it is found in collisions of highly accelerated clusters with hard surfaces, fullerenes impacted by high velocity projectiles, intermediate-energy nuclear collisions, etc. [1].

In particular in the case of collisions of energetic Ar clusters with hard walls it has been shown [2] that a transition from evaporation to shattering takes place in the range of incident energy which corresponds very closely to the corresponding boiling temperature.

Also, in recent experiments with energetic hydrogen-ion clusters colliding against 60 carbon fullerenes [3] the distribution of the detected largest mass and its variance versus multiplicity, when properly scaled, closely resemble the results obtained in Au-on-Au collisional experiments [4].

For the case of nucleus-nucleus collisions a rather huge amount of both experimental and theoretical work has been performed. The essential features of such a process can be summarized by stating that as the energy of the colliding system is increased the fragment size distribution of the resulting clusters goes from U shaped to exponentially decaying. Somewhere between these two extremes a power law can be detected. This sequence of shapes in the mass distributions is predicted by the Fisher droplet model [5] and could be related to a phase transition. Because neither the nuclear interaction potential nor the corresponding equation of state (EOS) of nuclear matter is known, the possibility of facing a phase transition has triggered a lot of work in this area; in particular the determination of the caloric curve and of the critical exponents has attracted a lot of attention (for recent reviews see [6,7]).

The possibility of a phase transition from a solid-like to a liquid-like state for finite systems, is well founded and can be related to the usual solid-liquid phase transition of infinite systems. This can be understood as follows: at low excitations small systems are self sustained, hence they have enough time to develop correlations of the kinetic type [8,9]. The characterization of the fragmentation process has been rather elusive, instead. In particular in this case we face a system out of equilibrium that develops a collective radial mode and fragments in small clusters. Among recent results in this area, we would like to mention the calculation of an extended caloric curve that describes the behavior of the temperature of the system at the breakup time with the excitation energy [10,11] (hereafter refereed as I), the calculation of global Lyapunov exponents [12], generalized entropies, and fractal dimensions [13].

In this work we analyze three-dimensional (3D) drops of 147 particles interacting via a 6-12 Lennard-Jones (LJ) potential, which are excited enough to undergo fragmentation or evaporation.

A way to characterize the dynamics in the phase space of this process is to calculate the maximum Lyapunov exponent (MLE). The MLE gives an idea of the velocity at which the system explores the available phase space.

In I it has already been shown that in the early stages of the evolution of highly excited drops, due to interparticle collisions, a collective radial motion develops. The emergence of such an ordered motion has important consequences, as, for example, it behaves as a heat sink giving rise to a constant fragmentation temperature. In this way the competition between ordered motion (collective expansion) and chaotic motion (interparticle collisions) plays a major role in the dynamics of fragment formation and thermal

properties of the system. A way to properly characterize such a competition is to study the time evolution of the MLE.

Given two very close initial conditions in phase space, the MLE is defined as

$$\lambda = \lim_{t\to\infty} \lim_{d(0)\to 0} \left[\frac{1}{t} \ln \frac{d(t)}{d(0)} \right],$$

where $d(t)$ is the distance in phase space between their corresponding trajectories at time t and $d(0)=d(t=0)$. The MLE is positive for chaotic systems.

The MLE has been used to study the solidlike to liquidlike phase transition in LJ clusters [14–16]. It also has been used to study the problem of fragmentation, analyzing the occurrence of a possible liquid-gas phase transition [12].

The MLE can be understood as an average, along the different stages of the evolution, of the behavior of the system along an infinite trajectory in phase space. In the case of fragmentation (as the name indicates) the nature of the system varies as it irreversibly evolves from a highly excited, unstable, piece of matter into a set of noninteracting, stable, clusters. As such the average over the infinite trajectory erases the relevant information, i.e., what happens while the system fragments. In order to avoid this feature we calculate the local-in-time Lyapunov exponents at a sequence of temporal intervals of the evolution of the excited drops (see Sec. IV), which can be considered as a short path estimate of the MLE. This magnitude has already been used, for example, in [17] to study the properties of small Ar clusters, in particular the evolution of its ergodic properties with energy.

Our calculation allows us to obtain a characteristic time [the time at which maximum local (in time) Lyapunov exponent (MLLE) attains its asymptotic value τ_{mle}] which can be used to distinguish the two well-differentiated stages in the evolution of the drop: (i) the highly collisional one, at early times, which is characterized by the absence of strong collective effects, and (ii) the asymptotic one, dominated by the collective radial motion.

We compare this result with other relevant time scales obtained from analysis of the dynamic process, like the characteristic times for fragment formation in phase space, the emergence of the radial collective motion (see I), and the one corresponding to the mean momentum transfer (MMT) between particles in the collisional process.

In the asymptotic state the system can be viewed as a mixture of noninteracting free particles and almost stable fragments of varying sizes. As such, it is not necessary to perform a local analysis (calculating the MLLE) and the standard MLE approach averaging over a long trajectory can be used.

Nevertheless, during this stage the almost stable clusters might decay via the evaporation of, mainly, free particles. Such a process will induce further cooling of the primary fragments [18] and this cooling should give rise to more regular configurations and thus reduce the effective MLE.

In this stage we calculate the dependence of the MLE with the mass of the fragments, and using the resulting information, we analyze the MLE of the whole system in terms of the MLE of its fragments. This study shows us that the MLE of the drop is driven by the size of the biggest fragments. In addition, we can calculate the fluctuations of the MLE, and when we plot these fluctuations as a function of the excitation energy, they display a sudden increase at the energy for which a power law asymptotic distribution of fragments is obtained.

This paper is organized as follows. In Sec. II we describe the model used to simulate the fragmentation process. Section III is devoted to the analysis of the properties of the drop: the mass spectra as function of the energy and the caloric curve. We also discuss the times scales relevant for the fragmentation process and the formation of the radial collective mode. In Sec. IV we deal with the definition of the MLE and the different ways to calculate it. In Sec. V we analyze the results obtained for the MLLE and the behavior of the MLE for the asymptotic state. Finally, in Sec. VI conclusions are drawn.

II. COMPUTER EXPERIMENTS

We study the fragmentation of excited Lennard-Jones drops. The two-body interaction potential is taken as the truncated Lennard-Jones (6-12) potential:

$$V(r) = \begin{cases} 4\epsilon \left[\left(\frac{\sigma}{r}\right)^{12} - \left(\frac{\sigma}{r}\right)^{6} - \left(\frac{\sigma}{r_c}\right)^{12} + \left(\frac{\sigma}{r_c}\right)^{6} \right], & r < r_c, \\ 0, & r \geqslant r_c. \end{cases} \tag{1}$$

We took the cutoff radius as $r_c = 3\sigma$. Energy and distance are measured in units of the potential well (ϵ) and the distance at which the potential changes sign (σ), respectively. The unit of time is $t_0 = \sqrt{\sigma^2 m/48\epsilon}$. We used the velocity Verlet algorithm to integrate the classical equations of motions [19] taking $t_{int} = 0.001 t_0$ as the integration time step. As a result, energy was conserved to an accuracy of 1 part per 10^6.

We simulate explosions of $N=147$ particles, closed-shell three-dimensional drops. The initial configurations are constructed by cutting a spherical drop from a thermalized periodic Lennard-Jones system with $N=512$ particles in each periodic cell. The degree of excitation can be easily controlled in this way by varying the density and temperature of the periodic system. The initial state of our drops is macroscopically characterized by their energy and density (taken as that of the periodic system). We studied a broad energy range which encompasses very different behaviors regarding the fragmentation pattern, going from $E=-2.8\epsilon$ to $E=+3.0\epsilon$. The density was taken as $\rho = 0.85\sigma^{-3}$. The temperature of the periodic system used for constructing the initial configurations is in the range ~ 1.4 to $\sim 4.3\epsilon$. It can be seen from the equation of state of the Lennard-Jones system [20] that our initial drops are hot and compressed.

III. PROPERTIES OF THE EXCITED DROPS: MASS SPECTRA, CALORIC CURVE, AND RADIAL COLLECTIVE MODE

As stated in the Introduction, many systems differing greatly in size, interaction potential, etc., exhibit fragmentation. Thus, a good way to characterize the excitation energy is according to the asymptotic mass spectra. If the energy of the system is high, it will break into several small fragments.

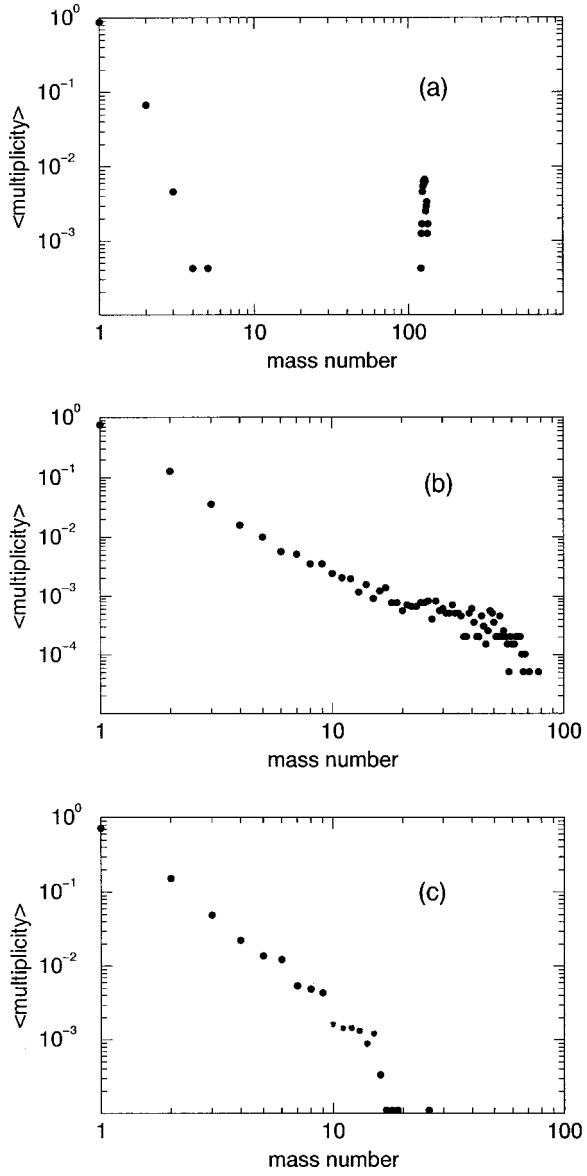

FIG. 1. Asymptotic mass spectra for three different energies in the fragmentation regime: (a) $E=-2.0\epsilon$, (b) $E=+0.5\epsilon$, and (c) $E=+3.0\epsilon$.

The asymptotic mass spectrum will show rapid decay for large masses. On the other hand, for low excitation energies, the systems will evaporate monomers and small clusters while a big drop, comprising most of the mass of the system, will remain bound. In this case, the mass spectrum is U shaped. A third case is usually found: for a given intermediate energy, the mass spectrum will show a power law behavior. This last case is quite important. Taking into account that a power law implies scale invariance and that power law mass spectra are found in second order phase transitions (e.g., percolation at the critical probability [21] or liquid-gas phase transition at the critical point [5]), this kind of spectrum was associated with the system undergoing a second-order phase transition [22,23]. This conjecture, however attractive, has not been confirmed yet.

The sequence of shapes of the mass spectra is the same as the one predicted by Fisher's model of liquid-gas phase transitions. In the latter model, the probability of having a drop of size A in the vapor is given by $P_r(A) = Y_0 A^{-\tau} e^{[-(\mu_l-\mu_g)A+4\pi r_0\sigma(T)A^{2/3}]}$ where μ_l and μ_g are the chemical potential of the liquid and vapor phases and σ is the surface tension.

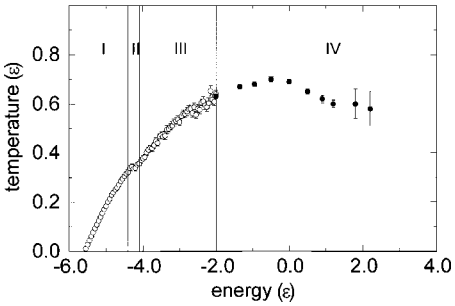

FIG. 2. Extended caloric curve $T(E)$. In regions I, II, and III the system is self-sustained. In region IV the system is in the fragmentation regime. Details of the calculation of the temperature can be found in I.

In Fig. 1 we show the mass spectra resulting from our numerical experiments, for three different energies per particle that display the above-mentioned behavior: $E=-2.0\epsilon$, $E=+0.5\epsilon$, $E=+3.0\epsilon$ (the ground-state energy for this system is $E=-5.8\epsilon$). For $E=-2.0\epsilon$ [Fig. 1(a)] the asymptotic spectrum consists of a big fragment of about 130 particles plus some free particles (U-shaped mass spectra). The mass spectra of $E=+0.5\epsilon$ [Fig. 1(b)] is ''power law like'' and the one corresponding to $E=+3.0\epsilon$ [Fig. 1(c)] is mainly composed of small clusters.

The process by which the drop is disassembled in a set of noninteracting fragments is characterized by the time scales of fragment formation. In [11] the time scales related to the time of fragment formation (τ_{ff}) and the time of fragment emission (τ_{fe}) were calculated. The time of fragment formation is defined as the time at which the fragments are formed in phase space and the time of fragment emission when they are formed in configurational space. The τ_{ff}'s obtained goes from $\sim 20t_0$ for $E=1.8\epsilon_0$ to $\sim 75t_0$ for $E=-0.5\epsilon_0$. The times scales obtained for fragment emission (τ_{fe}) goes from $\sim 40t_0$ for $E=1.8\epsilon_0$ to $\sim 100t_0$ to $E=-0.5\epsilon_0$.

In the Appendix we include a brief description of the fragment recognition algorithms which allow us to calculate τ_{ff}'s and τ_{fe}'s. (See I and references therein for details.)

In order to gain insight into the description of the fragmentation process, we show in Fig. 2 the extended caloric curve calculated in I. The caloric curve is the temperature at fragmentation time (τ_{ff}) as a function of the energy of the system. It displays two well-differentiated regions: the low-energy one, where the systems is self-sustained, and the high-energy region, where the cluster is in the multifragmentation regime. In the low-energy region, the temperature rises with the excitation energy. The drop goes from a solidlike phase to a liquidlike phase and its behavior resembles the one of macroscopic systems, although there are some important differences; see [9]. It is clear that an isolated liquid drop cannot be heated without limits. Once a certain temperature is attained, which depends on the size of the system, additional energy supplied to the system will evaporate particles but will not produces an increase of temperature at τ_{ff}; the associated temperature is called the limit temperature T_{lim}. For energies higher than that of the evaporating drop, the

system undergoes the nonequilibrium process of fragmentation. In the fragmentation region $E > -2.0\epsilon$ the temperature is approximately constant for a wide range of energies, decreasing slowly for the higher ones. Note that the initial drops in our computer experiments are artificially constructed hotter than the limit temperature. In its evolution the system cools down, while the expansion builds up, until it reach its limit temperature.

An important ingredient to understand the dynamical evolution of system is the collective radial mode of expansion. Because initially the drop is hot and compressed, it expands, developing the collective mode mentioned above. This has been fully analyzed in I and we recall that it was found that the radial flux is formed at $\tau_{flux} \simeq 10 t_0$ for the four energies analyzed: $E = 1.8\epsilon$, $E = 0.9\epsilon$, $E = 0.5\epsilon$, and $E = -0.5\epsilon$.

IV. LYAPUNOV EXPONENTS: DEFINITIONS

The maximum Lyapunov exponent, as already mentioned, is a measure of the sensitivity of the system to initial conditions and also gives an idea of the velocity at which the system explores the available phase space. Given two very close initial conditions in phase space, the MLE $\hat{\lambda}$, is given by the following relation:

$$\lambda = \lim_{t\to\infty} \lim_{d(0)\to 0} \left[\frac{1}{t} \ln \frac{d(t)}{d(0)} \right].$$

In order to calculate the MLE, we generate at time $t=0$ a initial configuration of the system (main point) characterized by a given energy. We also generate another initial configuration (the ''son''), which differs from the main point by a small amount d_0 in momentum space (the velocity of the particles are slightly different). Following [12], the distance between the trajectory of the main point and the trajectory of the ''son'' $d(t)$ can be defined as

$$d(t) = \left(\sum_{i=1}^{N} \{ a[\mathbf{r}_m(t) - \mathbf{r}_s(t)]^2 + b[\mathbf{p}_m(t) - \mathbf{p}_s(t)]^2 \}_i \right)^{1/2}, \tag{2}$$

where $\mathbf{r}$ and $\mathbf{p}$ refer to the positions and momenta of N particles at time t. The indices m and s refer to the two trajectories differing by d_0 at $t=0$ (main and son). a,b are two arbitrary parameters which express the fact that the LE's are independent of the particular metrics in phase space. In this work, when we calculate the MLE, we will choose $a=0$, $b=1/m$ with m the mass of the particles; i.e., we take distances in velocity space only. When calculating numerically the time evolution of $d(t)$ by solving the classical equations of motion (CEOM), we find an exponential increase followed by a saturation in v space [12,13]. This saturation takes place for $t \gg \lambda^{-1}$, allowing a reliable calculation of λ. The saturation distance d_∞ brings information about the effectively explored phase space. This way of calculating the maximum Lyapunov exponent will be referred to as the ''maximum global Lyapunov exponent,'' because by following a trajectory, information about the different stages of the evolution will be present in the resulting value of λ (of course λ is to be calculated for $t < \tau_{sat}$). The characteristic time at which saturation is achieved is $\tau_{sat} \sim 30 t_0$, a time shorter than the typical times scale of fragment formation for energies lower than $E = 0.9\epsilon$ (see I for details).

In [24] a method to calculate the MLE which avoids the above-mentioned saturation was developed. In this method, after a time step $\tau \ll \tau_{sat}$, the distance $d(\tau) = d_1$ is rescaled to d_0 in the maximum growing direction and the quantity $\ln[d_1/d_0]$ is saved. Repeating the procedure at every time step τ, the logarithmic increments $\ln[d_i/d_{i-1}]$ are collected. The MLE is defined as

$$\lambda = \lim_{n\to\infty} \frac{1}{n\tau} \sum_{i=1}^{n} \ln \left| \frac{d_i}{d_{i-1}} \right|. \tag{3}$$

The ratio d_i/d_{i-1} is a measure of the exponential divergence between two initially nearby orbits along the maximum growth direction at time $i\tau$. In this way, Eq. (3) can be understood as an average of ''local Lyapunov exponents'' along a infinite trajectory, each one measuring the chaoticity (in the sense of divergence of initially nearby orbits in the phase space) in each time interval, at times $i\tau$. [This local exponent, as we will define in Eq. (4), can be considered as a short-path estimation of the MLE and has been used in [17] to analyze the evolution of ergodicity of three- and seven-particle Argón's clusters. Also in [25], for the same kind of system, the complete spectrum of local Lyapunov exponents has been used to analyze the dynamics of the system in terms of the local structure of the potential energy surface of the clusters.]

If we use Eq. (3) to calculate the MLE for our case of interest, i.e., a drop that fragments, we would be averaging over the local behaviors, erasing the information we are interested in. Because the system is changing quickly in the first stages and we want to follow the evolution with time of the local MLE, we define a maximum local Lyapunov exponent associated with the ith interval of size $(N_i\tau)$, beginning at $t_i = i\tau$ as

$$\lambda_i' = \lambda'(t_i) = \frac{1}{N_i\tau} \sum_{j=i+1}^{i+N_i} \ln \left| \frac{d_j}{d_{j-1}} \right| = \frac{1}{N_i\tau} \ln \left| \frac{d_{i+N_i}}{d_i} \right|, \tag{4}$$

with N_i finite. This is valid for the given main point. Because the system we study is characterized by its macroscopic state (i.e., the energy), we average over an ensemble of main points at t_i corresponding to the set of trajectories macroscopically equivalent (i.e., with the same energy) at $t=0$.

The MLLE can also be calculated with a slightly different method. It consists also in generating a set of initial conditions that evolve in phase space, but after a period of time $\tau \gg \lambda^{-1}$, one randomly generates a new set of ''sons,'' erasing the information about the previous correlations. In this way we characterize different regions of the phase space visited by the system with its maximum local Lyapunov exponents calculated as

$$\lambda_i = \lambda(t_i) = \frac{1}{N_i\tau} \ln[d(t_i + N_i\tau)/d(t_i)]. \tag{5}$$

Once again an average over macroscopically equivalent main points is performed. We have found that both ways of calculating the MLLE [using Eqs. (4) and (5)] give equivalent results.

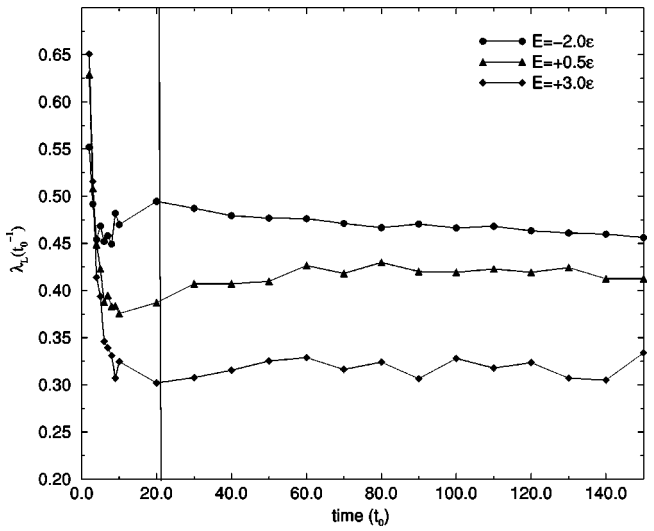

FIG. 3. Maximum local Lyapunov exponent (λ_L) as a function of time for the same three energies of Fig. 1: solid circles for $E=-2.0\epsilon$, solid triangles for $E=+0.5\epsilon$, and solid diamonds for $E=+3.0\epsilon$.

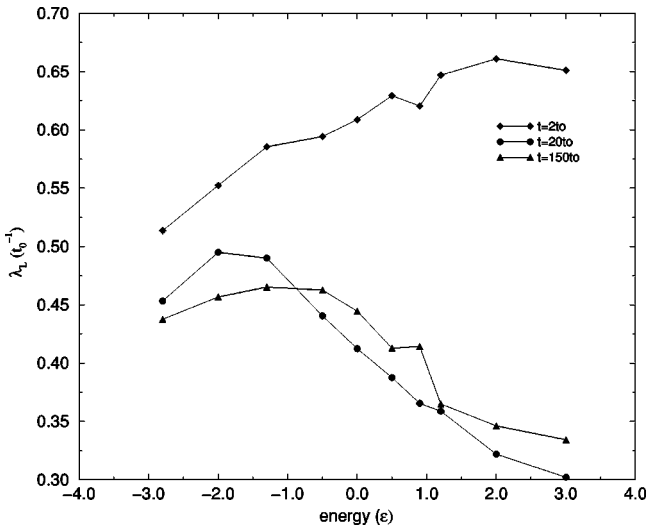

FIG. 4. Maximum local Lyapunov exponent (λ_L) as a function of energy for three different times: $t=2t_0$ (solid diamonds), $t=20t_0$ (solid circles), and $t=150t_0$ (solid triangles).

V. RESULTS OF NUMERICAL COMPUTATIONS

A. MLLE as a function of time and energy

In this work, we calculate the MLLE's using Eq. (4) applied to the dynamical evolution of the N=147 Lennard-Jones excited drops, with $\tau=0.01t_0$ and $N_i=1000$, so we have a value of the MLLE every $10t_0$, except for the first $10t_0$ of the evolution for which we use $N_i=100$ and then $N_i\tau=1t_0$. We calculate the MLLE in a range of energies from -2.8ϵ to $+3.0\epsilon$. The behavior of the MLLE as a function of the time can be seen in Fig. 3 for the same three energies analyzed in Fig. 1. The same behavior for the three energies analyzed can be seen: the MLLE's decrease sharply in the first $20t_0$ of the evolution, reaching an almost stationary value after this time.

This behavior shows us that we can classify the time evolution of the system in two well-differentiated stages: the early one, when the chaoticity of the drop is decreasing quickly, and the asymptotic one, characterized by the stability of the values of the MLLE. We can define a characteristic time τ_{mle} as the time at which the MLLE's reach their asymptotic values. In this system $\tau_{mle}=20t_0$.

It can also be seen that the dependence with energy is different at very early times than at asymptotic times. In order to illustrate this, we plot in Fig. 4 the MLLE as function of the energy at three relevant times. At $t=2t_0$, the MLLE is an increasing function of the energy. This behavior can be understood because the drop has not developed radial flux yet; it is still compressed and fragments are not formed yet. Due to the absence of collective radial motion, all the excitation energy goes into ''chaotic motion of the particles'' and the system behaves as an infinite one. We can denote this kind of behavior as ''infinite system like.'' We have found that for $t\geqslant 20t_0$ (at this time the system is fully in the asymptotic regime) the MLLE displays a maximum. This asymptotic behavior depends mainly on the size and the temperature of the biggest fragment of the fragmented drop, as we will see in the next section, but if we look at the mass spectra in Fig. 1, we can see that at the energy for which the maximum of the MLLE occurs, the asymptotic biggest fragment is as big as the whole system. Moreover, this peak is centered at about $E=-2\epsilon$, which is the value of the excitation energy for which the caloric curve reaches its maximum temperature's plateau (this temperature is called T_{lim}; see Fig. 1). As we said in Sec. III, such a plateau signals the onset of the fragmentation regime. In this case we can talk about ''finite-size-like behavior'': the asymptotic MLLE brings information about the dominant fragment in the asymptotic regime.

In the next subsection, we shall calculate the asymptotic values of the MLLE of the drop, as a function of the MLE of its fragments. This calculation will allow us to understand the role of the biggest fragment in the asymptotic value of MLLE.

According to the dependence with time of the MLLE, we can infer that the chaoticity of the system is related to the collisional process between particles, which depends strongly on the radial collective mode. In order to quantify the strength of the interparticle collisions and understand the behavior of the MLLE, we calculate the mean momentum transfer between particles as a function of the energy and time $[C(t)]$. It is defined as

$$C(t)=\frac{1}{N_{conf}}\sum_{j=1}^{N_{conf}}\sum_{i=1}^{N} m\left|\vec{v}_i(t+\delta)-\vec{v}_i(t)\right|, \qquad (6)$$

where N_{conf} is the number of configurations analyzed for each energy, i.e., an average over the ensemble of initial conditions.

We work in a range of energies from $E=-5.0\epsilon$ to $E=+3.0\epsilon$. The behavior of the MMT is exemplified in Fig. 5 where we show the time dependence of the MMT for the same three excitation energies analyzed in Figs. 1 and 3. We find that the MMT behaves in a similar way as the MLLE: at early times the collisions decrease abruptly until they reach their almost stable asymptotic values, and the dependence with the energy in this figure is similar to the one showed by the MLLE in Fig. 3.

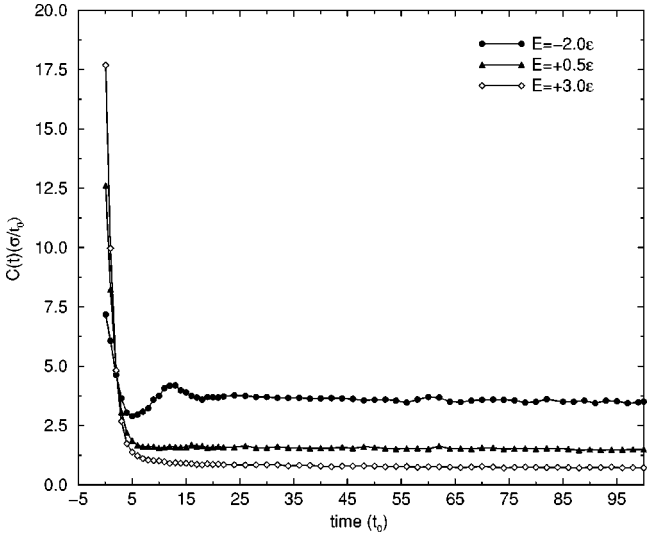

FIG. 5. Mean momentum transfer between particles $[C(t)]$ as a function of time for the same three energies of Fig. 1: solid circles for $E=-2.0\epsilon$, solid triangles for $E=+0.5\epsilon$, and open diamonds for $E=+3.0\epsilon$.

Moreover, if we look at the behavior of the MMT for $E=-2.0\epsilon$, it displays a loop between $5t_0$ and $15t_0$. We have found this behavior for our simulations in the range $-2.0\epsilon<E<-1.3\epsilon$. This feature can be explained if we study the behavior of the root mean square radius (R_{rms}) of the biggest fragment as a function of the time. In Fig. 6 we can see how for one of the mentioned energies ($E=-2.0\epsilon$) the R_{rms} displays a loop but inverted with respect to the MMT: when the R_{rms} rises, the MMT decreases and vice versa. This can be interpreted as follows: first the system expands while evaporating a few particles, then contracts and heats up, and finally expands again, reaching the asymptotic state.

To obtain the dependence of the MMT with all the energies analyzed for the different stages of the evolution we plot in Fig. 7 the MMT as a function of the energy for three relevant times: at very early time ($t=2t_0$), at an intermediate time ($t=20t_0$), and at very long time, when the fragments are well into their asymptotic regime, i.e., $t=150t_0$. At $t=2t_0$ the MMT is an increasing function of the excitation energy. On the other hand, for $t\geqslant 20t_0$ the MMT displays a maximum for energies at which evaporation is the dominating decay mode. This maximum is due to the fact that after evaporation of a few monomers we are left with a big fragment at the maximum temperature it can sustain (see I).

We can also see that for $t\geqslant 20t_0$ the dependence of MMT with the energy remains essentially constant. It allows us to define a characteristic time scale, i.e., the time at which the MMT reaches its asymptotic behavior. This time is $\tau_{mmt}\simeq 10t_0$ for almost all the energies except those in the range $-2.0\epsilon<E<-1.3\epsilon$ (the range at which the R_{rms} displays a loop), for which $\tau_{mmt}\simeq 20t_0$.

In this context, we can see that the MLLE's are strongly dependent on the collective radial mode: At very early times, the drop is highly chaotic because all the excitation energy goes to interparticle collisions (the radial flux is not formed yet), but when the system expands the collisions between particles decrease and the system becomes less chaotic. This process ends when the radial collective mode is fully formed.

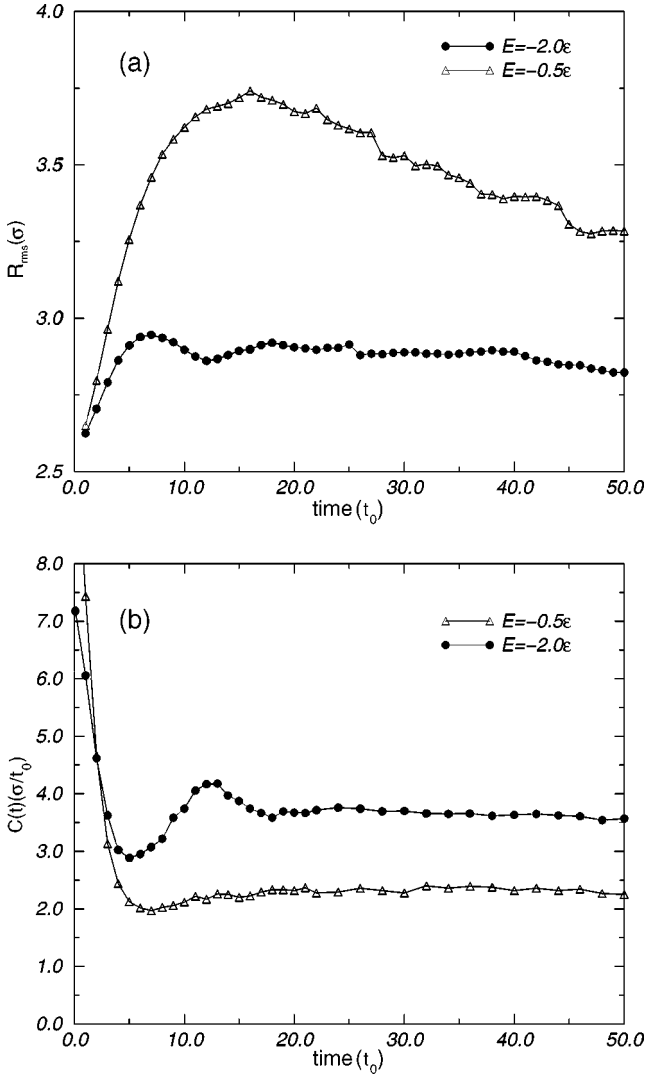

FIG. 6. Root mean square radius of the drop (R_{rms}) (a) and mean momentum transfer $[C(t)]$ (b) as a function of time for $E=-2.0\epsilon$ (solid circles) and $E=-0.5\epsilon$ (open triangles).

B. MLE in the asymptotic stage

Our main task in this section is to understand the behavior of the asymptotic MLLE found in the previous section, in terms of the MLE of the constituent fragments of the system. In order to do that, we calculate the MLE of the whole fragmented system and the MLE of its fragments at their maximum (limiting) temperature T_{lim}. We prepare the system in a broad energy range and solve the CEOM for times long enough to be very close to the asymptotic regime; i.e., the fragments are in long-lived metastable states. We then classify the clusters according to mass and calculate the MLE for each size using Eq. (3). The resulting MLE's are displayed in Fig. 8. It can be readily seen that the MLE is very well fitted by

$$\lambda = a(1-N^{-1/3}), \tag{7}$$

with $a=0.592t_0^{-1}$.

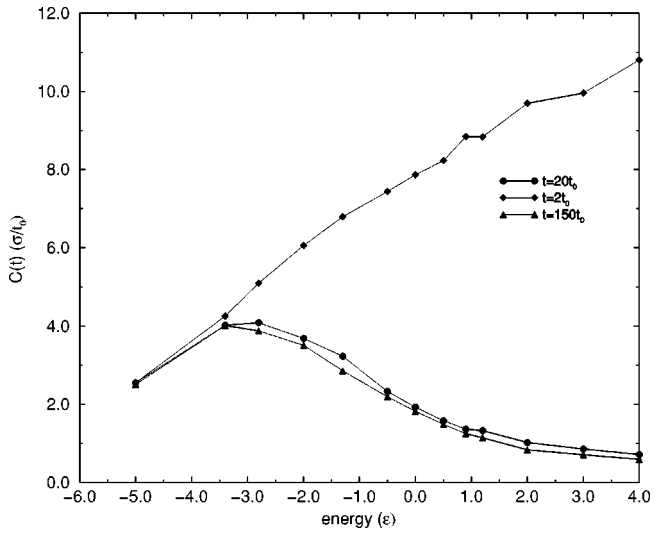

FIG. 7. Mean momentum transfer between particles $[C(t)]$ as a function of energy for three different times: $t=2t_0$ (diamonds), $t=20t_0$ (circles), and $t=150t_0$ (triangles).

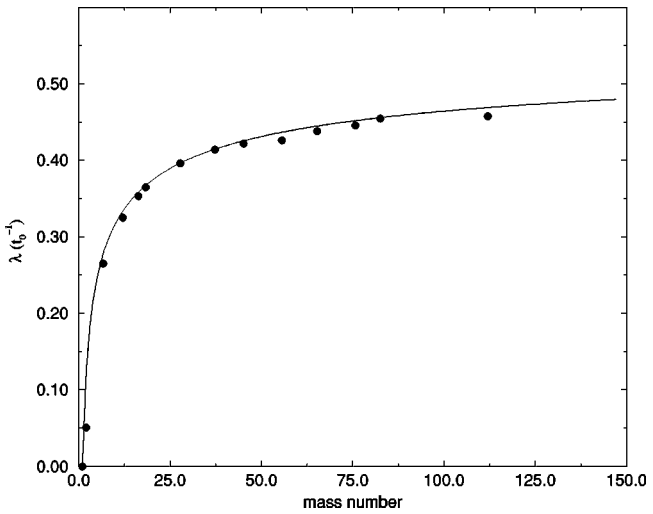

FIG. 8. Maximun Lyapunov exponent (λ, circles) as a function of mass of the asymptotic fragments together with the fitting function (see text for details).

This can be expressed as $\lambda=a(N-N^{2/3})/N$, where the first term represents the dependence on the volume of the fragment and the second term the dependence on the surface. In this way, if the size of the system goes to infinity, we are left with the MLE for an infinite system with temperature T_{lim}. Such a behavior has already been found in other systems [26,27].

We now focus on the whole fragmented system. Following Eq. (2), the expression for the distance between the trajectory of the main point and the trajectory of the ''son'' in the velocity space (d_v) can be cast in the following way:

$$d_v=\sqrt{\sum_{j=1}^{N_{clust}}\sum_{i=1}^{n_j}[(v_{xi}^{(2)}-v_{xi}^{(1)})^2+(v_{yi}^{(2)}-v_{yi}^{(1)})^2+(v_{zi}^{(2)}-v_{zi}^{(1)})^2]}, \tag{8}$$

with N_{clust} the number of clusters and n_j the number of particles in cluster j, from which it is immediate that

$$d_v=\sqrt{\sum_{j=1}^{N_{clust}}[d_0^j e^{\lambda_j t}]^2}, \tag{9}$$

which can be further approximated using $d_0\simeq cn_j$ (with c constant):

$$d_v=\sqrt{\sum_{j=1}^{N_{clust}}c^2(n_j)^2e^{2\lambda_j t}}. \tag{10}$$

By using the information of the asymptotic mass spectra (for the values of n_j) and the values of λ_j obtained from Eq. (7), we can fit accurately $d_v(t)$ in Eq. (10) with an exponential function. So we obtain the MLE of the whole system in terms of the MLE of its fragments. If we compare the results of this approximation with the standard calculation of the asymptotic MLE for the final stable configuration [using Eq. (3)], the results agree within 1%. But more information can be extracted by using our approximation. It allows us to understand the role of the biggest fragments in the final value of the MLE: as the biggest fragments drive the value of the MLE of the system, the small fragments practically do not contribute to it. The values of the MLE of the biggest fragments obtained from Eq. (7) are very close to the value of the MLE of the system calculated from Eq. (3) or (10).

With this result we can understand that the MLE of the whole asymptotic system is maximum at the excitation energy for which the biggest asymptotic fragment is as large and hot as possible. And the MLLE will reach its asymptotic value when its biggest fragment is almost formed in phase space.

Using Eq. (10) we can easily calculate the mean values and fluctuations of the asymptotic overall MLE's, in particular, the fluctuations given by $\langle\Delta\lambda\rangle=\sqrt{(\langle\lambda^2\rangle-\langle\lambda\rangle^2)}/\langle\lambda\rangle$.

In Fig. 9 we show the fluctuations of the overall MLE's as a function of the initial energy of the system. It is of special interest that this magnitude presents a noticeable change in slope at precisely the excitation energy at which the system displays a power-law-like asymptotic mass spectra.

Taking into account the dependence of the MLE with mass, it is rather easy to understand this behavior: at low energy we have basically a big fragment and a few very small fragments, but as we increase the energy the number of small fragments increases and big fragments are replaced by medium-mass ones. Finally at large enough energies the final configurations are mostly light fragments. When this behavior is combined with the behavior of the MLE as a function

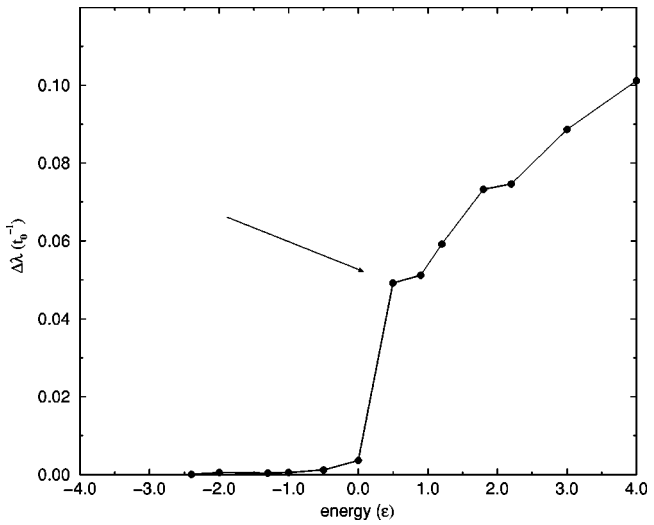

FIG. 9. Fluctuations of the maximum Lyapunov exponent of the 147-particle system ($\Delta\lambda$) as a function of the total energy. The arrow signals the energy for which a power law in the mass distribution is obtained.

of the mass, the fluctuations are amplified when the asymptotic state of the system is a mixture of fragments of all sizes, i.e., power law mass distribution.

VI. DISCUSSION

In this work we have analyzed the time evolution of fragmenting hot drops in terms of the MLLE. It is found that the early dynamics is highly collisional and it is characterized by MLLE values which are an increasing function of the energy as the MMT values are. In this regime the system behaves as an infinite system; all the excitation goes into chaotic motion of particles. For later times, as the system develops its radial collective mode, this trend is reversed. Rather early in the evolution, even before the fragmentation process is completed in phase space, the MLLE and the MMT attain their asymptotic behaviors. This can be understood because the system expands and fragments, thus diminishing the effect of collisions. For infinite systems one expects the MLE to be an increasing function of energy [28] but for finite systems with a free surface that develop a radial expansion this does not happen. In this case the system fragments, giving in the asymptotic regime a set of noninteracting fragments at their limit temperature (T_{lim}), and then the only source of trajectory divergence comes from interparticle collision of the constituents of the, not too hot, fragments. The rest of the energy goes into the collective motion which makes no contribution to the MLLE.

It is then clear that the system begins in a highly chaotic state and ''decays'' to a more ordered one. In this way, fragmentation can be viewed as a chaos-to-order transition.

If we now focus on the asymptotic regime, we find that the value of the MLLE is dominated by the one corresponding to the biggest fragment. We also found that the fluctuations of the asymptotic MLLE display a noticeable jump at the energy for which a power law mass spectra is found, signaling that in this case we find fragments of all sizes.

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APPENDIX

In order to study the mechanisms that lead to fragmentation it is important to know the time at which the asymptotic fragments form and the one at which they are emitted.

In previous papers the main fragment recognition algorithms currently in use have been fully analyzed [29]. The simplest definition of clusters is basically a group of particles that are close to each other and far away from the rest. The fragment recognition method, known as the minimum spanning tree (MST), is based on the last idea (I). In this approach a cluster is defined in the following way: given a set of particles $i,j,k,\ldots,$ they belong to a cluster C if

$$\forall i \in C, \quad j \in C \| \mathbf{r}_i - \mathbf{r}_j | \leqslant R_{cl}, \tag{A1}$$

where $\mathbf{r}_i$ and $\mathbf{r}_j$ denote the positions of the particles and R_{cl} is a parameter usually referred to as the clusterization radius, and is usually related to the range of the interaction potential. In our calculations we took $R_{cl}=3\sigma$.

On the other hand, the early cluster formation model (ECFM) [30] is based on the next definition: clusters are those that define the most bound partition of the system, i.e., the partition (defined by the set of clusters $\{C_i\}$) that minimizes the sum of the energies of each fragment according to

$$E_{\{C_i\}} = \sum_i \left[\sum_{j \in C_i} K_j^{\text{c.m.}} + \sum_{j,k \in C_i} V_{j,k} \right], \tag{A2}$$

where the first sum is over the clusters of the partition, and $K_j^{\text{c.m.}}$ is the kinetic energy of particle j measured in the center-of-mass frame of the cluster which contains particle j. The algorithm [early cluster recognition algorithm (ECRA)] devised to achieve this goal is based on an optimization procedure in the spirit of simulated annealing [30].

It has long been known that the ECRA algorithm finds that the asymptotic clusters are formed, in phase space, long before they separate in coordinate space, and become recognizable with the MST algorithm, i.e., long before they are emitted [10,11,29,31]. We then associate the time at which the ECRA method finds the asymptotic clusters to the fragment formation time scale $\tau_{ff}(E)$ and the one related to the MST analysis to the fragment emission time scale $\tau_{fe}(E)$.

An important quantity in this analysis is the microscopic stability of the clusters. In order to achieve this goal the microscopic persistence coefficient was defined. At a given time t the system will be formed by a set of clusters $C_i(t)$ which will become, for long times, the asymptotic fragments which we will denote C_i. Let us consider a given cluster $C_i(t)$ with mass number $n_i(t)$; let $b_i(t)=n_i(t)[n_i(t)-1]/2$ be the number of pairs of particles in the cluster. Its constituent particles might be at the asymptotic time in two or more different clusters. We define $a_i(t)$ as the number of pairs of

particles that belong to $C_i(t)$ and also are together in a given asymptotic cluster. We are now able to define the microscopic persistence coefficient

$$P(t)=\frac{1}{N_{ev}}\sum_{ev}\frac{1}{\sum_{cl} m_i(t)}\sum_{cl} m_i(t)\frac{a_i(t)}{b_i(t)}, \qquad \text{(A3)}$$

where the first sum runs over the different events for a given energy, N_{ev} is the number of events, and the other two sums run over the clusters at time t. It is clear that the persistence coefficient is equal to 1 if the microscopic structure of the clusters is equal to the asymptotic one. On the other hand, this coefficient approaches 0 when the two partitions under study bear little similarity. This coefficient can be defined for ECRA clusters as well as for MST ones.

Then we can define the $\tau_{ff}(E)$ related to the ECRA partition differing from the asymptotic one by an evaporation like process and the τ_{fe} related to the MST partition differing from the asymptotic one by an evaporationlike process.

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