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Critical dynamics in clusters of noble gas atoms

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Abstract. The multi-fragmentation dynamics of noble gas atomic clusters is considered for different statistically distributed deposited energies. The conditions giving rise to the development of criticality in the cluster evolution are revealed from an analysis of the signals in the fragment mass distribution. The time dependence of the observables related to critical exponents is studied. It is demonstrated that in a certain regime the cluster exhibits a behavior which can be identified as the precursor of a second-order liquid-gas phase transition.

PACS. 05.70.Fh Phase transitions: general studies – 05.70.Jk Critical point phenomena – 64.70.-p Specific phase transitions

Significant effort has been devoted in recent years to the study of critical behavior (*e.g.* a phase transition in infinite matter) in finite systems [1]. In the vicinity of critical conditions, the systems experience fluctuations at all scales; as a consequence, the relevant phenomena display universal features, being insensitive to the details of finite range forces and, therefore, important for the understanding of general characteristics of various objects.

In recent papers [2,3] we have performed an extensive study of the asymptotic fragmentation pattern of hot rare-atom clusters. By employing the analytical tools based on the analysis of statistical, correlation and fluctuation properties of fragment mass distributions we found strong evidence for a finite-size precursor of a second-order liquid-gas phase transition. However, in order to identify the detected behavior with the phase features of a finite system an analysis of the time evolution of criticality development is required [4]. Therefore, in this contribution we consider the time evolution of the observables which are related to the critical exponents, as an important aspect of dynamical effects. In addition, we also explore the criticality signal which is associated with the variance of the size of the biggest fragment. This quantity makes use of the finiteness of atomic clusters and cannot be employed in the analysis of an infinite system.

In the present study we use the dynamical model which has been described in reference [2]. We briefly recall that the cluster evolution is treated within Classical Molecular Dynamics. The atoms of the cluster move classically under the influence of a modified two-body Lennard-Jones potential; its parameters correspond to those of the argon liquid, $\{\epsilon, r_0, m\} = \{119\ \mathrm{K},\ 3.83\ \text{Å},\ 40\ \mathrm{amu}\}$. All observables are measured in these units.

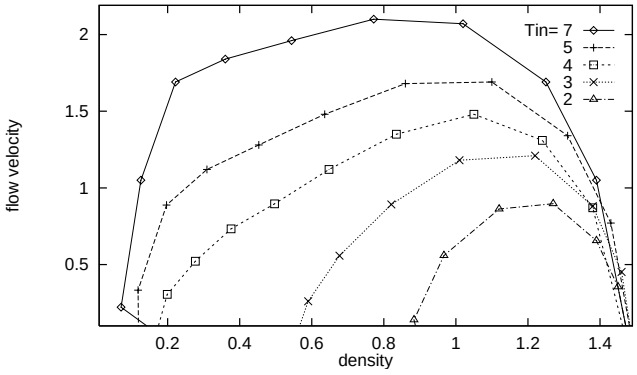

Fig. 1. The average flow velocity as a function of density in the expansion of an Ar_{300} cluster. For each T_{in}, the system starts at the highest density and proceeds along the respective curves to lower density.

The cluster is initialized in a local potential energy minimum by employing the frictional cooling method [5]. Using Metropolis sampling [6] it is then excited to a temperature T_{in} (giving a Maxwellian velocity distribution to the atoms), and the total energy of the cluster is kept fixed. In the initial stage the cluster dynamics display small fluctuations [2] implying that correlations perturbatively affect the system evolution which is based on a mean-field picture (*cf.* [7]). Such a picture leads to a time-reversible isentropic expansion process which is conveniently described in terms of the collective flow (*cf.* [8]). Figure 1 shows the density dependence of the average collective flow velocity of the biggest fragment,

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defined as

$$v_{\rm fl} = A_{\rm cp}^{-1} \sum_{\rm cp} \mathbf{v}_{i,\rm cm} \mathbf{n}_{i,\rm cm};$$
$$\mathbf{n}_{i,\rm cm} = \mathbf{r}_{i,\rm cm}/r_{i,\rm cm}, \quad (1)$$

where the index "$i,\rm cm$" indicates that the respective quantities are related to the center-of-mass of the fragment, and "cp" denotes that the sum is running over only those 50% ($A_{\rm cp}$) of fragment atoms which belong to the central region of the fragment. The flow velocity sharply increases and changes only weakly (during the isentropic regime of cluster evolution) until the cluster is decomposed into fragments of various masses in the second (dynamical) regime of its evolution [2]. This behavior is typical; we find the same features for the evolution of Ar_{100} and Ar_{500} clusters as well.

Figure 2 presents a few examples for the time evolution of the inclusive fragment mass distributions, averaged over at least 1000 cluster fragmentation events. At low temperatures the mass distribution remains "U" shaped until it stabilizes at asymptotically large times. At high temperatures the time evolution shows within a short time interval a dramatic change from a "U" shaped distribution to a monotonic decrease. According to Fisher's droplet model [9], such an average mass distribution can provide a first clue as to the occurrence of a critical behavior. For instance, in the case of overcritical evolution (*i.e.* vaporlike behavior at temperatures beyond the critical temperature) the inclusive mass distribution can within the thermodynamic limit be written in the form [2,9]

$$N(A) \propto A^{-\tau} \exp\Big(\frac{\Delta\mu}{k_{\rm B}T} A\Big); \quad (2)$$

where $\Delta\mu = \mu_{\rm g} - \mu_{\rm l}$ represents the difference of the gas and the liquid chemical potentials, respectively, and τ is related to some critical exponents through scaling laws of critical phenomena; the respective term accounts for a reduction of the available phase-space volume of surface fluctuations because the fragment surfaces are closed on themselves [2,9]. Since at the critical point the difference of the chemical potentials vanishes, the fragment mass distribution formed at the critical conditions follows a power law. This is a consequence of the strong large-scale density fluctuations which favor the formation of fragments with fairly wide mass spectra. Such a behavior, in conjunction with the value of the exponent τ, suggests a signature of critical evolution, *i.e.* the precursor of a second-order liquid-gas phase transition. Indeed, from Figure 2 one sees that at the initial temperature $T_{\rm in} \approx 4\epsilon$ the asymptotic inclusive mass distribution is very close to a power law, with $\tau \approx 2.23$, which is the value for condensation near the critical point as expected from Fisher's droplet model [2,9].

To further analyse the time development of the fragmentation process and to identify a possible critical behavior we use the reduced variance

$$\gamma_2 = \frac{M_2 M_0}{M_1^2}, \quad (3)$$

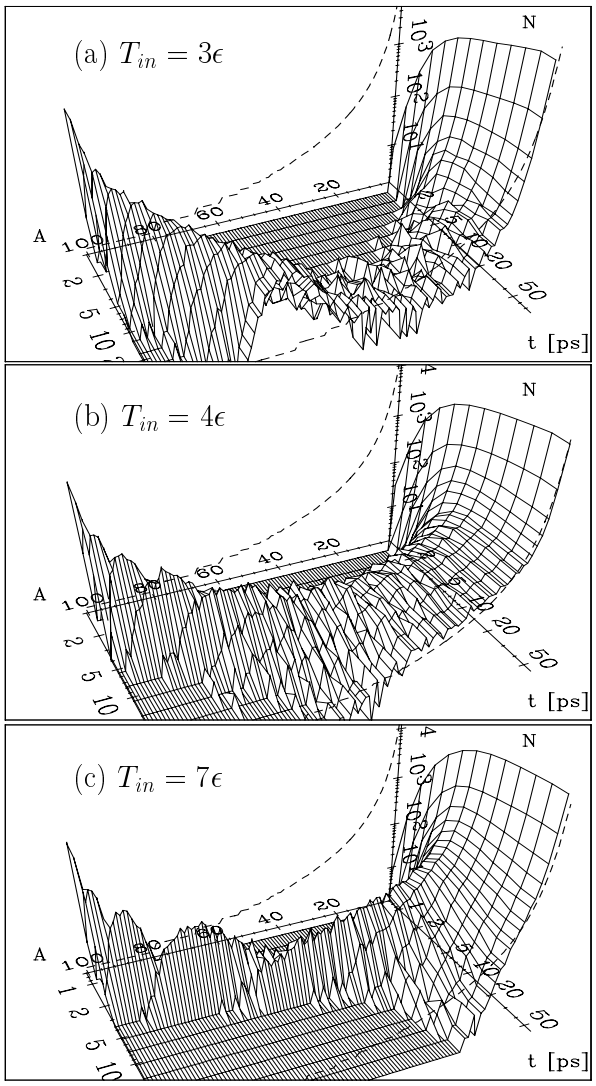

Fig. 2. Mass distributions at different time steps during the expansion of an Ar_{100} cluster with initial temperatures $T_{\rm in}/\epsilon =$ 3 (a), 4 (b) and 7 (c). The dashed lines give a power law yield $N \sim A^{-\tau}$ with $\tau = 2.23$.

where the conditional moments $M_k^{(j)}$ are defined through the fragment mass distribution $n^{(j)}(A)$ for the jth event as [10]

$$M_k^{(j)} = \sum_A A^k n^{(j)}(A). \quad (4)$$

It is an important feature of the quantity γ_2 that it displays a peak at conditions corresponding to the critical evolution of the system [2,10]. Using equation (2) and replacing the summation in equation (4) by an integration, the moments near the critical point in infinite matter can be estimated as $M_k \propto |\Delta\mu|^{-(1+k-\tau)}$. By assuming that at distance δ from the critical point the difference between the chemical potentials of the liquid phase and the gas phase vanishes as $\Delta\mu = \delta^\nu$, one recovers Campi's formula [10] for the moments at the critical point, $M_k \propto |\delta|^{-\nu(1+k-\tau)}$. Since τ is limited [2,9] between 2

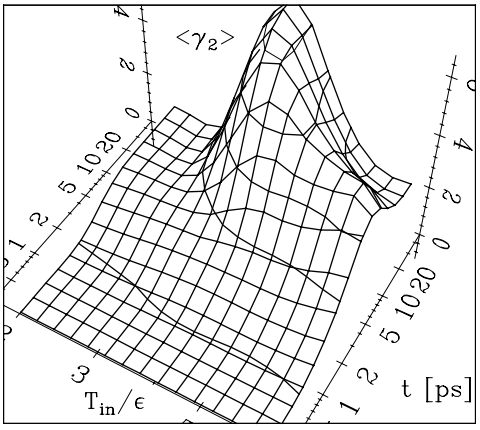

Fig. 3. Reduced variance γ_2 *versus* initial temperature, $T_{\rm in}$, and time for the expansion of Ar_{300}.

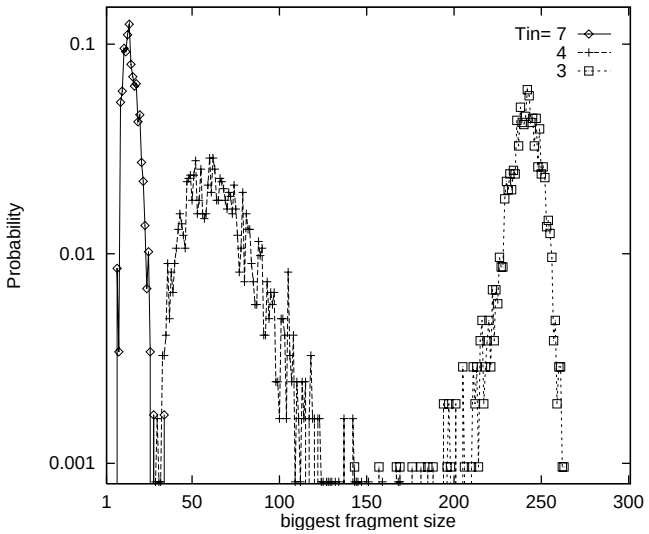

Fig. 4. The probability distribution of the size of the biggest fragment at three different initial temperatures for the fragmenting Ar_{300} cluster.

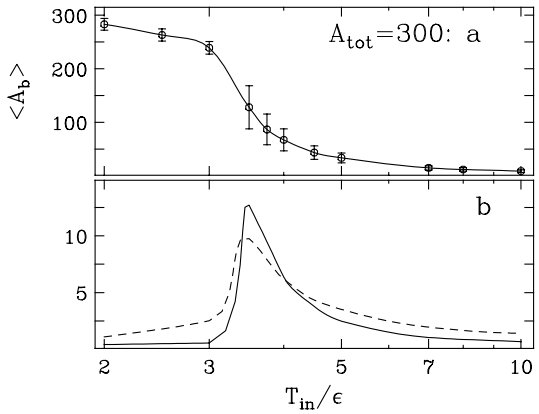

Fig. 5. The average mass of the biggest fragment (a, the errorbars display the quantity $\Delta_{A_{\rm b}}$), the NVB (b, solid line) and the asymptotic values of $\langle\gamma_2\rangle$ (b, dashed line) as a function of the initial temperature $T_{\rm in}$ for the fragmentation of Ar_{300}.

and 2.5, the zeroth and first moments are finite, while the higher moments diverge as the infinite system approaches the critical point.

Although in a finite system all moments M_k remain finite, they can nevertheless keep some qualitative features (if any) of critical behavior in an infinite system [2]. To trace such a criticality signal we calculate the averaged reduced variance

$$\langle\gamma_2\rangle(T_{\rm in},t)=\frac{1}{N_{\rm ev}(T_{\rm in})}\sum_j\gamma_2^{(j)}(t), \qquad (5)$$

where $\gamma_2^{(j)}(t)$ is determined for each event j of $N_{\rm ev}(T_{\rm in})$ events with the initial temperature $T_{\rm in}$. From Figure 3 one sees that at low initial temperatures (liquid-like events at $T_{\rm in}<4\epsilon$) the reduced variance γ_2 grows monotonically with time and saturates at large times. In contrast, for high initial temperatures (vapor-like events) we observe a pronounced non-monotonic behavior of γ_2 as a function of time, displaying a maximum in the dynamical regime of the system evolution. This property indicates that the fragments which are produced during the dynamical regime still contain a considerable amount of energy and are cooled by evaporation of mostly monomers (compare the time evolution of the mass distribution in Fig. 2). Note that at transitional temperatures ($T_{\rm in}$ between 3.5ϵ and 4ϵ) the reduced variance γ_2 peaks at asymptotic times as well as in the dynamical regime; this further corroborates the assertion of critical behavior at these temperatures.

It has been suggested [11] that a possible signal of critical behavior could be found in the size fluctuations of the biggest fragment. Supposedly, cluster size distributions for critical events show a maximum of fluctuations since the entire system becomes strongly correlated. As we have seen above, the maximum of the reduced variance $\langle\gamma_2\rangle$ corresponds to those events which are transitional between the liquid-like and vapor-like configurations. Due to the conservation of the number of atoms, the size of the biggest fragment should then display large fluctuations (*cf.* [12]) as well. Figure 4 shows the probability distribution of the mass of the biggest fragment for different initial temperatures. We observe that the width of the distribution attains its maximum at transition temperatures, $T_{\rm in}\approx 4\epsilon$. At these temperatures it is interesting to note the pronounced fluctuations in the probability distribution which are apparently not of statistical origin.

For the following, we thus consider the normalized variance of the size of the biggest fragment, NVB,

$$\gamma_{\rm NVB}=\frac{\Delta^2_{A_{\rm b}}}{\langle A_{\rm b}\rangle},$$
$$\Delta^2_{A_{\rm b}}=\langle A_{\rm b}^2\rangle-\langle A_{\rm b}\rangle^2, \qquad (6)$$

as a signal of criticality. Here again the brackets $\langle\cdot\rangle$ indicate ensemble-averaging as in equation (5). Figure 5 displays the initial temperature dependence of the average mass of the biggest fragment (Fig. 5a) and the reduced variance $\langle\gamma_2\rangle$ (Fig. 5b, dashed line) together with the values of NVB (Fig. 5b, solid line) taken at asymptotically

large times. One sees that the NVB signal shows a maximum located at $T_{\rm in}$ between 3.5ϵ and 4ϵ which is quite close to the value obtained from the analysis of the other signals [2].

In summary, we have considered the time trace of rare-atom cluster multi-fragmentation depending on the value of statistically distributed deposited energy (*initial temperature*). Conditions can be identified which correspond to the occurrence of critical behavior; the time evolution of characteristic observables in the dynamical regime confirms the vicinity of critical conditions. Finally, the normalized variance of the biggest fragment size distribution shows almost the same features as the reduced variance γ_2 of the fragment mass distribution, and, therefore, can be employed as an additional analytical tool for studies of critical behavior in a finite system.

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