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To be published in Physical Review letters
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Abstract

Using a recently developed multi-coincidence technique, the hydrogen cluster fragmentation resulting from a collision between a high energy $\text{H}_2^+$ ion with a $\text{C}_{60}$ target is investigated on an event by event basis. For a sample of 6000 collisions, statistical methods (conditional moments, scaled factorial moments) are applied to these data sets. The results obtained provide strong experimental evidence for the presence of a critical behavior in these finite systems.

PACS Numbers: 36.40.Ei, 5.70.Fh, 36.40.Qv
Due to the general relevance in science, interest in the observation and characterization of critical behavior in finite systems, in particular in nuclei and in clusters, has increased rapidly in the past years. Although small clusters do not exhibit exactly the first order phase transition known in infinite matter, Berry and co-workers have demonstrated that clusters may show a dynamic "solid-liquid" phase equilibrium which becomes a first order phase transition for large cluster sizes. Furthermore, by using molecular dynamics simulations to study cluster fragmentation the question on the existence and the nature of a finite-system analogue of a second-order phase transition (i.e., the correlation length becoming infinitely large at the critical point of a phase transition in normal macroscopic systems) has been addressed. Moreover, as such critical behavior is expected to manifest itself similar to the nuclear case in the characteristics of the fragment mass distribution resulting from hot fragmenting clusters, several experimental studies have been performed on the fragmentation of clusters induced by collisions under different experimental conditions. In all of these, a power-law fall-off as a function of fragment cluster size \( p \), i.e., \( p^{-x} \), has been observed with a critical exponent in close proximity to the critical exponent 2.6 found in nuclear fragmentation experiments and with prediction (=2.23) from Fisher's droplet model. Although these observations have been taken as a strong hint for the occurrence of critical behavior reminiscent of a second order phase transition in an infinite system, the mass yield shape alone cannot be considered as a conclusive proof. There exist several further predictions concerning the outcome of collisions close to the critical point (for instance the critical exponents, the intermittency signal, etc.), but these predictions can only be verified with data sets where complete analysis of the fragments is available on an event by event basis. These extended analysis methods have been proved to be valuable tools in several theoretical cluster fragmentation studies where the data have been generated by simulations. They have been already applied successfully to the analysis of nuclear collisions. However, to date no experimental data from cluster collision experiments were available.

Using in the present study a recently developed multi-coincidence technique for the measurement of the fragment size distribution induced by the collision of high energy \( H_{25}^+ \) ions with thermal \( C_{60} \) targets on an event by event basis, we have been able to obtain a first set of approximately 6000 completely analyzed collision events. This allows us to apply for the first time the method of conditional moments and of scaled factorial moments to measured
cluster fragment distributions. We find ample experimental evidence for the presence of a critical behavior in these molecular systems.

The apparatus used here is shown in Fig.1. Mass selected hydrogen cluster ions with an energy of 60 keV/amu are prepared in a high-energy cluster ion beam facility consisting of a cryogenic cluster jet expansion source combined with a high performance electron ionizer and a two-step ion accelerator. After momentum analysis by a magnetic sector field, the mass selected high energy projectile beam consisting in the present study of H$_{25}^+$ cluster ions is crossed perpendicularly by a C$_{60}$ effusive target beam (see Fig.1) produced by evaporation of pure C$_{60}$ powder in a single-chamber molybdenum oven at about 675 °C. One meter behind this collision region the high energy hydrogen collision products (neutral and ionized) are passing a magnetic sector field analyzer approximately 0.3 μs after the collision event. The undissociated primary H$_{25}^+$ cluster projectile ions or the neutral and charged fragments resulting from reactive collisions are then detected with a multi-detector device consisting of an array of surface-barrier detectors located at different positions, at the exit of the magnetic analyzer (see Fig.1). This allows us to record simultaneously neutral and charged fragments detected in coincidence for each single collision event (for more experimental details see Ref. 10 and 20) irrespective of the nature of the collisional interaction, i.e., small or large impact parameters leading to rather "gentle" or "violent" collisions.

The data sets thus obtained (=6000 events) are then analyzed by first determining for each event the number of fragments n(p) of size p and by constructing the conditional moments of the fragment size distribution as introduced by Campi

$$M_i = \sum_p p^i n(p)$$  \hspace{1cm} (1)

where i is the order of the moment. The summation is over all the fragments in the event except the heaviest one. One important test to gain a first insight into the shape of the fragment size distribution and to indicate the extent of critical behavior is to then generate the relative variance $\gamma_2$ defined as

$$\gamma_2 = \frac{M_2 M_0}{M_1^2}$$  \hspace{1cm} (2)
and to plot this quantity versus the temperature of the fragmenting system (the amount of energy in the fragmenting system). Since this quantity is not directly available, we are plotting here $\gamma_2$ versus the number of ions produced "NI" (see Fig.2) as this number is indicative for the average energy in the excited cluster ion before fragmentation.

Fig.2 shows the present $\gamma_2$ data which resemble the earlier results reported by Campi$^8$ who considered the properties of a three-dimensional bond percolation system (cubic lattice with 125 sites). In fact Campi$^9$ was the first to suggest that the methods developed in percolation theory may be of relevance to the analysis of multifragmentation data. In particular it was shown that quantities that display, close to the critical point, divergent behavior in macroscopic system (i.e. a phase transition of second order involving an infinitely large correlation length) are at least exhibiting a resonance like behavior in finite size systems (with the corresponding correlation length being of the size of the system itself).

In accordance with these considerations $\gamma_2$ has been demonstrated not only for percolation systems but also for simulated fragmentations of nuclei$^{10}$ and clusters$^{2}$ to have a peak around the critical point implying that the fluctuations are largest for critical conditions. The present experimental data are in good agreement with these predictions and results, and the occurrence of a peak in Fig.2 at around NI$=7$ confirms that at around this NI value the corresponding decaying hydrogen cluster ions are within the critical zone. At this point we can already conclude therefore that hot cluster ions are fragmenting similar to finite-size systems which show a second order phase transition for infinite size.

Because we are dealing in the present case with finite systems, additional methods are necessary to distinguish between dynamical fluctuations and statistical fluctuations. Thus in a next step we apply to our data sets one of the most convincing methods developed to analyze and categorize fluctuations and correlations, i.e., the analysis of event by event data in terms of intermittency. We now briefly outline the procedure. Intermittency is a statistical concept and it corresponds to the existence of non-statistical fluctuations which have self-similarity over a broad range of scales. The relevant information can be deduced from the horizontally scaled factorial moments (HSFM) which measure the properties of dynamical fluctuations without the bias of statistical fluctuations (deviations of the fluctuations from a Poissonian distribution)$^{11,22}$ given by

$$\text{(HSFM)}_i = M^{i-1} \sum_{k=1}^M \frac{n_k(n_k-1)...(n_k-i+1)}{N(N-1)...(N-i+1)}$$  \hspace{1cm} (3)