Cluster multifragmentation and percolation transition: A quantitative comparison for two systems of the same size

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Using a recently developed multicoincidence technique, fragmentation resulting from collisions between high energy (60 keV/u) H_{27}^+ ions with an He target are investigated on an event by event basis. The data obtained are analyzed in terms of fluctuations in the fragment size distribution. A comparison with results from two different three-dimensional (3D) lattice bond percolation models of the same size (27 constituents) indicates the presence of a critical behavior in the decaying finite system and the importance of accounting in the percolation model for the specific binding situation in the hydrogen cluster ion.

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Fragmentation covers a wide range of phenomena in science and technology, including polymers, colloids, droplets, and rocks. Therefore, fragmentation can be observed on different scales ranging from the femtometer size in the case of nuclear decay reactions to astronomical dimensions for collisions between galaxies [1]. Despite intensive research in these various fields, complete analysis and understanding of fragmentation has not yet been achieved. Nevertheless, it has been recognized recently that some features of this phenomenon are rather independent of the decaying systems and underlying interaction forces [2]. Thus it is highly desirable to identify those features which are common to all kind of fragmentations and independent of the size and interaction forces in order to identify more general principles. This can be achieved by studying either experimentally or theoretically (using appropriate models such as percolation models) a specific decaying system in as much detail as possible, and then comparing the various features obtained with other differently sized and bonded systems. It has been shown recently that a particular useful system for this purpose are clusters of atoms or molecules [3].

The hydrogen cluster ions, as the simplest ionized molecular systems, have recently attracted great interest and it was thus possible to clarify their structure and other properties [4–7]. Their structure has been calculated to consist of a central H_3^+ core which is solvated by shells of unperturbed H_2 molecules. One important feature of these cluster ions is the large difference between the strong intramolecular interaction (covalent bonding with an energy between 4 to 5 eV) and the rather weak intermolecular interaction (ion–dipole interaction with an energy close to 0.1 eV) [6,7]. Recently, fragmentation of hydrogen cluster ions induced by collisions with either atomic helium or fullerenes has been studied using a newly developed multicoincidence technique [8]. With this technique it was possible to detect and to identify, in terms of mass (to charge ratio), all neutral and (charged) fragments produced in a collision on an event by event basis [8]. Under all experimental conditions a power law falloff in the fragment mass distribution, i.e., $p^{-\tau}$, with p the fragment mass and τ an exponent, has been observed in these experiments with a τ derived which was close to (i) the critical exponent 2.6 found in nuclear fragmentation experiments and (ii) predicted values ($\simeq 2.23$) using Fisher's droplet model [9–14]. Based on these and other results, the question about the existence of a critical behavior (i.e., the occurrence of a second-order phase transition) in the fragmentation of hydrogen cluster ions has been addressed recently [15].

The percolation model enables the generation of events under conditions where the presence of a critical behavior has been demonstrated [12,16]. In this model, neighboring sites can be connected by bonds. Each of these bonds is randomly activated with a probability q. A fragmenting particle (cluster) can be viewed as an ensemble of neighboring sites connected by active bonds. By varying the parameter $q(0 \leqslant q \leqslant 1)$, the shape of the fragment size distribution changes. If the system is infinite this shape will approach for $q \rightarrow 0.25$ a power law resulting from a second-order percolation transition [16]. Moreover, Campi [12] was able to demonstrate the existence of a similar (power law) behavior also for finite system. As a matter of fact, he showed that the existence of a second-order percolation transition can also be seen in the characteristic behavior of plots of the average size $P_{\max}$ (where $P_{\max}$ is the size of the largest fragment produced in a single event for a given multiplicity) versus multiplicity, and by the presence of fluctuations in $P_{\max}$ in the vicinity of the critical point.

Here, we report on a comparison between (i) experimental results obtained by fragmentation of H_{27}^+ cluster ions induced by collisions with an He target and (ii) the results obtained

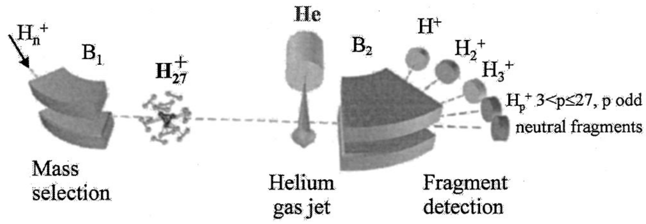

FIG. 1. Schematic diagram of the experimental setup.

by a calculation using a lattice bond percolation model (with the same number of bonds as the number of constituents in the hydrogen cluster ion) to simulate critical behavior. From the overall agreement between the two cases in plots of the average size $P_{\max}$ versus the multiplicity m, and also related plots concerning the presence of fluctuations, we can conclude and confirm (thereby extending the earlier conclusions based on the existence of power laws [13,15,17]) the existence and presence of a critical behavior in the fragmentation of hydrogen cluster ions. Slight deviations of the experimental data from the theoretical predictions by the standard percolation model used can be reconciled by the use of a novel modified (two-bond type model) percolation model, which takes into account the fact that in the hydrogen cluster ions two types of bonds are present and will influence the decay pattern.

The apparatus used here is shown in Fig. 1. Mass selected hydrogen cluster ions with an energy of 60 keV/u 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 two step ion accelerator. After momentum analysis by a magnetic sector field, the mass selected high energy projectiles consisting in the present study of H_{27}^+ cluster ions are crossed perpendicularly by a helium effusive target beam. 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_{27}^+ cluster projectile ions or the neutral and charged fragments resulting from reactive collisions are then detected with a multidetector device, consisting of an array of surface barrier detectors located at different positions at the exit of the magnetic analyzer. This allows us to record for each event, simultaneously, the sum of the masses of all the neutral fragments and, in coincidence, each charged fragment resulting from the interaction (for more experimental details, see Refs. [8], [9] and references therein).

After a thorough event by event data analysis, we are considering here the dependence of the average size $P_{\max}$ of the largest fragment produced in a single event on the total number of fragments, usually termed multiplicity m. Campi already showed that the shape of this dependence of $P_{\max}$ on multiplicity m will be different in the presence or absence of a critical behavior [18]. As the fluctuations in the fragment size distribution are largest near the critical point, it is interesting to also plot (as a measure for these fluctuations) the standard deviation of $P_{\max}$ versus the multiplicity and thus obtain a further characteristic fingerprint of the presence or absence of a critical behavior type of situation. In Fig. 2 we have plotted these two types of correlations (i.e., $P_{\max}$ versus m and the standard deviation of $P_{\max}$ versus m, both designated by filled circles) for 29041 analyzed events resulting from the interaction between H_{27}^+ and the He target. It can be seen that, in general, the average size of the largest fragment, $P_{\max}$, decreases with increasing multiplicity m. The data show a distinct change of slope at around $m=13$. In contrast, the standard deviation of $P_{\max}$ versus multiplicity curve has a bell-shaped form with a maximum peak value of 1.5 at $m=5$.

In order to simulate a critical behavior in a finite system, we have used here, as mentioned above, a three-dimensional (3D) percolation model with the same number of bonds in the lattice as the number of constituents in the hydrogen cluster ion studied, i.e., a cubic system with 27 sites $(3\times3\times3)$. We have carried out these calculations for differing probabilities q between 0 and 1 in steps of 0.01 and by creating 10 000 ''fragmentation events'' for each step. The corresponding results in terms of $P_{\max}$ versus multiplicity m and the standard deviation of $P_{\max}$ versus multiplicity for this model calculations are plotted in Fig. 2 and designated as open triangles. On a first sight there exists quite a good agreement in the general shape between the experimental data obtained in the H_{27}^+–He collision experiment for $P_{\max}$ versus m and the standard deviation of $P_{\max}$ versus m and the calculated data from this percolation model. As mentioned above, from this good agreement we can conclude and confirm (thereby extending the earlier conclusions based on the existence of power laws) the existence and presence of a critical behavior in the fragmentation of hydrogen cluster ions. This is the more convincing as calculations performed with a simple one-dimensional (1D) percolation model yield quite different results, e.g., not showing the peak in the standard deviation function so characteristic for the presence of critical behavior [18].

Moreover, it is interesting to note that there exist only two other experimental data sets allowing such an analysis as the present one. One data set (comprising about 400 events) comes from a nuclear fragmentation experiment, i.e., consisting of a nearly complete fragment charge analysis of 1 GeV/amu Au ions bombarding an emulsion [19]. The data are exhibiting a similar behavior as the present cluster collision data and 3D percolation model [18,20]. It should be mentioned, however, that according to DeAngelis *et al.* [20], this nuclear fragmentation data set is very likely biased towards low multiplicity events as the experimental setup records only those events in which all 79 charges are detected. In contrast, the present cluster collision experiment includes all events and the comparison with the percolation model is unbiased from the experimental detection probability. Moreover, using similar plots as in the present case, Bonasera [21] has recently compared two systems of different nature and scale finding a surprising similarity between experimental results for gold–gold nuclear collisions [22] and cluster ion-fullerene collisions [3]. In passing, Bonasera notes in this essay that simulations of fragmenting systems based on classical molecular dynamics give predictions that are similar to these experimental results obtained [3,22].

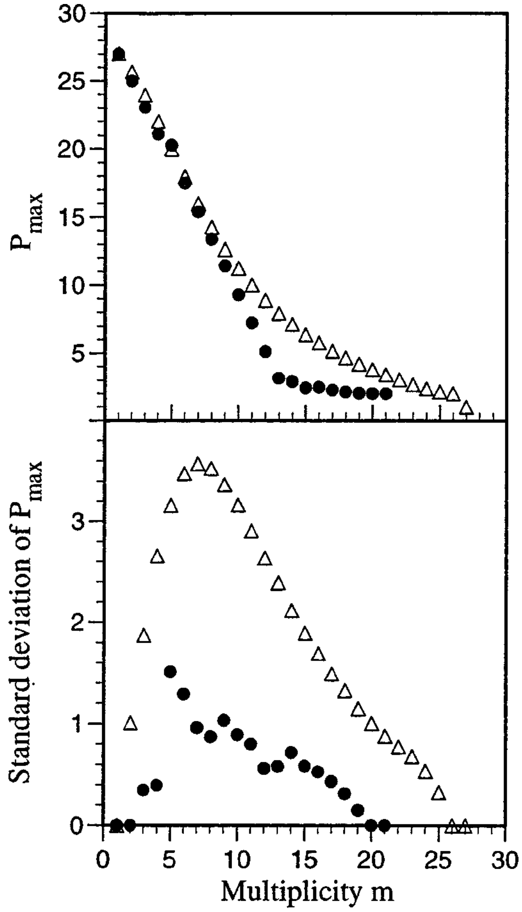

FIG. 2. Comparison of the fragmentation behavior of an H_{27}^+ cluster ion designated by filled circles and the results from a 3D lattice bond percolation model designated by open triangles. Upper part the average size of the largest fragment, P_{max}, versus the multiplicity, m, and lower part standard deviation (fluctuation) of P_{max} given in the upper part versus m. The standard deviation is defined as $\sqrt{(\langle P_{max}^2\rangle - \langle P_{max}\rangle^2)}$.

Nevertheless, as can be seen in Fig. 2, there exist slight deviations of the experimental data from the theoretical predictions by the 3D standard percolation model used. First of all, P_{max} of the experimental data decreases faster with increasing m than the calculated points and, in addition, the calculated points do not show the clear change in slope exhibited by the experimental data. Moreover, the absolute magnitude of the standard deviation of P_{max} is much smaller in the case of the experimental data (close to 1.5) than in the percolation model (close to 3.6). In order to reconcile these deviations of the model calculations from the experimental data, we have modified the percolation model taking into account the special type of binding situation in the hydrogen cluster ions. We have noted already above that one of the most striking features regarding protonated hydrogen clusters (as well as bulk molecular hydrogen) is the large difference between the very strong intramolecular interaction within the neutral cluster constituents H_2 and H_3^+ (4–5 eV, $r_{eq}=0.74$ Å), and the weak intermolecular potential between the cluster constituents (0.1 eV, $r_{eq}=1.8$ Å for $H_3^+(H_2)_3$) [6,7].

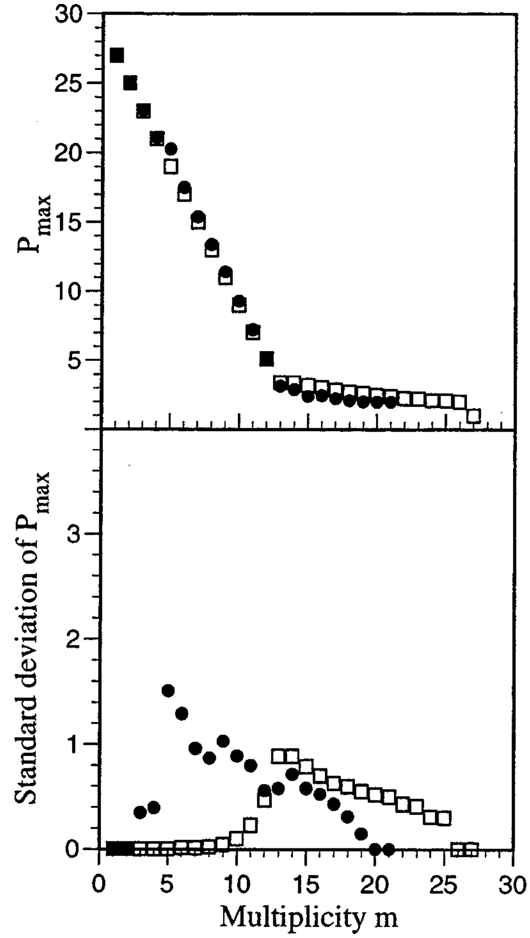

FIG. 3. Comparison of the fragmentation behavior of a H_{27}^+ cluster ion designated by filled circles and the results from a modified 3D two bonds percolation model (see text) designated by open squares. Upper part the average size of the largest fragment, P_{max}, versus the multiplicity, m, and lower part standard deviation (fluctuation) of P_{max} given in the upper part versus m.

Thus we have developed a novel type of percolation model (including two types of bonds) in order to take into account the fact that there are these two kinds of bonds in the hydrogen cluster ions, i.e., modeling the decay of the H_{27}^+ cluster ion [which is equivalent to $H_3^+(H_2)_{12}$] by a percolation lattice where the two differing bond strengths are characterized by two different probabilities q_s and q_w representing the two bond strengths: a strong one and a weak one. In the hydrogen cluster ion H_{27}^+ there exist 15 strong bonds: three strong bonds in the core H_3^+ and one bond associated with each covalent bond in the 12 H_2 molecules. Neglecting the very weak interaction between the neutral hydrogen molecules, there exist, in addition, also 12 weak bonds corresponding to the ion–dipole interaction between the core H_3^+ and each of the H_2 molecules.

In order to determine the relation between q_s and q_w, taking into account that the energy of the strong bond E_s is forty times the energy of the weak bond E_w in the hydrogen cluster ion, let us consider a thermodynamical system with these two kinds of bond in a heat bath of temperature T. In a canonical ensemble, the probability for activation of a strong bond can be written as

$$q_s = 1 - \exp\left(-\frac{E_s}{kT}\right). \quad (1)$$

As an analogy, the probability for activation of a weak bond is

$$q_w = 1 - \exp\left(-\frac{E_w}{kT}\right). \quad (2)$$

Therefore, we obtain a relation between the probabilities of

$$q_w = 1 - \exp\left(\left(\frac{E_w}{E_s}\right)\ln(1-q_s)\right). \quad (3)$$

''Events'' are generated in a similar manner as in the single bond percolation model, however, for each of the values the associated q_w is first calculated with Eq. (3) using $E_s = 40E_w$. In Fig. 3, we have plotted the correlation between P_{max} and multiplicity for the events generated from this two bonds lattice percolation model (open squares). First, a very good agreement for the decrease of P_{max} with multiplicity is observed between the H_{27}^+–He collision experiment and this new percolation model. We can clearly see the presence of a change of slope at a multiplicity of $m=13$. Second, a peak is also observed in the standard deviation of P_{max} with a maximum in the same order of magnitude as the one observed in the case of our H_{27}^+ experimental data (close to 1 for the new lattice bond percolation model and close to 1.5 for H_{27}^+ fragmentation).

Nevertheless, the maximum of the peak of the standard deviation of P_{max} versus multiplicity curve is localized at different multiplicity values, i.e., at $m=5$ in the case of H_{27}^+ and at $m=13$, $m=14$ in the case of the two bonds percolation model. In order to understand this difference, it is helpful to analyze in detail the experimental events and ''events'' by the percolation model with a multiplicity of $m=3$. It turns out that 98% of the experimental events with $m=3$ proceed via ''evaporation'' of two hydrogen molecules, i.e.,

$$(H_{27}^+)^* \rightarrow H_{23}^+ + 2H_2, \quad (4)$$

whereas about 2% of the experimental events involve a situation where one of the H_2 molecules is excited into a dissociative ionized state that subsequently decays into $H^+ + H$, i.e.,

$$(H_{27}^+)^* \rightarrow H_{25}^+ + H^+ + H. \quad (5)$$

The presence of two possible decay channels for $m=3$ leading to different P_{max} (i.e., H_{23}^+ and H_{25}^+, respectively) is the reason for a fluctuation in the experimental outcome and therefore resulting in a nonzero standard deviation value for P_{max}. For the two bonds lattice percolation model, however, all events are simple evaporation events (i.e., loss of two H_2 molecules) giving thus a zero standard deviation of P_{max} for $m=3$. Actually, in this two bonds percolation model no event is generated where a strong bond is broken unless also all the weak bonds are broken. This difference in the model and the experiment is responsible for the difference in the results obtained at low multiplicity. This difference, however, vanishes at higher multiplicity when, also in the percolation model, competing dissociation channels become available.

In conclusion, correlations between multiplicity and average size of the largest fragment and its standard deviation in single event data sets for a given multiplicity are good indicators for the presence or absence of critical behavior [18]. The present comparison of available experimental data obtained from the analysis of collisions between H_{27}^+ and an He target and results from a 3D bond lattice percolation model of the same size shows remarkable similarity in these correlations, in particular, when using instead of the simple cubic 3D lattice percolation model, a modified two bonds percolation model thereby taking into account some of the specific binding properties of the hydrogen cluster ion. From this similarity in correlations we conclude that in analogy to the situation for the percolation transition, also in the decaying hydrogen cluster ion we encounter the signature for critical behavior. The observation of a critical behavior has been interpreted for a long time as evidence for a second-order phase transition [14,17,23]. Recently, the critical behavior experimentally observed in the fragmentation of small systems has been demonstrated to be compatible with a first-order phase transition because of finite size effects [24]. Moreover, power laws have been reported along the Kertesz line [25] at higher densities and temperatures inside the single fluid part of the phase diagram [25,26]. Thus the presently observed transition is not necessarily a percolation transition. The detailed investigation of the caloric curve for these fragmentation reactions may give further insights about the true nature of this critical behavior.

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[1] R. P. Binzel, Nature (London) **388**, 516 (1997).
[2] *Fragmentation Phenomena*, edited by D. Beyson, X. Campi, and E. Pefferkorn (World Scientific, Singapore, 1995).
[3] B. Farizon, M. Farizon, M. J. Gaillard, F. Gobet, C. Guillermier, M. Carré, J. P. Buchet, P. Scheier, and T. D. Märk, Eur. Phys. J. D **5**, 5 (1999).
[4] I. Stich, D. Marx, M. Parrinello, and K. Tenakura, J. Chem. Phys. **107**, 9482 (1997).
[5] I. Stich, D. Marx, M. Parrinello, and K. Tenakura, Phys. Rev. Lett. **78**, 3669 (1997).
[6] M. Farizon, H. Chermette, and B. Farizon-Mazuy, J. Chem. Phys. **96**, 1325 (1992).
[7] B. Farizon, M. Farizon, H. Razafinjanahary, and H. Chermette, Phys. Rev. B **60**, 3821 (1999).
[8] B. Farizon, M. Farizon, M. J. Gaillard, E. Gerlic, R. Genre, S. Louc, N. V. de Castro Faria, and G. Jalbert, Chem. Phys. Lett.

252, 147 (1996).
[9] B. Farizon, M. Farizon, M. J. Gaillard, M. Carré, J. P. Buchet, P. Scheier, and T. D. Märk, Int. J. Mass Spectrom. Ion Processes **164**, 225 (1997).
[10] J. E. Finn *et al.*, Phys. Rev. Lett. **49**, 1321 (1982).
[11] A. Hirsch *et al.*, Phys. Rev. C **29**, 508 (1984).
[12] X. Campi, Nucl. Phys. A **495**, 259c (1989).
[13] V. Latora, M. Belkacem, and A. Bonasera, Phys. Rev. Lett. **73**, 1765 (1994).
[14] M. E. Fisher, Rep. Prog. Phys. **30**, 615 (1967).
[15] B. Farizon, M. Farizon, M. J. Gaillard, F. Gobet, M. Carré, J. P. Buchet, P. Scheier, and T. D. Märk, Phys. Rev. Lett. **81**, 4108 (1998).
[16] *Introduction to Percolation Theory*, edited by D. Stauffer and A. Aharony (Taylor and Francis, London, 1992).
[17] J. A. Hauger *et al.*, Phys. Rev. Lett. **77**, 235 (1996).
[18] X. Campi, Phys. Lett. **208**, 351 (1988).
[19] C. J. Waddington, P. S. Freier, Phys. Rev. C **31**, 888 (1985).
[20] A. R. DeAngelis, D. H. E. Gross, R. Heck, Nucl. Phys. A **537**, 606 (1992).
[21] A. Bonasera, Phys. World , 20 (February 1999).
[22] M. D'Agostino *et al.*, Phys. Rev. Lett. **76**, 2646 (1999).
[23] J. B. Elliott *et al.*, Phys. Lett. B **418**, 34 (1998).
[24] F. Gulminelli, P. Chomaz, Phys. Rev. Lett. **82**, 1402 (1999).
[25] J. Kertész, Physica (Amsterdam) **161A**, 58 (1989).
[26] X. Campi, H. Krivine, Nucl. Phys. A **620**, 46 (1997).