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Analyzing fragmentation of simple fluids with percolation theory

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Abstract. We show that the size distributions of fragments created by high energy nuclear collisions are remarkably well reproduced within the framework of a parameter free percolation model. We discuss two possible scenarios to explain this agreement and suggest that percolation could be an universal mechanism to explain the fragmentation of simple fluids. Experiments with atomic clusters would help to confirm this hypothesis.

PACS. 36.40.Ei Phase transitions in clusters – 36.40.Qv Stability and fragmentation of clusters – 25.70.Mn Projectile and target fragmentation

1 Introduction

Fragmentation phenomena concern a wide diversity of objects in nature, at many scales of distance and time. A natural question is to ask what is generic and what is specific in these phenomena. Most theoretical efforts made to understand fragmentation apply only to specific objects or, on the contrary, concern simple models with few links with reality [1]. Moreover, experimental data [2–5] are rather sparse and often the experimental conditions are ill defined. As a consequence, the question of the possible existence of universal fragmentation mechanisms remains an open problem [6].

The arguments in favour of the existence of universal mechanisms are of various orders. For instance, in aggregation phenomena, which can be considered as the opposite of fragmentation, it is possible to define universal classes [7,8] (diffusion limited aggregation, clustering of clusters...) in terms of the initial conditions (number of seeds) and the motion of the aggregating particles (ballistic, Brownian). The fractal structure (dimension) of the aggregation cluster is the fingerprint of these universal classes. In many fragmentation processes the experimental observation, over many orders of magnitude, of power law (scale invariant) fragment size distributions is another possible indication of universal classes. In this case, the value of the power law exponent would be the corresponding fingerprint [2,6].

Collision induced fragmentation of small fluid drops (atomic nuclei [9], atomic aggregates [10,11], liquid droplets [12]) is a field of experimental research particularly active because it offers the best possibilities of complete identification of the fragmentation products. We present in this paper an analysis of the fragmentation of atomic nuclei in high energy collisions. We show that random percolation theory accounts for experimental data without any adjustable parameter and we discuss two possible explanations for this agreement, depending if one assumes or not that thermal equilibrium is reached before fragmentation occurs. It is then suggested that this *percolation type* fragmentation mechanism could be universal for simple fluids, *i.e.* fluids made of structureless particles interacting with short range potentials.

The main aim of this paper is to draw the attention of the atomic cluster community to the possible universality of fragmentation processes. In this respect, the availability of fragmentation data from atomic clusters made of noble atoms would help to test the validity of this hypothesis.

The structure of the paper is the following. In Section 2, a brief description of a fragmentation experiment with atomic nuclei precedes the analysis of data. Section 3 is devoted to the interpretation of the results. Some final remarks and conclusions are in Section 4.

2 Analysis of fragmentation data

A proper identification of this *percolation type* fragmentation mechanism demands a detailed examination of the data. In particular (see below), one needs an identification of the fragments event by event. To our knowledge, no such data is available for the fragmentation of atomic clusters[1]. In order to illustrate with real data these processes

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[1] With the exception of the data on the fragmentation of hydrogen clusters of Farizon *et al.* [11]. See the comment in Section 3.

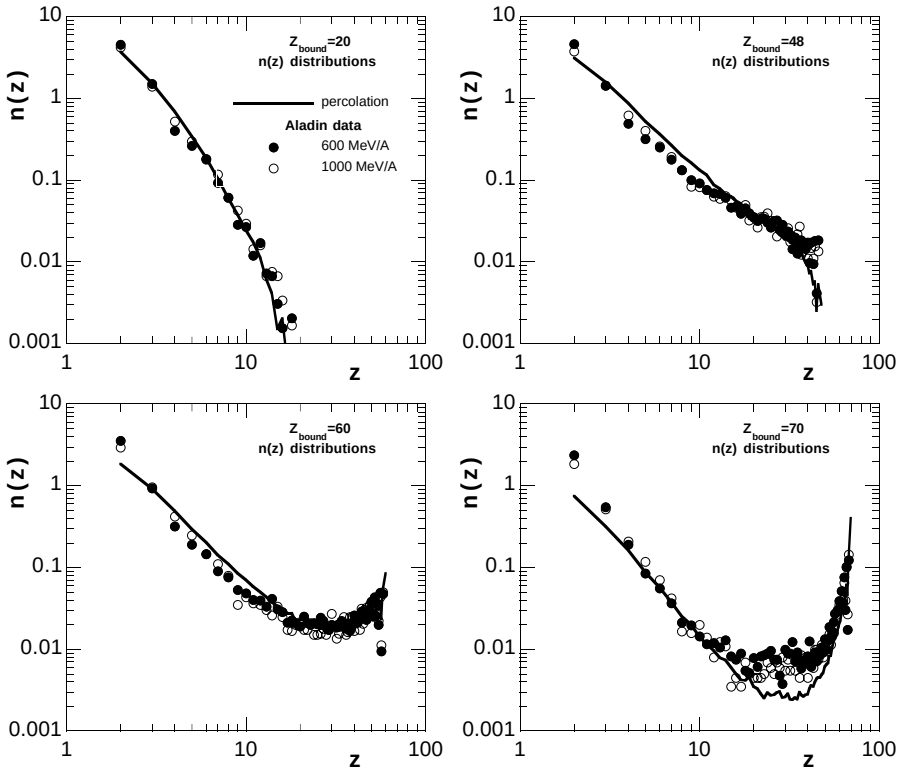

Fig. 1. Experimental fragment size distributions $n(z)$ at four values of the control parameter $Z_{\rm bound}$ (see text). The lines correspond to the percolation calculation. A log-log representation has been chosen to emphasize the power law behaviour at $Z_{\rm bound} = 48$.

(the identification of this mechanism), we discuss the analysis of a fragmentation experiments performed with atomic nuclei.

2.1 The experiment

The experiment was performed by the *Aladin* collaboration at *Gesellschaft für Schwerionenschung* (G.S.I.), Darmstadt [13]. Beams of gold ions (Au, $Z = 79$) at 600 and 1000 MeV/nucleon incident energies were used to bombard targets made of thin copper foils. Data for more than 3×10^5 events were collected. At these high energies[2], the commonly admitted scenario of the collision is the following: the part of the projectile that does not geometrically overlap with the target at the point of closest approach is thought to decouple from the rest and form a sub-system called the *projectile spectator* (PS). The size of this system and its excitation energy E^* are therefore dependent on the impact parameter. The overlapping part of the system is completely vaporized and therefore does not contribute to the formation of fragments with nuclear charge (z) greater than one.

In the present experiment, the *Aladin* device detected with very high efficiency all spectator fragments (those resulting from the decay of the unstable PS) with nuclear charge $z > 1$. Neither hydrogen isotopes nor neutrons were detected, event by event, with any significant efficiency. The initial size of the spectator system ($Z_{\rm PS}$) is not experimentally measured and can only be inferred from the comparison with a nuclear reaction model. Empirically, this size can be estimated, on average, from the following relation [14]:

$$\langle Z_{\rm PS} \rangle = 25 + Z_{\rm bound} - 0.004 Z_{\rm bound}^2 \qquad (1)$$

where $Z_{\rm bound}$ is equal to the sum of all the charges of products with $z \geq 2$. In all the studies of these experiments, the parameter $Z_{\rm bound}$ is used as control parameter. Notice that $Z_{\rm bound}$, which is closely related to the impact parameter, decreases when the violence of the collision increases.

2.2 Analysis of data

To analyze the *Aladin* data, the following very simple procedure has been used. For a given value of $Z_{\rm bound}$, the size of the PS was deduced from equation (1) and percolation calculations on a lattice of corresponding size were performed by varying the bond breaking parameter (random-bond percolation). Only those events with the proper $Z_{\rm bound}$ were kept and the fragment size distribution of these compared to the corresponding experimental data. In this manner the comparison is parameter free. Notice that by fixing a value of $Z_{\rm bound}$, the maximum value $z_{\rm max}$ of z is also constrained by this value.

Figure 1 shows, for $Z_{\rm bound}$ values of 20, 48, 60 and 70, the experimental fragment size distributions. One observes three different regimes of fragmentation. For large $Z_{\rm bound}$, large impact parameter and low excitation energy, only a heavy residue and light particles are produced. This is the "evaporation" regime. For small $Z_{\rm bound}$ (most violent collisions) only small fragments are produced and

[2] High energies compared with the 8 MeV/nucleon binding energies or the 35 MeV/nucleon Fermi energies of nuclei.

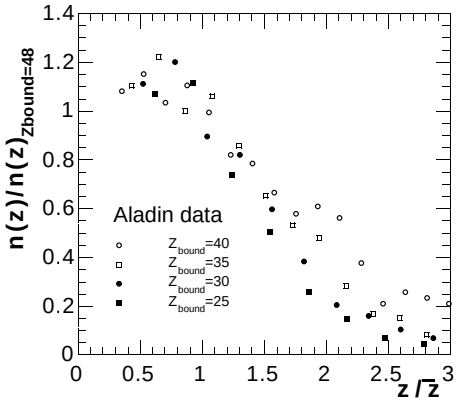

Fig. 2. Test of the scaling assumption (Eq. (2)), that requires that all the ratios $n(z)/n_c(z)$ follow the same curve.

no heavy residue is left. The distribution of this "vaporization" regime decreases exponentially. Remarkably, at $Z_{\rm bound}$ around 48 an intermediate regime exists, for which the distribution can be fitted by a power law $n(z) \sim z^{-2.2}$. The mean excitation energies *per particle* can be estimated for the fragmenting systems as a function of $Z_{\rm bound}$ [14]: they correspond to about $\langle E^* \rangle \simeq 14, 6, 3, 1$ MeV per particle for $Z_{\rm bound} = 20, 48, 60, 70$ respectively.

The full lines in Figure 1 represent the corresponding distributions obtained from the percolation calculation. The calculation reproduces quantitatively, and over several orders of magnitude, the data in the three regimes[3]. We are naturally led to associate the experimental power law behaviour at $Z_{\rm bound} \simeq 48$ to the percolation critical behaviour.

Close to the critical regime, percolation theory predicts typical scaling properties of the cluster size distributions [15]. The size distributions can be reduced to an universal function, $f(z/\bar{z})$, by the following formula:

$$n(z) = n_{\rm c}(z) f(\frac{z}{\bar{z}}) \tag{2}$$

where $n_c(z)$ is the size distribution at the critical point, taken here as the size distribution observed for $Z_{\rm bound} = 48$. The "characteristic size" $\bar{z}$ is defined by:

$$\bar{z} = m_z/m_2 \tag{3}$$

with $m_k = \sum_{z \geq 2} z^k n'(z)$, where $n'(z)$ is the mean fragment size distribution obtained excluding by event, the largest fragment. One observes in Figure 2 that, within the scattering of the data, this rule is rather well satisfied, keeping in mind that this corresponds to the ratio of two quantities varying over more than three orders of magnitude. Similar study has been performed on Au + C data at 1 AGeV [16].

The size of the largest fragment plays in percolation theory the role of the order parameter. For an infinite system, it is of infinite size in the percolating phase and finite in the non-percolating one. In a finite system, this transition is smooth, as illustrated in Figure 3. This figure on the left compares, again as a function of $Z_{\rm bound}$, the experimental measured size of the heaviest fragment $z_{\rm max}$ to the one obtained from the percolation calculations. The figure on the right shows the fluctuations of $z_{\rm max}$,

$$\sigma^2_{z\rm max} = \frac{\langle z^2_{\rm max} \rangle - \langle z_{\rm max} \rangle^2}{\langle z_{\rm max} \rangle} \,. \tag{4}$$

As expected [17], these functions show a maximum around the "critical" value of $Z_{\rm bound}$. In Figure 4, the full distributions of $P(z_{\rm max})$ are shown and compared to the calculation. As recommended by the *Aladin* collaboration, data for $Z_{\rm bound} > 70$ have not been considered for this analysis as they are contaminated by experimental triggering problems [18].

By any standard, the agreement observed in Figures 1 to 4, between the experiment and the calculations is very good. In the analysis presented here, as already stressed, no adjustable parameters are used.

3 Interpretation of the results

The choice of percolation theory to analyze the experimental results is motivated by the following reasoning. A fully microscopic description of nuclear fragmentation is, *a priori*, out of scope of theory. Atomic nuclei behave in their ground state as small drops of Fermi liquids composed by particles strongly interacting mainly with a two-body, short range force. This interaction is ill defined at short distances and the techniques used to solve this complicated many body problem are not fully under control. The description of collisions, using transport equations for example, is even more difficult. In view of the above remarks and with the stated goal of understanding the universal features of the data, we believe that a more fruitful point of view is to tackle the problem with the minimum number of assumptions.

The simplest hypothesis would be to assume the equiprobability of all partitions of the integer number $Z = 79$. This is equivalent to a "maximum entropy principle" [19,20]. However, the resulting $n(z)$ are always exponentially decaying functions, in contradiction with experiments.

A step further is to consider topological constraints, by considering fragments in a 3-dimensional space. Among the infinity of models one can imagine to make fragments, random-bond percolation seems a good candidate because despite its simplicity it retains the essential constraints. For example, the shape of the fragments is not assumed *a priori*. It turns out that, as shown in Section 2, it suffices to reproduce very well the experimental data. How does one understand this agreement? One can imagine at least two scenarios.

In the first scenario, one idealizes the nuclear fluid as an ensemble of particles connected by bonds. The bond between a pair of particles is active as long as the magnitude

[3] A close inspection of the data for $Z_{\rm bound} = 70$ shows an excess of fragments produced around $z = 30$ due to the presence of fission events. This is understandably beyond the scope of percolation theory.

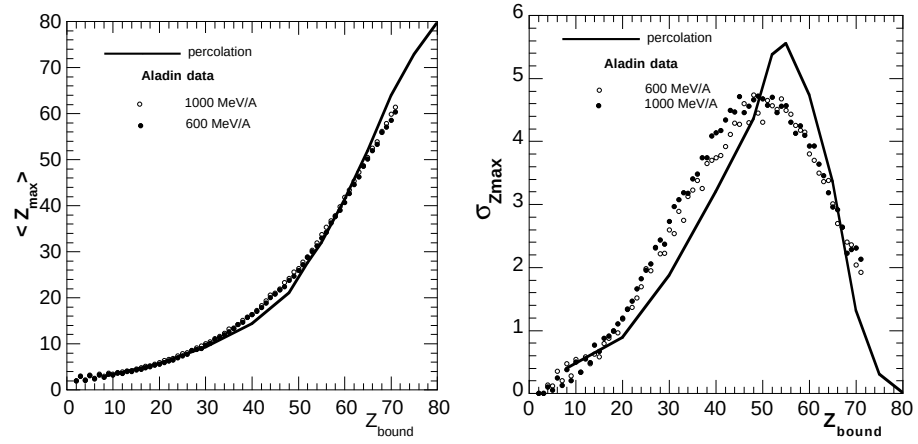

Fig. 3. Mean value of $z_{\max}$ as function of $Z_{\rm bound}$ for percolation calculation and experimental data from reference [13] (left). Fluctuations of $z_{\max}$ as function of $Z_{\rm bound}$ for percolation calculation and experimental data from reference [13] (right).

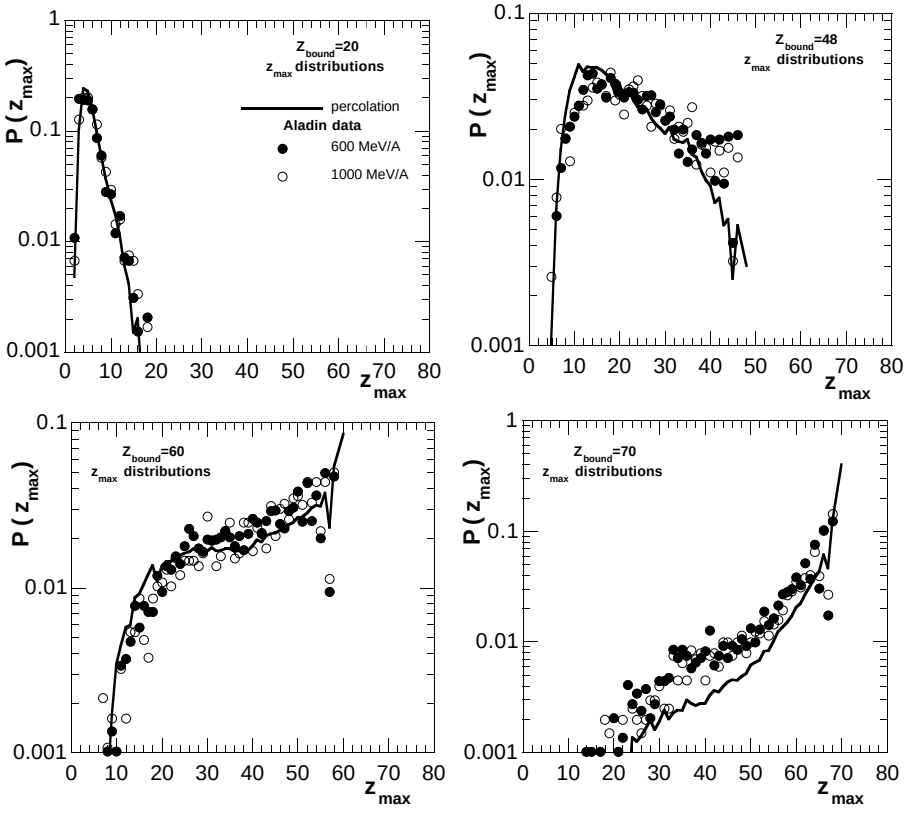

Fig. 4. The distribution $P(z_{\max})$ of the largest fragment at four values of $Z_{\rm bound}$. The circles correspond to the data from reference [13] and the lines to the percolation calculation.

of their potential energy is greater than the relative kinetic energy. During the collision some of these bonds break because of the change in the position and/or the velocity of the particles. The simplest assumption is that bonds are broken randomly, which corresponds to the usual uncorrelated bond percolation model [15]. Particles connected by unbroken bonds form the fragments. These fragments separate rapidly, pushed away by the long range Coulomb force between protons.

In a second scenario, one assumes that after the collision phase, equilibrium thermodynamics applies: the system expands until a "freezout" density is reached, at which the fragments cease to interact by the strong nuclear attractive force and their size distribution is "frozen". Then, the Coulomb force accelerates the fragments, as before. In order to calculate its distribution, we have to define first what a "fragment" is. In the present context, it seems natural to call "fragment" a self-bound ensemble of particles [21,22][4]. With these definitions, one finds in the $\rho - T$ diagram a percolation line (sometimes called the Kertész line [26]) that separates a percolating and a non-percolating phases. The line joins the thermodynamical critical point to the random bond percolation critical

[4] Another possibility is to impose the stability by monomer evaporation only [23]. At the present level of accuracy, both definitions are operationally very similar. Definition [23] is equivalent to the ones proposed by Hill [24] and by Coniglio and Klein [25].

point. On it the fragment size distribution is a power law $n(z) \sim z^{-\tau}$, with $\tau \simeq 2.2$ (see Figs. 1 and 2 of Ref. [23]). In a small system like an atomic nucleus, rather than a sharp critical line, one finds a "critical zone" on which the $n(z)$ approaches this behaviour (see Fig. 4 of Ref. [27]). The agreement of these calculated $n(z)$ with the experimental ones shown in Figure 1 is also very good. Therefore, by inspection of the $n(z)$ alone, it is not possible to disentangle between these two scenarios. However, one could hope that extra information on, for example the fragment kinetic energies, could indicate if thermalization is present or not.

More generally, the main difficulty in analyzing nuclear fragmentation data is due to the very small size of the system, which fundamentally limits the extraction of the universal critical exponents. Indeed, a proper characterization of the physical process requires, apart from the τ exponent, the determination of the other critical exponents [15] associated with the moments m_k of the fragment size distribution. Such measurements are, in principle, possible for simple fluid systems (*i.e.* made of structureless particles subject to short range forces) of larger sizes such as atomic aggregates or macroscopic pieces of matter, such as liquid drops [12].

We consider as very plausible the possibility to observe this *percolation type* fragmentation in simple fluids. Indeed, the arguments that we have developed to explain the success of percolation theory should apply without restriction to any system of structureless particles interacting with a short range potential. In fact, attempts have been made to show experimentally this behaviour in the fragmentation of hydrogen aggregates [11]. In these experiments, the multiplicity of fragments m_0 is used as control parameter. As a function of m_0 the mass of the largest fragment and its fluctuation, evolve qualitatively as expected in percolation theory. However, in reference [11], the data are compared with a percolation system of improper size and no definite conclusions can be drawn.

Other experiments on the fragmentation of noble metals have been performed, for example the fragmentation of gold aggregates by high energy collisions with xenon ions [28] or the production of copper and silver clusters in sputtering sources [29]. In both cases one observes fragment size distributions of power law type (with odd-even and shell effects superposed on it) with a decay exponent compatible with the prediction of percolation theory. Unfortunately this is not conclusive because no selection was made on the different classes of fragmentation events.

On the theoretical side, we are performing large scale classical molecular dynamics simulations of a Lennard-Jones fluid, for both the sudden disassembly of an equilibrated system and for the collisions of drops. We clearly find in these calculations the fragment size distributions predicted by percolation theory [30].

In fact, a close examination of earlier calculations of the fragmentation of *equilibrated* Lennard-Jones droplets shows qualitatively this percolation type behavior. For example, Pandharipande and collaborators considered the fragmentation of equilibrated droplets [31,32] made of a few tens to a few hundreds of particles. The parameters of the potential are adjusted to describe the interaction between argon atoms. One clearly sees in Figures 10-15 of reference [32] an evolution of the $n(s)$ of percolation type. The same behaviours are found in references [21,22,33]. Pandharipande *et al.* also considered the fragmentation of colliding droplets [32] and showed (by looking at the velocity distributions of the fragments and at their size distributions) that it is possible to describe this process as the fragmentation of a single equilibrated system of the same total mass and of excitation energy corresponding to the center of mass collision energy. The best equilibrated systems are obtained with the collision of two nearly equal mass droplets. It is also important to notice that the above mentioned fragment size distributions are the *asymptotic* ones, *i.e.* calculated once the fragments have evacuated their internal excitation energy by particle evaporation. These asymptotic fragments are cold, otherwise they would not be stable by particle evaporation.

These remarks are interesting, in view of the possible experimental observation of the percolation line in collisions between atomic clusters of noble atoms. For these clusters, at the high excitation energies considered here, the classical molecular dynamics with a Lennard-Jones potential is a reliable guide to define the optimal experimental conditions.

Some other calculations of the fragmentation of Lennard-Jones droplets seem to contradict our claim on the existence of a percolation line in the supercritical phase. The reasons of this apparent discrepancy are of various orders. (a) The calculations do not explore the correct points of the phase diagram, *i.e.* close enough to the Kertész line [34–36]. (b) The initial conditions are extremely out of equilibrium [37] and do not correspond to a realistic physical situation (see the above remark concerning the collision of argon droplets). (c) Clusters are simply defined as ensembles of particles close enough in space [36,37], without any energetic consideration. Serious errors are then introduced if the analysis of the cluster distribution is made at a too earlier stage during the expansion of the system. (d) Artificial boundary conditions are introduced, evolving with the expansion of the system and allowing for a partial re-equilibration [35,36].

4 Final remarks

Other experiments of nuclear fragmentation at high bombarding energies have been successfully interpreted with percolation theory [38–41]. However, at lower bombarding energies (30–50 MeV/nucleon) the agreement is less satisfactory [42,43]. The shape of the $n(z)$ evolves qualitatively as in Figure 1, but a closer examination shows a systematic over-production of intermediate size fragments. The origin of these discrepancies is still unclear. Different explanations can be considered:

(a) the identification of the source of the fragments is more difficult at lower collisionnal energies where

the separation between the projectile, the target, or the fused system is not as clear as in the case of high energy collisions, leading thus to possible contaminations between these different sub-systems;

(b) different fragmentation mechanisms could result from the smaller relative velocities between projectile and target, particularly in the case of fusion at small impact parameter, inducing possible compression effects.

These discrepancies, more generally, could just signal the fact that the assumption that fragmenting nuclei behave as "simple fluids" breaks down at these lower incident energies.

Nuclear fragmentation experiments are often analyzed with the so-called Statistical Multifragmentation Models [44,45]. In brief, these models deal with the equilibrium thermodynamics of ensembles of *spherical* drops of *nuclear matter* confined in a "freezout" volume. Drops interact with each other only by the long range Coulomb force and their internal partition function is taken from empirical mass formulas or from experiment. These models[5] are successful in describing the above mentioned low bombarding energy experiments when fixing the size, the density and the excitation energy of the fragmenting source. For the high energy *Aladin* data similar analysis have been performed but with a larger set of input parameters [39].

The use of percolation theory concepts is however more comprehensive while much easier to handle. It provides an excellent agreement with the data without requiring any adjustable parameter.

We hope that the present results will encourage both theoretical and experimental studies of the fragmentation of simple fluids. The fragmentation by collisions of very large aggregates seems particularly promising, because it combines the experimental possibility to detect fragments together with a reduction of finite size and surface corrections effects. Systems that could be studied could range from hydrogen aggregates (as in [11]) to those of noble metals. In view of the analysis that we proposed, the data of *each* fragmentation event should be characterized by the size of the individual fragments and possibly by their individual kinetic energy. The total excitation energy of the system should range from the regime of evaporation of monomers to the regime of total vaporization. This is certainly a difficult but rewarding challenge.

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[5] These models have been also implemented to study the fragmentation of atomic clusters [46].