Energy relaxation and vibrations in small 3D plasma clusters

T Antonova\textsuperscript{1}, B M Annaratone\textsuperscript{2}, H M Thomas\textsuperscript{1} and G E Morfill\textsuperscript{1}
\textsuperscript{1} Max-Planck-Institut für Extraterrestrische Physik, Garching, D-85741, Germany
\textsuperscript{2} UMR6633 CNRS-Laboratory PIIM, Centre de Saint Jerome case 321, University of Provence, PO BOX 321, Marseille F-13397, France
E-mail: antonova@mpe.mpg.de

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Abstract. The time evolution of three-dimensional (3D) plasma clusters containing 17 and 63 particles has been analyzed. Using a radiofrequency (rf) spot electrode, we were able to get almost un-stressed 3D clusters under gravity conditions on Earth. Fast 3D diagnostics of the particle positions allowed us to study the cluster structure and dynamics in detail. In particular, we were able to follow the evolution of the systems through rearrangement and particle evaporation to their final equilibrium state with minimum energy. The vibrations of the larger (63 particles) cluster were compatible with theoretical estimates for a liquid drop with surface tension. This indicates that macroscopic properties, normally associated with systems in the cooperative regime, provide an adequate description even for small (discrete) clusters.
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Small particles injected in laboratory plasmas can form structures resembling gaseous, fluid and solid like assemblies, which react to discharge conditions and the force of gravity. These plasmas are known as dusty or complex plasmas. The unique properties of these systems (relative simplicity of observing and controlling the parameters, self-organization and formation of ordered structures) make dusty plasmas very attractive systems in which to visualize multibody microscopic effects [1, 2]. The particle equilibrium positions inside the discharge are determined mainly by the electric potential distribution in the chamber and gravity (plus some smaller forces). Usually in laboratory experiments the electric field is strong enough to compensate for the gravitational force only in the plasma sheath region near the electrodes (unless the particles are very small, tenths of microns). In this region good ordered crystalline structures often form. The influence of gravity stresses these structures, relatively stress-free three-dimensional (3D) assemblies are obtained only under microgravity conditions [3].

Applying strong external confinement [4] or by using the specific shape of the discharge with a ‘secondary plasma’ inside the main plasma sheath [5], it was possible to build unstrained 3D plasma clusters in gravity. It is generally accepted that for small clusters the states of the matter: solid, liquid, etc lose their typical definitions because the equilibrium distance between particles is locally modified by the geometry. Phase transitions are then distributed on a large range of thermal energies. However, in such assemblies the particles can be strongly coupled; we can get perfectly ‘frozen’ clusters at high gas pressure and electrical power, similar and often identical to nanostructured solid state systems or ion traps. This is due to geometrical constraints and the existence of an equilibrium distance. However, in order to study the characteristics of individual interactions as well as the properties of the whole structure (as a macro-system) we have chosen a regime in which the particles are still somehow mobile. This allows us to follow the evolution of the system in search of the minimum energy. From this point of view the observed vibrations and the displacement of the particles inside the clusters can be used (at least in principle) to discover new features of 3D nanoclusters generally.

In this work, we present an analysis of 3D plasma cluster behavior and study their properties. The clusters have been obtained in a discharge produced by the so-called
‘adaptive electrode’ [6]. In previous work [7] the interaction forces among particles observed in a small four particle cluster were quantitatively derived from analysis of the particles’ spontaneous motion. It was found that the interaction force has an attractive part. The physical mechanism responsible for the attraction can be dipole–dipole interaction, the compatibility has been shown in [8], or ion and neutral shadow forces (to be published), or even a mixture of these two effects. Whatever the physical reason, the measured attractive force was stronger than the surrounding confining electric field. The clusters described in the present paper have been obtained in the same way as the clusters studied in [7], so the conditions are the same and the external confinement has a negligible influence on the processes inside the clusters discussed below (collective effects may generate a self confinement though). This makes it possible for the first time to conduct high-accuracy kinetic studies of generic importance.

Clusters with 17 and 63 particles have been studied in detail. In the larger cluster particle rearrangements due to ejected particles and characteristic oscillations have been observed. In the smaller cluster particle restructuring occurred after the injection of a fast particle. In this cluster vibrations are not strong and do not show characteristic frequencies.

1. Experiments and 3D diagnostics

In our experiments an argon plasma was generated by radiofrequency excitation, 13.56 MHz, of the upper of two parallel plate electrodes in the so-called PKE–Nefedov chamber [3], which has glass walls. The lower segmented ‘adaptive electrode’ is divided into 57 small pixels that can be independently driven in dc and rf (13.56 MHz) [6]. Spherical clusters have been obtained by applying an rf voltage to a single pixel driven in opposite phase with respect to the rf voltage on the upper electrode. The other 56 pixels of the electrode are kept floating at the local self bias, slightly negative w.r.t. ground. When the dc voltage of the chosen segment is kept grounded through an inductance, a region of enhanced ionization (∼32 mm³) appears in the sheath above the pixel. Hence, in the usual rf discharge we have an additional ‘secondary plasma’ in the sheath of the main plasma.

When the pixel is ‘off’ the injected melamine–formaldehyde particles of 3.4 µm diameter levitate in the plasma sheath. As soon as the ‘secondary plasma’ glow appears, particles are collected from the dust cloud in the sheath to this small glow where they form near-spherical clusters [5]. Particle distribution in the plasma chamber then look as follows: the dust cloud in the main plasma sheath above the ‘secondary plasma’ and particle cluster inside ‘secondary plasma’ glow (see [5], figure 20). Yet, particles from the above cloud can be collected one by one to the clusters by varying the amplitude of rf voltage applied to the pixel. This allows one to form and study different dust structures. Particles in such a small plasma volume are visualized by a system of three lasers of different wavelength. This is a 3D optical system, described in detail in [5], which together with programming analysis allows us to simultaneously measure the three coordinates of the particles in clusters with less than 100 particles. The particles’ velocity vectors are also measured in 3D, from the analysis of traces recorded during the CCD shutter opening time, 36 ms [7]. From the digital image sequences which contain 200 frames (8 s, 25 frames per second) all particle coordinates are derived. In the programming analysis each particle image is fitted by a Gauss function, which gives the coordinates and intensities for all sequences. Then images are correlated in y in order to get x, y and z for each particle in each frame. The experimental errors in the determination of particle positions are 3 µm in the x- and z-directions and 21 µm in the y-direction. By this technique fast processes in the clusters can be
detected. This possibility is indispensable in 3D cluster studies, because one can extract some information about the system only by observing ‘fast’ events (e.g. at the Einstein frequency).

2. 17 particle experiment: changing of the system energy after the arrival of a new particle

One recorded sequence shows the dynamical evolution of the cluster with 17 particles. On the first frame of the sequence one additional particle arrives vertically from the above dust cloud to an already formed cluster of 16 particles. This gives us the opportunity to observe the dynamics of a cluster which is adapting to a new state with an additional particle. This cluster was formed at 76 Pa (200 V_{p-p} on the upper rf driven electrode and 110 V_{p-p} on one pixel of the ‘adaptive electrode’). The incoming particle was injected into this cluster with a velocity of at least 12 mm s^{-1} (the velocity has been calculated from the trace left, but the beginning of this trace is beyond the frame). This particle penetrates the cluster, pushing and re-arranging the neighboring particles. The total kinetic energy imparted to the cluster is >2.4 \times 10^{-18} J. We were able to follow the dynamical evolution of the excited cluster to its new equilibrium. This turned out to be, in part, a collective sharing and dissipation process (the energy is redistributed, e.g. by phonons). Figure 1 shows the first and second frames overlapped. It is seen that after the new particle arrives in the second frame all particles as a whole shift their positions in the \( y \)-direction in comparison to the first frame. In the following frames, we observed a collisional
cascade effect, where a particle, displaced from its equilibrium position by the collision, moves inside the cluster, collides with and rearranges other particles, until, after 1.12 s, it comes again to the external surface of the cluster having lost the kinetic energy acquired in the collision (figure 2). Thus, this particle tries to find a stable position in an already formed structure, pushing the nearest neighbors which, in turn, push their neighbors.

3. Model

The rearrangement inside the structure can be explained by the motion of the particle to find an energetically preferable position. This provides a minimum energy for the whole system. In small clusters the bonds between particles are stretched/compressed by the geometrical constraints, making the energy per bond higher w.r.t. the energy of the same bond in an infinite crystal. Moreover, in the transient analyzed, the new particle moves among interstitial metastable states, while the other particles re-adjust their equilibrium positions, until a more stable configuration is reached. The additional potential energy due to stretched/compressed bonds can be calculated linearizing the interaction force for small displacement around the equilibrium positions: the additional potential energy due to the stretching/compression of the
Figure 3. The structure of shells in the clusters of 17 particles. (a) First frame of the sequence before additional particle arrives, (b) frame in the middle of the sequence after rearrangement of the cluster caused by the arrival of the new particle.

$n$ bonds in the $m$th frame can be defined as:

$$E(m) = \sum_n \frac{1}{2} K (d_{n,m} - d_{0,m})^2 + \text{Const}(m),$$

where $d_{n,m}$ is the distance between each first neighbors particle pair and $d_{0,m}$ is the averaged distance for all pairs in frame $m$. However $d_{0,m}$ varies from frame to frame and it is different from the equilibrium distance of the binary interaction $d_0$. For comparison, this equilibrium distance measured in [7] for the same particle size and the same Debye length was $d_0 = 175 \mu m$. The difference between $d_{0,m}$ and $d_0$ defines the constant in equation (1) that can be rewritten as:

$$\text{Const}(m) = +\frac{1}{2} K (d_{0,m} - d_0)^2. \quad (2)$$

There are two methods to estimate $d_0$: (i) we have minimized the energy of frames in which the structure is stable enough (no dramatic reorganizations are observed). The result gives $d_0 = 200 \mu m$. (ii) The second way is to make averages of the displacements around equilibrium (most of the bonds are in the linear range), this gives a very near result: $d_0 = 210 \mu m$. A further validation comes from the observation that the above values correspond also at the shell-dimensional separation as given in figure 3. As the interaction force tends rapidly to zero with increasing distance in our schematic model we introduce a cut-off distance so that equation (1) holds for $0 < d < d_{\text{cut-off}}$, for higher separations the interaction energy is zero. This cut-off distance can be estimated using the behavior of the interaction force [7] shifted to take into account the variation of the equilibrium distance (from 175 to 200 μm); we have chosen the value of 0.25 mm. This value also excludes interaction with the second neighbors, see figure 4. We also use the same force gradient near the equilibrium position as in [7]: $K = 1.5 \times 10^{-9} \text{N m}^{-1}$. An error in this latter value will result in a multiplicative constant that prevents us from observing qualitative variations of energy. The energy of all stretched/compressed bonds in the system is shown in figure 5(a) for 54 frames (2.16 s), which contain the rearrangement and transition to the equilibrium. The energy after the rearrangement (24th frame) is clearly lower. The peaks correspond to the frames in which larger changes of the displaced particle’s position were observed. We conclude, that after the arrival of one new particle, the system goes through a collisional transition state to a final state with minimum energy.

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Figure 4. The structure of shells in the larger cluster after three particles left, forming a 60 particle cluster.

Figure 5. The derived energies of the 17 (a) and 63 (b) particle clusters in time. (a) A fast particle arrives at frame 1 and pushes a neighboring one, which moves inside the cluster during frames 8–10, stays there (frames 11–20) and comes to the cluster top (frames 21–24), full timescale 2.16 s, energy is averaged in 5 points. (b) Particles leave at frames 28, 63 and 87, full timescale 4.08 s, energy is averaged in seven points. Time between frames is 40 ms.

We can explain the transition by interpreting the structure observed in the states with ‘magic numbers’. Clusters in these states are stable and have less energy per bond than they would have with a smaller or a larger number of particles. In principle the magic numbers depend on the nature of the interaction force; however, there are common features when the force is linear (or linearizable) around an equilibrium distance, due to attractive forces or to
external confinement counteracting Coulomb repulsion. In particular for small clusters, up to 20 particles, numerical simulations based on the minimization of the interaction energy show that the magic numbers coincide; to give an example, in [9] systems based on the Gupta potential and Murrell–Mottram potential give the same magic numbers as those derived by geometrical consideration. A 16 particle cluster can be seen as a 13 particle nucleus, with three particles added as ‘defects’. A cluster of 13 particles can be found in two structures: fcc and icosahedron [10]. The former has six first neighbors at 1(a) (a-equilibrium distance) and eight second neighbors at 1.71(a) while the latter has ten first neighbors at 1(a) and two second neighbors at 1.05(a) (with 5% stretch of the bond). In our case the original state shows one clear peak of 7 particles (magic number) in the first shell, and a second shell broadened by thermal motion, figure 3(a). This state becomes metastable with the arrival of the 17th particle and after 1.5 s we observe the transition to the icosahedral structure, clearly visible in figure 3(b). It is of interest to compare this result with the equilibrium configurations obtained by Ludwig et al for confined Coulomb clusters [11]. These simulations are based on particles of equal charge in an harmonic trap. According to this paper the probability of finding a 17 particle cluster empty (without the particle in the center) is not much different from the probability of finding the 16 particle cluster empty; the filled states are however preferable for both 16 and 17 particles. In these terms, we conclude that, for our system, the transition is caused by the reshuffling of the system due to the added energy of the collision that effectively induces a further minimization of the energy. The evolution of the energy during the transition measured in our experiment offers a model for the same transition observed in the formation of metallic nanoparticles [12] and plays a role in the melting of clusters [13].

4. 63 particle experiment: changing of the system energy after ‘evaporation’ of the particles

Three particles ‘evaporated’ from the top of the larger 63 particle cluster (57 Pa pressure, 300 $V_{p-p}$ on the upper rf driven electrode and 120 $V_{p-p}$ on the pixel) without any external manipulation during the recorded sequence (figure 6), leaving the cluster in the final structure, figure 4. Variations in the energy of the cluster were again calculated for all bonds using equation (1) with $d_0$, the equilibrium displacement that minimizes the energy in stable frames. Figure 5(b) shows the average energy of the cluster during 100 frames (4 s) which contain the evaporation of three particles and demonstrate the evolution of the system from the 63 to 60 particle state. After the first two particles escape (frames 28 and 63) the energy rapidly increases (peaks at frames 47 and 76) demonstrating an energetically unfavorable state. Presumably, the gain in energy is due to conversion of kinetic to potential energy, as the cluster initially rearranges. There is no increase in energy after the last particle has left (frame 87). The stable trend after frame 87 corresponds to the vibrations of the upper particle in the cluster, which however does not evaporate.

The results of [11] show that 57 and 60 are ‘magic’ numbers in 3D configurations both corresponding to the closure of the second shell, 60 is a lower number corresponding to two shells. Furthermore, the escape of the particle at the top of the cluster suggests the configuration of an electric field, which is overall weak in the plasma region of the cluster, and stronger at the position where particles evaporated to the above main plasma sheath. This supports the conception of weak external confinement for our clusters, which has been postulated in [7].
5. Analysis of vibrations

5.1. Dust acoustic waves

After the last particle has left the larger cluster the remaining upper particles exhibit large amplitude vibrations. In the lower part of the cluster the vibrations are less pronounced probably because of the influence of the electrode vicinity. A detailed analysis of the vibrations showed that the three particles which evaporated did so during such self-oscillations, which are mostly vertical. We have obtained the natural frequency of the system by Fourier transforming the vertical position of the particles. Figure 7 presents several modes of the cluster frequency, $f$ (the peak near 0 Hz in figure 7 is due to the transform technique). In the following these oscillations are compared to three models, dust acoustic waves, dust lattice waves and hydrodynamical self-mode of oscillation.

Smirnov [10] interprets the vibrations of particles as acoustic waves with a sound speed, which propagate inside clusters. As a first attempt the oscillations experimentally measured have been compared to dust acoustic waves. The contribution of the finite dimension and the spherical geometry of the cluster could be taken into account working in analogy to [14]. In [14], the authors analyzed the cylindrical geometry for applications to waveguides. They found that the dust acoustic frequency did shift to lower values but the effect is small for $R \sim \lambda_D$. For this kind of waves inertia of dust is very important and electrons and ions follow a Boltzmann distribution [15]. The Poisson equation has been considered together with the dust continuity
equation and dust momentum equation. In the case of the long-wavelength limit, \(k^2 \lambda_D^2 \ll 1\), where \(k = 1/D\), \(D\) being the dimension of the cluster, \(\lambda_D\) is combined Debye length, the wavefrequency can be found as:

\[
\omega = kZd_0 \left( \frac{n_{d0}}{n_{i0}} \right)^{1/2} \left( \frac{k_BT_i}{m_d} \right)^{1/2} \left[ 1 + \frac{T_i}{T_e} \left( 1 - \frac{Z_{d0}n_{d0}}{n_{i0}} \right) \right]^{-1/2},
\]

(3)

where \(m_d\) is the dust particle mass, \(n_{i0}\) and \(n_{d0}\) are the densities of the ions and dust, respectively, and \(Z_{d0}\) is the charge of particles. The value of this latter quantity deserves attention because our cluster is located inside a plasma that has been modified by the presence of the cluster itself, i.e. the plasma near the cluster border differs from the plasma far away. Since electrons and ions are absorbed by the solid spheres a space charge forms near the cluster. Matching the flux of particles in and out of the cluster, with zero net current, will create double layers, as in [16]; in that paper, at the same pressure and particle size as in this paper, for planar geometry, the double layer voltage was \(1.007 kT_e e^{-1}\). In our case the effect is reduced by the focusing of the ions near the cluster border due to the spherical shape, as for spherical Langmuir probes. In our slightly collisional regime the radial motion theory for the ion motion gives an upper estimate for the floating potential. For \(R/\lambda \sim D \simeq 1\), the effect of the geometry implies a reduction of 33% with respect to the planar geometry in argon [16]. Thus, we have a difference in the potential between the plasma surrounding the clusters and the plasma ‘far away’ equal to \(1.007(1 - 0.33) = 0.66 kT_e e^{-1}\). Inside the quasineutral cluster the particles have a charge strongly reduced with respect to the charge calculated using the vacuum approximation and the particle floating potential from the radial theory for electric probes [17]. In our collisionality regime (200 \(\mu m\) the distance between particles and the mean free path is 30 \(\mu m\)) we assume the charge \(z_d = 2500e\), somewhat lower than that estimated in [7], because of the higher pressure, [18] and the larger number of nearby particles in the present case. \(n_{d0} = 2.35 \times 10^{11} m^{-3}\) is the dust density in our cluster of 63 particles. The plasma density \(n_{i0}, n_{e0}\) in the ‘small plasma’ volume has been estimated from the spectroscopic analysis to be of the order of \(10^{16} m^{-3}\), higher than the main plasma density. Ions are considered to have room temperature \((T_i = 0.025 eV)\), the temperature of electrons is supposed to be about 5 eV (some how higher than in the main plasma). At these parameters the estimated angular frequency of

**Figure 7.** The Fourier transform of the vertical vibrations of the 63 particle cluster.
dust acoustic waves $\omega$ is 2.5 Hz, which is in agreement with the first peak of experimental frequency $f = 1.3$ Hz.

5.2. Dust lattice waves

Since the investigations performed have shown that our cluster is strongly coupled, the second attempt to understand the cluster vibrations is comparing with dust lattice waves. The frequency of longitudinal dust lattice waves is expressed as follows [15]:

$$\omega^2 = \frac{2Z_D^2}{m_d a^3} \left( 1 + \frac{a}{\lambda_D} \right) \exp \left( -\frac{a}{\lambda_D} \right),$$

with $m_d = 3.1 \times 10^{-14}$ kg, $a$, the interparticle distance, is 200 $\mu$m and $\lambda_D$ is the linearized Debye length $1/\lambda_D^2 = 1/\lambda_{D1}^2 + 1/\lambda_{De}^2$ giving $\lambda_D = 30$ $\mu$m. For our parameters the frequency $f = 13$ Hz, while the experimental data give $f = 1.3$ Hz. However such discrepancy with the experimental data may still be explained by very small cluster size and border effects. Unfortunately no theoretical analysis of the effect of the geometry is available in the literature to the authors’ knowledge.

5.3. Hydrodynamic approach

A third possibility to understand the natural global modes in the case of strongly coupled systems is the hydrodynamic approach. The cluster has the shape of an oblate sphere, therefore the frequency of the system has been compared with the resonance mode frequencies of a liquid drop with surface tension. According to [19] the self-frequency of oscillations of a spherical drop is given by the following equation:

$$\omega^2 = \frac{T l(l - 1)(l + 2)}{\rho R^3}.$$  

In the case of a cluster $R = 0.4$ mm is the cluster radius, $T$ is surface tension, $\rho$ is the density of the particles in the cluster, $\rho = 63\text{mass}_{\text{part}}/\text{Volume}_{\text{cluster}}$. $l$ takes positive integer values; the value $l = 0$ corresponds to radial oscillations, i.e. to spherically symmetric pulsations of the drop, which we do not observe. For $l = 1$ the motion is simply a translation. The smallest possible frequency of oscillations of incompressible drops corresponds to $l = 2$ (elongation–oblateness) and is expressed by $\omega_{\text{min}} = \sqrt{8T/\rho R^3}$. Since the oscillations are more obvious in the upper part of the cluster one can compare the cluster with a liquid drop with one fixed point (at the lower part) and can expect some modes to coincide with those of a free drop. If we identify the minimum frequency for $l = 2$ with the first peak in figure 7 ($f = 1.3$ Hz) the next frequency modes can be calculated.

In table 1, the experimental peaks of frequency (figure 7) are compared with the frequency modes predicted by equation (5).

| Modes | $l = 2$ | $l = 3$ | $l = 4$ | $l = 5$ |
|-------|--------|--------|--------|--------|
| $f$, Hz (experiment, figure 7) | 1.3 | 2.6 | 3.57 | 5.6 |
| $f$, Hz (equation (5)) | 1.3 | 2.5 | 3.9 | 5.44 |

Table 1. The frequency modes from equation (5) for $l = 3, 4$ and 5 and peaks found in figure 7.
Thus, it can be seen that frequency modes for $l = 3$ and 5 coincide with experimental peaks obtained in figure 7. There is some discrepancy for $l = 4$ and there is a peak, $f = 3.2$ that is not foreseen in the theory. Here one should take into account the experimental errors in the determination of the particle positions, the fact that the cluster does not have ideal spherical shape and some instrumental noise.

In the hypothesis that the vibrations are due to surface tension we could estimate the surface tension of the cluster by using the first peak at $f = 1.3$ Hz. $T$ is found to be $3.3 \times 10^{-12}$ J m$^{-2}$. This value may look very small but should be compared with the energy of the external bonds in the cluster. From the analysis of shell structure (figure 4) we see that there are roughly two shells in the cluster of 60 particles: theoretically 12 particles in the first shell and the other 48 in the second shell. The number density of bonds between the first and second shells, $N$, is the ratio of the number of particles in the external shell to the surface area of the cluster, $48/4\pi R^2$. The ‘equivalent’ surface tension is obtained by multiplying $N$ by the energy per single bond, $1/2k\overline{\Delta d}^2$, with $\overline{\Delta d}$ the average value of the displacement w.r.t. the equilibrium. Equating the derived surface tension with $N \times 1/2k\overline{\Delta d}^2$ yields a value of $\overline{\Delta d} = 18 \mu$m, which matches well with the experimental, direct measurements of the particle displacement, namely 10–20 $\mu$m.

6. Conclusion

The possibility of producing 3D plasma clusters under gravity conditions and obtaining the three space coordinates as well as the velocities of the particles simultaneously allowed us to perform a kinetic analysis of the cluster structure and dynamics. We have demonstrated that the observed reorganization of a 3D plasma cluster is governed by the principle of minimum energy. In general these experiments allow us to follow in detail the evolution of the energy towards stable configurations, to compare and validate the huge simulation effort produced in the field. [9], [11]–[13], to quote only the most recent. A cluster of 16 particles was found in a metastable state and evolved to a fundamental state by the addition of an energetic particle. A cluster formed by 60 particles appears to be stable in accordance with the ‘magic’ number found in the simulation of [9, 11]. Accordingly, three particles evaporated from a 63 particle cluster so reducing the energy per bond providing an energetically stable state. The analysis of the cluster vibrations using fast Fourier transform shows resonance modes, the frequencies of which have been compared to dust waves and to the hydrodynamic approach. In this paper, we have provided the readers with the information for them to decide on the quality of the agreement. However no such vibrations were observed for the 17 particle cluster where the individual interactions remain more important. Sixty is the lowest number of particles to allow a 3D two closed-shells structure and is the smallest cluster, for the chosen conditions, where collective effects have been observed. It appears to us that the model for fluid like cooperative behavior can describe, with some approximation, the dynamics of such small clusters in spite of the obvious ‘discrete’ nature of these systems.

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