

Interparticle potential barriers in soft matter systems

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We discuss how a system of particles with bounded interaction potentials responds to increased densities. In particular we elaborate the ability of a system to form potential barriers between the particles, to allow a so called cluster formation. These theoretical considerations are complemented by computer simulations that allow us to observe the onset of clustering at low particle densities. We discuss the simulation results with regard to the presence of potential barriers and establish a formulation to explain the effect of potential barriers by the use of the Fourier transform of the potential function.

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I. INTRODUCTION

Within the field of computational physics, systems are usually described by the pair potential of their constituent particles. Under certain conditions, for example when studying specific macromolecules in a solvent, the pair potential can be approximated by a function which is bounded at its origin (e.g., [1]). This implies that the centers of mass of such macromolecular aggregates can—in contrast to conventional particles (e.g., Argon [2])—fully overlap.

The importance of systems with bounded interaction potentials, especially the Gaussian core model (GCM) [1,3–7], has become evident within the last years as theoretical and experimental studies have been performed [8,9]. It has been shown that different dendrimers [10,11] and polymer chains [12] can be very well approximated by such effective interaction potentials.

When increasing the density within a system of particles with bounded pair potential they can behave in two different ways. Depending on the pair potential they show either reentrant melting behavior or their particles arrange themselves in clusters. Within this context clusters are an aggregation of overlapping particles, which can form crystalline systems.

The effect of clustering takes place because the solid always lowers its free energy by allowing multiple occupied crystal sites. This effect was first discovered when simulating the penetrable spheres model (PSM) [13]. On the other hand, the GCM model shows fluid and solid phases as well as reentrant melting [8].

In order to explain the different mechanisms of soft matter systems at high densities, considerable effort has been put into experimental and theoretical studies. Within [14] Mladek *et al.* introduced the generalized exponential model with index n (GEM- n), which is defined as $v(r) = \epsilon e^{-(r/\sigma)^n}$. This model becomes the GCM with $n=2$ and approaches the PSM for $n \rightarrow \infty$, thus it is a model that allows us to observe the phase behavior of the system when the steepness of the potential is tuned.

Likos *et al.* [15] derived a criterion to determine whether a system shows reentrant melting or clustering, by employing a mean-field–density-functional theory. Within their work they distinguish between Q^+ and Q^\pm classes of potentials. The Fourier transform of Q^+ potentials contains only positive values, while the Fourier transform of Q^\pm potentials

contains positive and negative values. The criterion found by Likos *et al.* states that Q^+ potentials show reentrant melting behavior, while Q^\pm potentials will freeze at all temperatures in a clustered formation at high densities [15].

At first glance the formation of clusters seems to be counterintuitive, especially for systems with purely repulsive pair potentials, as one would expect attractive forces to be needed to keep the particles concentrated in a cluster.

Although purely repulsive potentials lack an attractive force, they are able to build an overall potential field within a system that provides less repulsion at a particle position than at a free space. In this case an attractive force is merely provided by the lack of repulsion.

Within our paper we try to explain the mechanisms of clustering by two different approaches: First we observe the pair potential and the resulting potential distribution in a solid formation with regularly spaced particles. We will formulate a necessary criterion for generic pair potentials to build up “quasiattractive” potentials. These considerations are explained in detail in Sec. II.

On the other hand, the formation of clusters is not only determined by the potential energy but rather a result of the interplay between the system’s potential energy and its entropy. Therefore the observation of the potential is important to qualitatively understand the mechanisms better, but it is not sufficient to quantitatively determine the structures a system will form. Therefore other methods have to be used. In this paper we document simulations of a system with a purely repulsive pair potential that give also insight into the mechanics of clustering.

Some of these simulations will directly show that the simulated system has a potential distribution according to the considerations presented in Sec. II, while other simulation results can be well explained by the theory. The simulations and their results are described in Sec. III.

A short conclusion is given in Sec. IV.

II. THEORY OF POTENTIAL BARRIERS

Within this section we consider systems with bounded potentials as a solid formation and observe the resulting potential of all particles. These summed potentials strongly influence the behavior of additional particles entering the system.

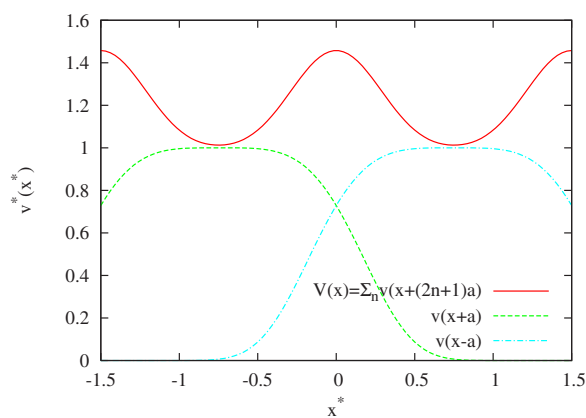


FIG. 1. (Color online) The pair potential of two particles and the summed pair potential of all particles. The particles are located with a distance of $2a^* = 1.5$ to each other, which is a cluster distance observed in previous simulations [14,16].

Within Fig. 1 pair potentials of the GEM- n system with index $n=4$ are drawn. This system was recently discussed and simulated [14,16]. The particles are placed at $x=(2m+1)a$ within a one dimensional system (m is any integer value), thus the distance between two neighbor particles is $2a$. Unless otherwise noted, all physical quantities in the figures of this paper are drawn in reduced units which is indicated by the superscripted asterisk, as defined in [17].

The dashed lines show the contributions of each individual particle, while the solid line shows the summation of all particles. It can be clearly seen that the summed potential has a higher value between the particles than at the positions where particles are placed.

An additional particle brought into the system will therefore observe a lower potential if it places itself onto another particle than in the case that it resides between the particles. In case the kinetic energy of this particle is low compared to the potential barrier, it will most likely move to the next minimum and stay there trapped.

Of course the particle could also stay at the top of a potential barrier; the probability to find the particle at a certain place within the system is defined by the Boltzmann distribution. That means the probability is not only dependent on the height of the potential barrier but also on the width of it compared to the width of the minimum, i.e., its geometric layout.

A potential distribution with very few or narrow minima will lead to an increased probability to find the additional particle away from the minimum. In other words, the fewer places with a potential minimum are available in a system the higher will be the entropy of the system with trapped (i.e., clustered) particles.

This is an important fact as it shows how the free energy is included in our model. Actual results of the interplay between the entropy and the potential energy are documented in Sec. III.

The height and even the occurrence of such a potential barrier is dependent on the distance $2a$, which is inversely proportional to the particle density of the system. We therefore expect the following behavior within a GEM-4 system:

Starting from a solid with a very low density we increase the density until potential barriers build up. When the density is further increased the barriers become high enough to allow clustering if any further particles are placed into the system.

Such a potential barrier cannot be generated with just any kind of pair potential. If we would, for example, try the same experiment with a potential function $v(r) = \epsilon e^{-r/\sigma}$ we would observe no higher potential than the one at the particle positions, no matter how we choose the distance a .

From these considerations we can intuitively conclude that only particles with a pair potential that makes potential barriers as illustrated in Fig. 1 possible can show clustering at high densities, while systems missing this potential barrier will fail to form a stable cluster formation.

As we are interested to find an answer to the question of whether a specific system can build up a potential barrier, we would have to check this for *any* distance $2a$. As this is a nontrivial task, we are looking for an analytical expression that can quickly give the answer to this question. An expression using the frequency domain representation of the effective pair potential also allows us to compare the result with the criterion derived by Likos *et al.* [15].

Within the following discussion we suppose an even, real-valued Fourier transformable potential function $v(x)$. To find out whether the sum of the potentials is higher in between the particles ($x=0$) or at the position of the particle ($x=a$) we calculate the difference

$$V(a) - V(0) = \dots + v(-2a) - v(-a) + v(0) - v(a) + \dots; \quad (1)$$

using the Kronecker δ [19] this summation can be rewritten as

$$V(a) - V(0) = \sum_{l=-\infty}^{\infty} v(la) \cos(\pi l) = \int_{-\infty}^{\infty} v(x) \sum_{l=-\infty}^{\infty} \delta(x - la) \cos\left(\frac{\pi}{a}x\right) dx. \quad (2)$$

As we required $v(x)$ to be an even and real function its Fourier transform will also be even and real. Let us call $\hat{v}(Q) = \sqrt{2\pi} \mathcal{F}[v(x)]$ where $\mathcal{F}[\]$ denotes the Fourier transform. Equation (2) is then identical to

$$V(a) - V(0) = \frac{1}{a} \sum_{l=-\infty}^{\infty} \hat{v}\left(\frac{\pi}{a} + l \frac{2\pi}{a}\right) \quad (3)$$

because the multiplication with the sum of Kronecker functions within Eq. (2) is a discretization within the spatial domain and corresponds to the periodic repetition of its Fourier transform as expressed in Eq. (3).

It is obvious that the difference (3) will always be positive for Q^+ potentials, i.e., no distance $2a$ can be found at which the potential between two particles is higher than at the particle position itself. Only Q^+ potentials can build up a barrier as depicted in Fig. 1 for certain values of a .

We can also learn something about the possible values of a from the following considerations: As the spectral bandwidth of conventional pair potentials is practically limited

we can define a value Q_0 in a way that $Q \cdot \hat{v}(Q)=0$ holds true with adequate accuracy for $Q \geq Q_0$ [20]. Therefore we derive a lower limit for a from Eq. (3):

$$a \geq \frac{\pi}{Q_0}. \quad (4)$$

On the other hand, we can obviously do the same in the spatial domain to obtain an upper limit for a . If $v(x)=0$ holds true for $x \geq x_0$ we derive

$$a \leq x_0 \quad (5)$$

as an upper limit for a . Following the notation of Likos *et al.* [15] we call Q_* the value of Q at which $\hat{v}(Q)$ attains its minimum, thereby yielding a negative value.

The result of the summation (3) will generally be dominated by the expression $1/a \cdot \hat{v}(\pi/a)$, the minimum of this expression refers to the maximal height of the potential barrier and will generally be reached at a particle spacing that is close to $2a=2\pi/Q_*$, which is the particle spacing at which a Q^\pm system will freeze according to [15].

These considerations can also be applied to systems of higher dimensionality, in particular to a three dimensional system. Let us assume that the particles are placed at $x=(2j+1)a$, $y=(2k+1)a$, and $z=(2l+1)a$ with the integers j, k, l . Within Eq. (6) the term $V(a)$ again refers to the potential which can be observed at a particle position [e.g., $(a \ 0 \ 0)$], and $V(0)$ is the potential at the point symmetrically placed between the particles at $(0 \ 0 \ 0)$. Equations (2) and (3) can then be rewritten as

$$\begin{aligned} V(a) - V(0) &= \int v(\vec{x}) \sum_{j,k,l} \delta(\vec{x} - a\vec{j}) \cos\left(\frac{\pi}{a} \vec{e} \cdot \vec{x}\right) d\vec{x} \\ &= \frac{1}{a^3} \sum_{j,k,l} \hat{v}\left(\frac{\pi}{a} \vec{e} + \frac{2\pi}{a} \vec{j}\right). \end{aligned} \quad (6)$$

with

$$\vec{j} = \begin{pmatrix} j \\ k \\ l \end{pmatrix} \quad \vec{e} = \begin{pmatrix} 1 \\ 1 \\ 1 \end{pmatrix}. \quad (7)$$

We obtain the same result within the three dimensional system as in the one dimensional: Q^+ potentials will always require the difference (6) to be positive and thus they will not be able to form a stable clustered configuration.

The argument presented here only takes into account the internal energy U , whereas at finite temperatures it is rather the free energy $F=U-TS$ that matters. Whereas energy favors cluster formation, entropy plays a dual role. On the one hand, a cluster consisting of n_c particles entails an entropic cost of $k_B \ln(n_c)$, thus clusters are disfavored. On the other hand, cluster formation opens up new free space, so that the free volume entropy increases and cluster formation is favored anew [14]. The simulation results presented in Sec. III demonstrate that cluster formation does indeed bring about an overall free energy gain, a fact that has also been confirmed by analytical calculations based on density-functional considerations [18].

III. SIMULATION RESULTS

To obtain better insight into the mechanisms of clustering we carried out a large number of simulations within the GEM-4 system. These simulations were performed on a high performance reconfigurable computing platform, on which we implemented a lattice Monte Carlo algorithm.

For this purpose the configuration space was discretized to allow the pair potential values to be tabulated. The resulting lookup tables were copied into a so-called “field programmable gate array” (FPGA), where their content was read out in a parallelized fashion to allow considerable speedup for the calculation of the total potential. We chose the discretization in a way that its effect became negligible compared to other sources of error, which we verified experimentally and analytically [16]. All simulations were performed in the NVT ensemble with a varying number of particles to keep the size c of the simulation cube constant.

To set up simulations that support the theory presented in Section II, we had to evaluate first the phase diagram in a wide density range. Using the phase diagram we were then able to set up a system exactly at the onset of clustering, which is described later in this section. We determined the phase diagram by simulated annealing and melting simulations. This would be a relatively straightforward task within a system that does not build clusters, but is somehow more complicated when the number of particles is not known that will form a cluster.

To allow a fluid to crystallize it is necessary to simulate the correct number of particles in the system, which build up the correct number of clusters to form either a bcc or fcc lattice. Therefore we chose an iterative approach, where we set up a system at a high density ($\rho^*=9$) and temperature $T^*=2$ and cooled it slowly down until a solid state was reached. The cluster density $\rho_c^*=0.48$ (the number of clusters within the volume unit) of this solid was taken as a starting value for the following iterations at lower densities: (i) Set up a system to obtain 108 clusters (this would allow an fcc crystal for the clusters) with a given ρ_c^* at a temperature where it is in a fluid phase. (ii) Slowly cool down the system until it reaches a solid state. (iii) If 108 clusters in fcc configuration are met the iteration has finished. Otherwise calculate the obtained ρ_c^* and start over at point 1.

The same procedure was performed to allow a bcc crystallization (128 clusters). The fcc iterations converged only at one density after two iterations, at all other densities the iterations did not converge at all. The result of the annealing process were disordered, glasslike configurations of clusters.

The bcc iterations converged at six different densities after the first, at four different densities after the second, and at three different densities at the third iteration.

Although it cannot be definitely said which of the crystal configurations (bcc/fcc) is the “natural” choice of the system at the fluid to solid transition, as the free energy was not calculated within the simulations, these simulations give evidence that the system prefers a bcc configuration when building up clusters.

The results of these simulations are summarized in Fig. 2; the dashed line in this figure is the function for a system that shows no clustering (i.e., the particle density is always equal

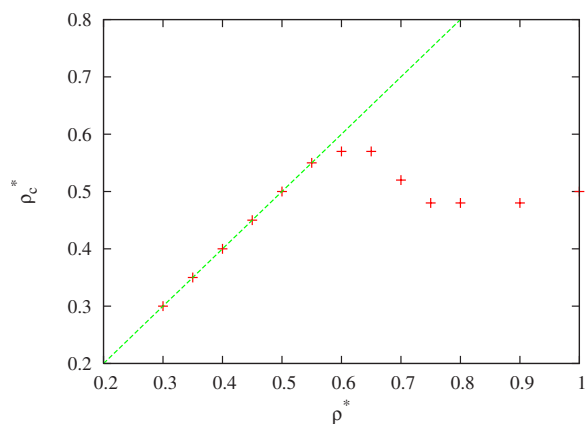


FIG. 2. (Color online) The cluster density of a GEM-4 system dependent on the particle density, obtained by iterative simulated annealing simulations. The dashed line shows the function $\rho_c^* = \rho^*$.

to the cluster density). It seems that the system “tries to avoid” clustering as long as possible by lowering the particle spacing. But starting at $\rho^* = 0.55$ more and more particles find their place “on top of other” particles.

This behavior can be explained by the fact that the potential barriers need a certain distance between the particles to build up. As long as the distance between two particles is too low to build up sufficiently large potential barriers, the particles do not form clusters. When the system reaches a certain density, where each particle is surrounded by a potential barrier, the clustering sets in.

At $\rho^* = 1.0$ we found all clusters occupied with two particles in the system (i.e., ρ_c^* increases to 0.5). Again the system avoids as long as possible to build three particle clusters by lowering the particle spacing. For densities $\rho^* > 1$ the cluster density remained at $\rho_c^* = 0.48$. This corresponds to a widely constant lattice spacing for all densities where clustering can be observed, with a slightly smaller lattice spacing at lower densities where clustering sets in. As potential barriers can only be built up at certain lattice spacings the constant cluster density is a prerequisite to form clusters at all densities. The effect of constant lattice spacing was already predicted in [15].

After determining the function $\rho_c^*(\rho^*)$ it was straightforward to obtain the phase transition temperature as a function of the density. We set up the system at different densities ranging from $\rho^* = 0.3$ to $\rho^* = 7$ with a number of particles that allows the system to crystallize in the appropriate formation. The system was then slowly cooled down from the fluid state until a solid formation was reached.

To verify whether the cooling rate was low enough, we also performed simulated melting. For this purpose we set up crystalline systems with the expected values of ρ^* and ρ_c^* and heated them up with the same rate as the simulated annealing was performed. The melting and the annealing temperatures were found to match with only small deviations. Thus the cooling and heating rates are considered to be low enough for accurate results.

The result of these simulations is drawn in Figs. 3 and 4. The curves agree very well with the results from Mladek *et al.* [14], who obtained the phase diagram by use of a density

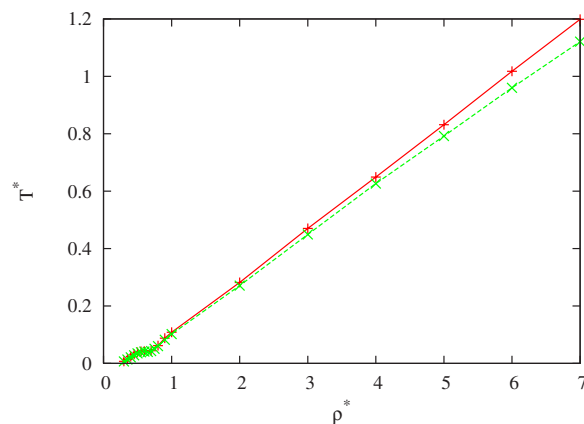


FIG. 3. (Color online) The phase diagram of the GEM-4 system, as derived by simulations within an NVT ensemble (redrawn from Ref. [16]). The phase transition points were obtained by simulated annealing (x) and simulated melting (+).

functional theory for high densities. For densities $\rho^* > 1$ we can see that the phase transition temperature increases linearly with the density. In Fig. 4 we can observe that the clustering, which sets in at a low density of $\rho^* = 0.55$, reduces the melting temperature by a certain value.

In our case the phase transition was determined by observing the potential of the system while varying the temperature. Such a curve can be found in Fig. 5 for a density where no clustering takes place. The step at the phase transition can be clearly seen.

In Fig. 6 the system was set up at a density where clustering takes place, the step at the phase transition is softened in the solid and the fluid phase within the system. The softening within the fluid phase is probably due to the building of temporary clusters in the fluid, as they are described in [18].

The softening in the solid phase is related to the higher mobility of the particles in and between clusters at higher temperatures. Some of the particles are acting like a fluid within the solid.

At low temperatures the particles rested on each other, resulting in a small expansion of the cluster. At higher temperatures, we observed that the individual particles became

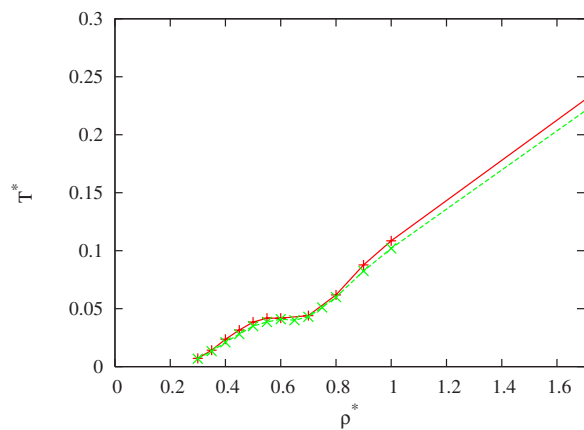


FIG. 4. (Color online) A detail of Fig. 3 showing the onset of clustering at low densities.

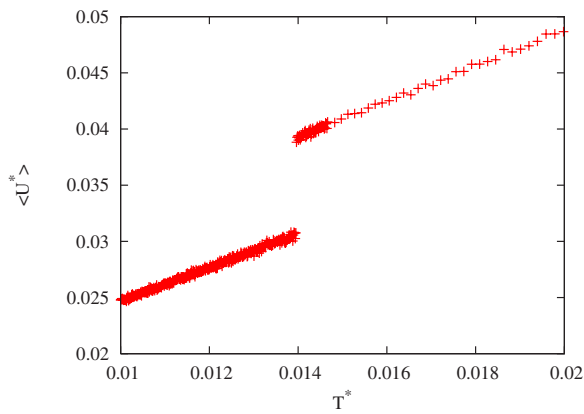


FIG. 5. (Color online) The mean energies of a system at $\rho^* = 0.35$ set up in an fcc configuration. At this density no clustering takes place. During the simulations the temperature was slowly changed from $T^* = 0.01$ to $T^* = 0.02$. The melting point can be clearly seen as a step in the potential energy of the system.

more and more mobile within and between the clusters, i.e., the geometric cluster dimension increased whereas the crystalline arrangement of them was preserved.

This behavior can be explained if we assume a potential function with a minimum at the center of a cluster (as depicted in Fig. 1 for a two particle cluster). At low temperatures all particles try to rest close to the minimum of the potential function. At high temperatures the particles can also reach positions with higher potential energy.

When the step at $T^* = 0.65$ was reached, the crystalline formation was destroyed and the system changed into the fluid state. When a system in such a fluid state is cooled down very quickly, it also forms clusters but they are in general not arranged in a regular lattice. Unregular solid formations of clusters were observed; an example is shown in Fig. 7.

Such glasslike formations have a stability against remelting (i.e., the number and position of the clusters are unchanged) up to a temperature that is slightly lower than for the crystalline formation. They also show cluster expansion

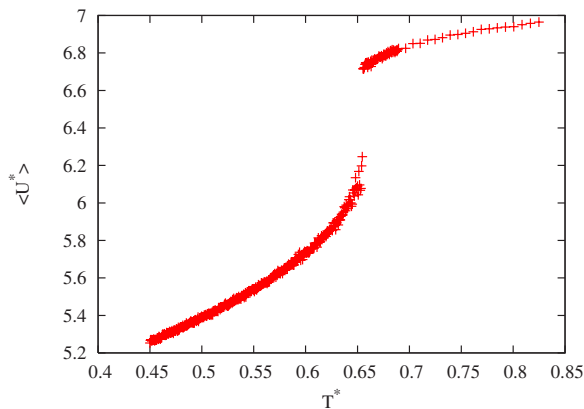


FIG. 6. (Color online) The mean energies of a system at $\rho^* = 4$ set up in a bcc configuration, forcing the system to build large clusters. During the simulations the temperature was slowly changed from $T^* = 0.45$ to $T^* = 0.83$. The energy step softened in the solid as well as the fluid formation.

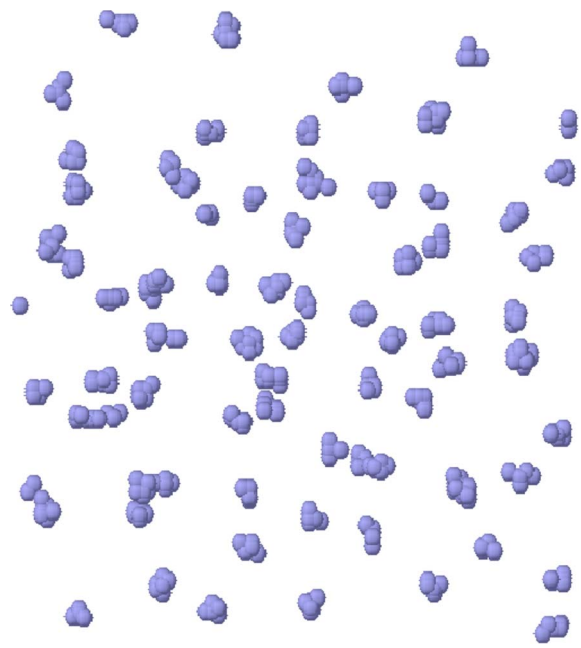


FIG. 7. (Color online) A system in a fluid state at $\rho^* = 4$ was cooled down very quickly to $T^* = 0.1$. The clusters are not located on a regular lattice, nevertheless, they maintained a fixed relative position to each other during the whole simulation time of 1 000 000 passes. Particle diameters are not drawn to scale but are chosen to optimize visibility of the structure.

at higher temperatures and the intercluster “tunneling” effect as described in more detail later on. This behavior was verified during simulated melting with very low heating rates of up to one million passes per temperature increment.

The potential barriers can be formed at different interparticle distances, as discussed in Sec. II. Glasslike configurations are therefore not contradictory to the theory of potential barriers, as long as the clusters maintain certain restrictions on minimum and maximum distance to each other. The simulation results show that also the glasslike configurations provide a constant cluster density, thus the average distance between the clusters is in the same order as in crystalline systems indicating that the minimum and maximum restrictions can be met.

To give our assumptions from section II experimental evidence, we set up a system at a density where clustering sets on. We chose a bcc lattice configuration, where every lattice site was occupied by one particle and put one additional particle between the lattice positions. The temperature of the system was chosen to be low to minimize the noise on the trajectories of the additional particle. The movement of this additional particle can be seen in Fig. 8. Within a few simulation passes the particle moves onto one of the nearest particles. We observed that the lattice structure remained unchanged. All positions are now occupied with one particle except one lattice site which hosts two particles. The energy that this additional particle observes during these simulation steps can be seen in Fig. 9. Clearly the energy drops when the double occupation of the lattice position is reached.

The increased potential energy of the particle while it is located between the regularly spaced particles indicates

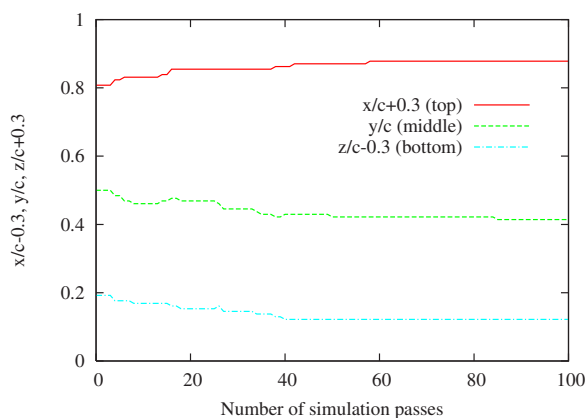


FIG. 8. (Color online) The trajectories of a particle placed within an fcc lattice at $\rho^*=0.55$ and $T^*=0.001$. The particle immediately moves onto another particle while the fcc lattice remains undisturbed. The coordinates of the particle are drawn in relation to the size of the simulation cube c .

clearly the existence of a potential barrier. The straight movement of the particle onto the next neighbor is an indicator that the repulsive force from the potential barrier is responsible for the clustering. With this experiment we also proved that the assumption of a static lattice is valid when the system is set up at the density where clustering takes place.

In another experiment we chose the same setup at a higher temperature and observed the movement of the additional particle over a longer time. After an arbitrary number of simulation passes the particle moves to another lattice site. This behavior can be seen in Fig. 10.

This sudden movement to other lattice sites often involves more than just one particle. The tunneling results in a chain

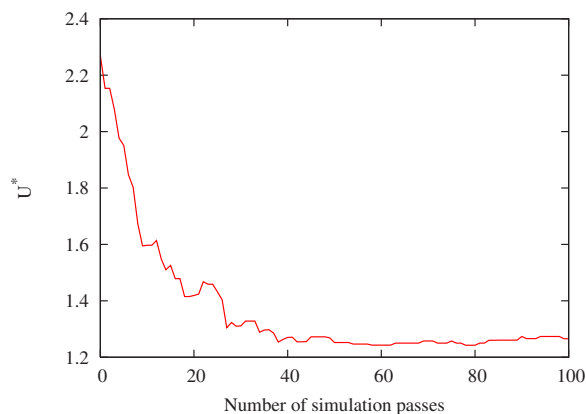


FIG. 9. (Color online) The potential energy of the additional particle from the experiment described in Fig. 8. Before the particle reaches the position onto another particle its energy level is clearly increased.

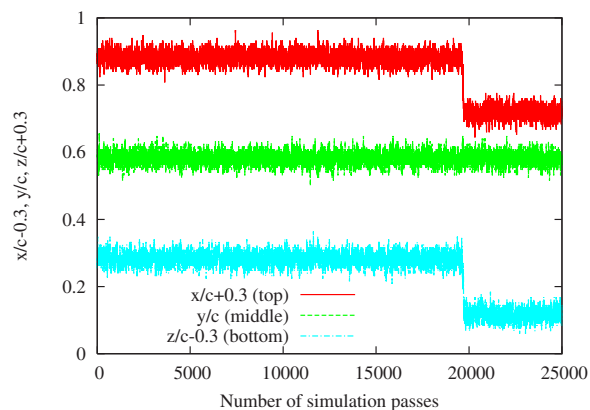


FIG. 10. (Color online) The trajectory of the additional particle at $\rho^*=0.55$ and $T^*=0.03$. At an arbitrary point of time the particle jumps to a different crystal site. The coordinates of the particle are drawn in relation to the size of the simulation cube c .

reaction where the particle of the target lattice site is knocked out of its position and has to look for a new place. It was experimentally verified that at higher temperatures the particles are changing the lattice site more frequently. The inter-cluster tunneling is not limited to low densities and was also observed at high densities in ordered and disordered solid phases.

Potential barriers require particles with a high kinetic energy to move from one cluster to another. Such particles are more often present in systems with a higher temperature. Thus the increased frequency of cluster tunneling at higher temperatures can be well explained with the theory of potential barriers.

IV. CONCLUSIONS

We considered systems made of particles that interact by means of a pair potential that is bounded. We defined the system to be in a solid state and observed the sum of the potential functions of all constituent particles.

We presented results of simulations demonstrating the clustering effect at low densities, by observing trajectories of individual particles and the crystalline structure of the system. We formulated the ability of the system to build potential barriers between particles by an equation using the spectral representation of the pair potential. The results we obtained are in line with the results from Likos *et al.* [15] and provide an alternative, simplified, and intuitive view on the effect of clustering.

The equations derived herein are not limited in their validity to potential functions that are purely repulsive. The only restrictions to the potential function are that it is even, real, and Fourier transformable.

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- [19] If we do not use a system with reduced units as, for example, described in [17], the Kronecker function has to have the same unit as $(1/x)^D$, where D denotes the dimensionality of the argument.
- [20] While one will always find an appropriate Q_0 for real world potential functions, this is not possible for functions containing discontinuities as, for example, the PSM, because the term $Q \cdot \hat{v}(Q)$ does not vanish for $Q \rightarrow \infty$. Therefore these models do have the ability to build up potential barriers without a lower limit for the particle distance $2a$, which can be easily verified for the PSM within the spatial domain.