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Calculation of correlation lengths in 2D Lennard-Jones fluids

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Abstract. The process of spinodal decomposition in two-dimensional system that was obtained by molecular dynamics simulation has been analysed using the correlation of reciprocal areas of the Voronoi cells. Correlation lengths dependence on the temperature and the critical exponent of the LJ12-6 system in the fluctuation region by the general renormalization group (RG) framework were calculated. We showed the agreement of the calculated critical exponent ν with both experiments and theory. The proposed methodology for correlations of the reciprocal areas of the Voronoi cells is well applicable in the experiments with 2D colloidal systems.

Keywords: soft matter, colloidal particles, tuned self-assembly, phase transitions, molecular dynamics

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Материалы конференции

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Вычисление корреляционных длин в двумерных жидкостях с потенциалом Леннарда-Джонса

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Аннотация. Проанализирован процесс спиноподального распада в двумерной системе, полученный методом молекулярной динамики с помощью корреляции обратных площадей ячеек Вороного. Рассчитана зависимость корреляционных длин от эффективной температуры системы с потенциалом взаимодействия LJ12-6 в области спиноподального распада. Показано соответствие вычисленного критического индекса ν как экспериментальным исследованиям фазовых переходов в двумерных ферромагнетиках, так и теоретическим исследованиям систем Изинга. Предложенный метод корреляций обратных площадей ячеек Вороного может быть применим в экспериментах с двумерными коллоидными системами.

Ключевые слова: мягкая материя, коллоидные частицы, управляемая самосборка, фазовые переходы, метод молекулярной динамики

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Introduction

Method for the characterization various phases of matter is one of the central tasks of condensed matter physics and materials science. In particular, the study of the phase transitions in 2D systems has played a crucial role for understanding of the phase transitions and it can be extended to other complex systems. Understanding the process of the phase transitions in 2D systems plays important role in a number of areas, from photonics and electronics [1–8], to novel materials [9, 10], and biophysics [11], since knowledge of the phase behavior opens way to designing systems with the necessary properties.

A great number of phase transitions phenomena studies are carried out by methods of the molecular dynamics (MD) and Monte Carlo (MC) simulation. One of the simplest model which is capable of reproducing gas, liquid, and solid phases behaviour is the Lennard-Jones (LJ) system. Such systems make it possible to study melting and crystallization [12–14], condensation and evaporation [15], spinodal decomposition [16–18].

One of the platforms mimicing molecular behavior are colloidal systems [19–23]. The studies in such systems include crystallization and melting [24–31], reentrant and solid-solid phase transitions [32–34], condensation and critical phenomena [35–37], molecular-like interactions [38, 39], sublimation [40], gelation, slow dynamics in glasses [41–44], the role of three-body interactions of phase transitions [45].

Tunable interactions between colloidal particles can be provided using different physical mechanisms [46]: electrostatic interactions in solvents, [47, 48], tunable interactions induced by external magnetic fields [27, 49], alternating anisotropic and rotating electric fields [50, 51]. Rotating electric fields make it possible to induce and to control interparticle interactions in colloidal systems, and collective dynamics of colloidal particles can be visualized in real time with the spatial resolution of individual particles. In rotating electric fields, interparticle interactions in a colloidal system monolayer are tuned by the following mechanism [51–54]: an external electric field polarizes colloidal particles, that leads to anisotropic interactions; a fast (compared to the particle diffusion time) rotation of the field in the plane of the system makes it possible to achieve isotropic tuned dipole attraction at large distances [51, 54]. The colloidal system in the rotating electric field provides a rich variety of different interactions, including repulsion, attraction, combination of short-range repulsion with long-range attraction, barrier-type interactions with short-range attraction and long-range repulsion, and double-scale repulsive (core–shell) interparticle interactions [55]. This makes it possible to use such system for particle-resolved study of phase transitions [17].

It is required a methodology to study critical phenomena in colloids. The divergence of the correlation length for a mixed phase transition in a 2D colloid model system with nearest neighbor interaction has been shown in Paper [56]. The authors in [57] attempted to calculate the correlation length using the effective interfacial thickness in 2D LJ fluids. In our work, we suggest using the correlation function of reciprocal areas of the Voronoi cells critical temperature vicinity. It allows us to calculate the dependence of the correlation length on the temperature and to determine critical exponent ν in 2D systems.

Materials and Methods

To study the efficiency of density-density correlation method (correlation of reciprocal areas of the Voronoi cells), we performed MD simulations of the system undergoing spinodal decomposition.

We considered system of particles interacting via the potential:

$$\phi_{12-6}(r) = 4 \epsilon \left[\left(\frac{\lambda}{r} \right)^{12} - \left(\frac{\lambda}{r} \right)^6 \right]. \quad (1)$$

Where $\phi_{12-6}(r)$ is the usual Lennard–Jones potential (LJ12-6 potential), ϵ and λ are the strength and range of the interaction, respectively. We use the dimensionless temperature $T/\epsilon \rightarrow T$, distance $r/\lambda \rightarrow r$, surface number density $n\lambda^2/m \rightarrow n$, and time

$$t \sqrt{\frac{\epsilon}{m}} \lambda^2 \rightarrow t$$

(here, m is the mass of the particle).

To obtain correlation length dependence on the temperature the MD simulations of a system containing $N = 10^4$ particles were performed in an NVT ensemble for different temperatures above the critical. Initially particles were distributed uniformly over the simulation region with periodic boundary conditions. Then at every temperature, it was integrated for 10^6 steps until the equilibrium state was reached. The time step of $\Delta t = 5 \times 10^{-3}$ was used. The last 10 snapshots of the system were used for correlation lengths calculations with a sampling rate of 5×10^{-3} steps. The cutoff radius was set to $r_c = 5$.

During spinodal decomposition a new state of reduced symmetry emerges continuously from the disordered or symmetric phase as the temperature is reduced. To analyze the fluctuations in the system, it is proposed to use correlation of reciprocal areas of the Voronoi cells $C(r, r')$.

Let us define the correlation function $C(r, r')$:

$$C(r, r') = \langle \phi(r) \phi(r') \rangle. \quad (2)$$

The correlator of the system was calculated as reciprocal areas product of the Voronoi cells of particles at the radius-vector $r - r'$, averaged over each particle $r - r'$. Figure 1 shows the mechanism for calculating the correlator $C(r, r')$. The area of the Voronoi cell $\phi(r)$ corresponding to the particle at the r' multiplied by the reciprocal area $\phi(r')$ corresponding to the particle at the $r - r'$.

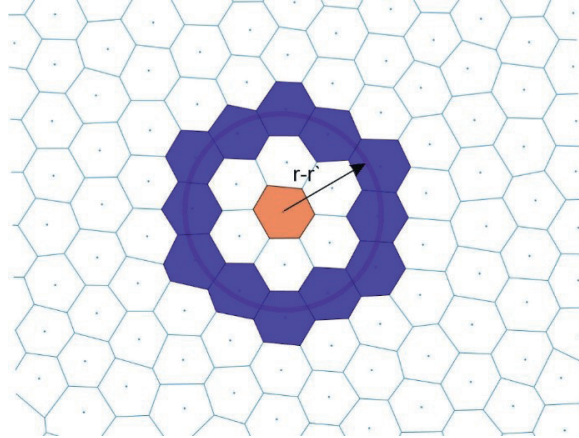


Fig. 1. The field of Voronoi cells for calculating correlator of the system. Voronoi cell corresponding to the particle at the radius vector r is colored orange; Voronoi cells corresponding to the particles at the radius vector $r-r'$ over which averaging is carried out are colored blue

The correlator $C(r, r')$ of the system decreases rapidly with distance above the critical point T_c and nonzero at and below T_c .

In the self-consistent field theory, from the Ginzburg-Landau free energy equation, the correlator $C(r, r')$ of the system is given as follows [58]:

$$C(r) = r^{1-\frac{d}{2}} K_{\frac{d}{2}-1} \left(\frac{r}{\xi} \right), \quad (3)$$

where due to symmetry we can write $C(r)$ instead of $C(r, r')$, ξ is the correlation length, d is the dimension of the system and K_n is the Macdonald function of the n -th order.

Results and Discussion

The correlator (eq. 2) was calculated for the MD system in the vicinity of critical point temperature range. The correlator dependence on the distance and its fit by (eq. 3) are shown in Figure 2.

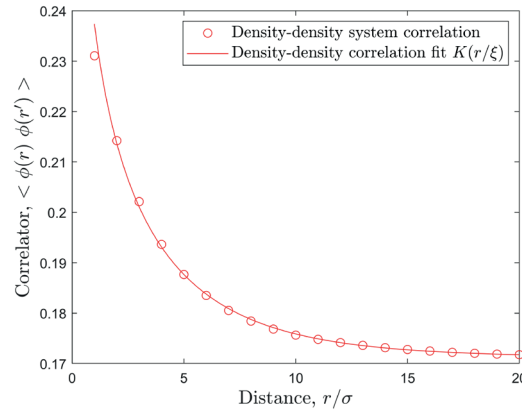


Fig. 2. The correlator dependence on distance of the 2D LJ12-6 system. Red symbols \circ correspond to calculated correlation, σ is the particle radius. Red solid line is the fit of the calculated correlator (eq. 2) by the equation (3)

From the approximation of the correlator $C(r)$ by the Macdonald function (eq. 3) the correlation length ξ for different temperatures were calculated. Several MD simulation timesteps were considered and Figure 3 shows the average correlation length dependence on reduced temperature $\tau = \frac{T - T_{cp}}{T_{cp}}$, where T_{cp} is the critical temperature.

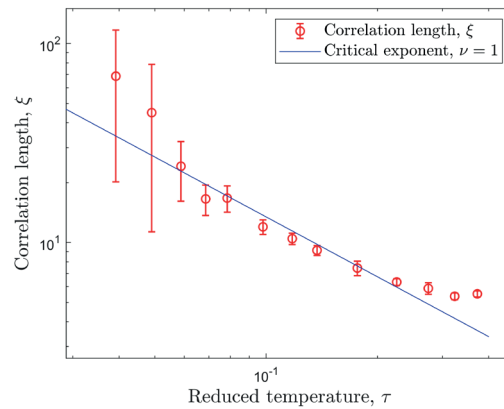


Fig. 3 The correlation length dependence on reduced temperature τ . Red symbols \circ correspond to correlation length; vertical bars correspond to confidence interval of the averaged correlation lengths at several timesteps (confidence level 0.95). Blue solid line is the fit of the correlation length critical behavior (eq. 4)

Critical exponents phenomenologically describe the behavior of many-body systems close to criticality. The behavior of the correlation length on reduced temperature in the fluctuation region is described by the critical exponent ν :

$$\xi = \tau^{-\nu}. \quad (4)$$

Using equation (4) the critical exponent for the 2D LJ system was determined to be $\nu = 1.00 \pm 0.07$. For comparison, Table 1 shows some estimations for the critical exponent ν of two-dimensional systems. One can see agreement with the proposed method.

Table 1

Experimentally measured critical exponents of some two-dimensional systems

System	ν	Class	Ref.
Rb ₂ CoF ₄	0.99(4)	Ising	[59], [60]
K ₂ CoF ₄	0.97(4)	Ising	[61], [60]
Fe/W(110)	0.93(14)	Ising	[62]
LJ12-6	1.00(7)	Ising	Current work

The LJ12-6 interaction demonstrates a 2D Ising-like critical behavior [63]. For two-dimensional Ising systems it is shown that the critical exponent $\nu = 1$ [64].

Conclusion

Thus, methodology for calculating the correlator of reciprocal areas of the Voronoi cells was tested on the LJ12-6 two-dimensional system. Correlation lengths in the area above the critical transition was estimated. The critical exponent ν describing the behavior of the correlation lengths in the fluctuation region was determined. Results were compared with both experimental and theoretical data. This methodology of critical exponent estimation is well applicable for two-dimensional systems and can be applied to experimental systems of colloidal particles in the external electric rotating fields.

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