Glass transition in a two-dimensional core-softened system
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Abstract. In the present paper we study a phase diagram of a model core-softened system in two dimensions. Previous studies of this system showed that the phase diagram of this system contains several ordered phases (low and high-density triangular phases, square phase and dodecagonal quasicrystal). However, there is a region of (P,T) parameters where the structure has not been identified. In the present paper we study the structure and dynamical properties of the system in this region and show that it experiences glass transition. Basing on the temperature dependence of the diffusion coefficient we estimate the glass transition temperature and place it on the phase diagram.

Keywords: two-dimensional core-softened system, phase diagram, ordered phases, glass transition

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1. INTRODUCTION
Glass transition is one of the most intriguing problems which does not find a solution already for about a century. One of the difficulties in investigation of glass transition is complex nature of glass-forming systems [1]. Experimentally one often uses some organic glass-formers (molecular liquids), metallic alloys (metallic glasses), network-forming liquid etc., i.e. rather complex systems. Importantly, these systems are typically multicomponent. One component systems usually demonstrate strong ability to crystallization which prevents their transformation to glass. However, glass transition can be observed in some model systems in frames of computer simulation methods [2-4]. Since one component systems are much simpler then the multicomponent ones their investigation allows to reveal some general features of glass forming systems which is important for the general understanding of the problem.

The situation becomes even more complex in the case of two-dimensional (2D) systems. It is well known that 2D systems can demonstrate long time tails of the velocity autocorrelation functions, which makes the diffusion coefficient to be finite even at low temperatures [5]. It makes them much worse glass-formers comparing to the three dimensional (3D) ones [6]. However, some models do demonstrate glass transition even in monatomic 2D system [7-9]. Refs. [7–9] studies the so-called Lennard-Jones-Gauss system (LJG). This is the system of particles interacting via the pair potential which consists of two parts: Lennard-Jones one and a Gaussian step. As a result the potential becomes a core-softened one. This system was widely studied in 3D [10-14]. However, its phase diagram has
not been calculated. Most of the calculations focused on the properties of LJG liquid [10-14]. In 3D this system demonstrates numerous liquid-state anomalies, including density anomaly [13], diffusion anomaly [14], structural one [13] and the anomalous behavior of viscosity [10, 11]. At some values of parameters of the potential LJG system can form quasicrystalline phases both in 2D and 3D [15, 16]. It also demonstrates high glass-forming ability [17].

The LJG potential belongs to a family of so-called core-softened potentials. It is well known that many core-softened systems demonstrate complex behavior [18]. Moreover, the behavior of these systems strongly depends not only on the functional form of the potential, but on the parameters of the potential too: even tiny changes of the parameters can lead to dramatic change in the behavior. A particular core-softened model introduced in our previous works is so called Repulsive Shoulder System (RSS) [3]. This is the system of particles interacting via a potential:

\[ U(r) = \left( \frac{\sigma_1}{r} \right)^{14} + 0.5(1 - \tanh(k(r - \sigma_1))). \]

The parameters \( \varepsilon \) and \( \sigma \) give the energy and the length scales in the system. Below we will use them as the units of energy and length respectively: \( U' = U/\varepsilon, \quad r' = l/\sigma. \) Since only these units will be used we will omit the prime marks. The parameter \( k \) is fixed to be \( k = 10.0. \) The parameter \( \sigma_1 \) determines the width of the repulsive shoulder. Fig. 1 shows the potential for \( \sigma_1 = 1.35. \)

The behavior of this system was studied in a number of papers in both 3D [3, 19–22] and 2D [23–27]. It was shown that at \( \sigma_1 = 1.35 \) the 3D system has complex phase diagram with several crystalline phases. Glass transition was also indicated in the system [3, 4]. Interestingly, a tiny change to \( \sigma_1 = 1.37 \) suppresses the glass transition and leads to formation of a dodecagonal quasicrystal [28, 29]. This system also demonstrates numerous liquid state anomalies, which also strongly depend on the parameters of the potential [20-22].

In Refs. [23-27] the phase diagram of RSS with \( \sigma_1 = 1.35 \) in 2D was studied. It was shown that this system forms three crystalline regions (low density triangular crystal, square crystal and high density triangular one). A dodecagonal quasicrystal phase appears in between of the square phase and the high density triangular one [27]. No stable ordered phase was identified in the region between the low density triangular phase and the square crystal.

In the present work we study the 2D RSS in the region of densities between the low density triangular phase and the square one. We show that the system does not demonstrate any ordered structure in this region within the simulation time. At the same time when the temperature is lowered the dynamics of the system becomes slower. Basing on this we conclude that the system undergoes a transition into a glassy state.

2. SYSTEM AND METHODS

In the present paper we study a 2D system of 20000 particles in a rectangular box with size ratio \( L_y/L_x = 3^{1/2}/2 \) by means of molecular dynamics method. Periodic boundary conditions are applied in both directions. The system is simulated in canonical ensemble (constant number of particles \( N \), square of the system \( A \) and temperature \( T \)) by means of molecular dynamics method. The time step is \( dt = 0.001. \) Firstly \( 5 \cdot 10^7 \) steps are made for equilibration of
the system. After that more $2 \cdot 10^7$ are performed for calculation of properties.

We calculate the equation of state of the system and its internal energy. The structure of the system is characterized by the radial distribution functions (RDFs) and the diffraction patterns. The dynamical properties of the system are probed by the mean square displacements. The diffusion coefficient is obtained by Einstein method.

3. RESULTS AND DISCUSSION

We start the discussion from the equation of state of the system in the region of interest. Fig. 2 shows the equation of state along several isotherms and the isochore $\rho = 0.7$. One can see that the isotherms demonstrate two peculiar regions: at the densities about $\rho = 0.6$ and $\rho = 0.77$. The nature of these peculiarities is well studied in our previous publications: the former region corresponds to the melting of triangular crystal with low density, while the later one is the melting region of the square crystal.

At the same time one can see that the isochor of the system experiences a bend at the temperature about $T = 0.06$. Apparently the high temperature phase of the system is liquid. Therefore, the liquid also experiences some transformation being cooled along this isochore. These observations mean that in the region between the triangular and square crystal another phase which has not been recognized in the previous works takes place. The goal of the present paper is to identify this phase and to describe its properties.

In order to characterise the structure of the system in the gap between the triangular and the square phases we calculate the Radial Distribution Functions (RDFs) at very low temperature $T = 0.001$ and several densities. The results are shown in Fig. 3a. The RDFs

**Fig. 2.** Equation of state of the system (a) along a set of isotherms and (b) along the isochore $\rho = 0.7$. The arrows in panel (a) mark the peculiarities due to the melting of triangular and square crystals.

**Fig. 3.** (a) Radial distribution function at several densities at $T = 0.001$. (b) Diffraction pattern at $T = 0.001$ and $\rho = 0.66$. 
of crystalline phases at such low temperature look like a set of $\delta$-peaks, which is exactly what we expect for the low temperature crystals. However, at intermediate densities the RDF of the system experiences a qualitative change (see the inset of Fig. 3a). Two relatively high peaks (however, much lower then the first peaks of the crystalline structures) are observed. However, at large distance the RDF looks liquid-like. We conclude that the system demonstrates a pronounced short-range ordering, but not the quasi-long-range, as it should be for a 2D crystal. Further evidence of this assumption is obtained from the diffraction pattern of the system given in Fig. 3b. No clear structure is observed, i.e. the system remains disordered. Basing on this we assume that the phase between the triangular and square crystals is glass.

In order to characterize the dynamical properties of the system we study the mean square displacement of the system. Fig. 4 shows the mean square displacement at the isochor $\rho = 0.7$ and different temperatures. One can see that at high temperatures is looks liquid-like, while when the temperature is lowered it becomes shoulder-like in the whole range of measured times. In order to estimate the glass transition temperature we fit the inverse diffusion coefficient $1/D$ to a well known Vogel-Fulcher-Tammann (VFT) law:

$$\frac{1}{D} = \frac{1}{D_0} \exp\left(\frac{A}{T - T_g}\right),$$

where $T_g$ is the glass transition temperature, and $A$ and $D_0$ are fitting coefficients. Fig. 5ab the obtained diffusion coefficients and fitting of $1/D$ by the VFT law. From the fitting we obtain that $T_g = 0.0328$.

From the results above we conclude that in the region of the phase diagram between the low-density triangular crystal and the square one the system experiences the glass transition. This result fills the gap of unrecognized phase in the phase diagram of the system. The complete phase diagram is shown in Fig. 6.
4. CONCLUSIONS
A simulation study of structural and dynamic properties of a 2d core-softened systems was performed. Previous studies of this system failed to recognize a phase between the low-density triangular crystal and the square one. The results of this work demonstrate that in this region the system experiences the glass transition.

REFERENCES
1. Binder K, Kob W. Glassy Materials and Disordered Solids: An Introduction to Their Statistical Mechanics. World Scientific Publishing Company, 2005.
2. Dzugutov M. Phys. Rev. A, 1992, 46:R2984-2987.
3. Fomin YuD, Gribova NV, Ryzhov VN, Stishov SM, Frenkel D. J. Chem. Phys., 2008, 129:064512.
4. Ryltsev RE, Chtchelkatchev NM, Ryzhov VN. Phys. Rev. Lett., 2013, 110:025701.
5. Hansen JP, McDonald IR. Theory of Simple Liquids, 2013, Elsevier.
6. Flenner E, Szamel G. Nature Comm., 2015, 6:7392.
7. Mizuguchi T, Odagaki T. Cent. Eur. J. Phys., 2009, 7(3):479-482.
8. Mizuguchi T, Odagaki T, Umezaki M, Koumyou T, Matsui J. AIP Conference Proceedings, 2008, 982, 234.
9. Thi Nhu Tranh D, Van Hng V, Thi Thu Hanh T. Mater. Research Express, 2008, 5:015205.
10. Fomin YuD, Ryzhov VN. Physics Letters A, 2013, 377:14691473.
11. Egorov SA. J. Chem. Phys., 2008, 128:174503.
12. Zhou S, Solana JR. Mol. Phys., 2017, 116:491-506.
13. de Oliveira AB, Barbosa MC, Netz PA. Physica A, 2007, 386:744-747.
14. Netz PA, Raymundi JF, Camera AS, Barbosa MC. Physica A, 2004, 342:4853.
15. Engel M, Trebin H-R. Phys. Rev. Lett., 2007, 98:225505.
16. Engel M, Trebin H-R. Philosophical Magazine, 2008, 88:1959-1965.
17. Vo Van Hoang, Odagaki T. Solid State Communications, 2010, 150:19711975.
18. Ryzhov VN, Tareyeva EE, Fomin YuD, Tsiok EN. Phys. Usp., 2019, DOI: 10.3367/UFNe.2018.04.038417.
19. Gribova NV, Fomin YuD, Frenkel D, Ryzhov VN. Phys. Rev. E, 2009, 79:051202.
20. Fomin YuD, Ryzhov VN, Gribova NV. Phys. Rev. E, 2010, 81:061201.
21. Fomin YuD, Tsiok EN, Ryzhov VN. J. Chem. Phys., 2011, 135:124512.
22. Fomin YuD, Tsiok EN, Ryzhov VN. Eur. Phys. J. Special Topics, 2013, 216:165173.
23. Dudalov DE, Fomin YuD, Tsiok EN, Ryzhov VN. Journal of Physics: Conference Series, 2014, 510: 012016.
24. Dudalov DE, Tsiok EN, Fomin YuD, Ryzhov VN. J. Chem. Phys., 2014, 141:18C522.

25. Tsiok EN, Dudalov DE, Fomin YuD, Ryzhov VN. Phys. Rev. E, 2015, 92:032110.

26. Dudalov DE, Fomin YuD, Tsiok EN, Ryzhov VN. Soft Matter, 2014, 10:4966.

27. Kryuchkov NP, Yurchenko SO, Fomin YuD, Tsiok EN, Ryzhov VN. Soft Matter, 2018, 14:2152-2162.

28. Ryltsev R, Klumov B, Chtcchelkatchev N. Soft Matter, 2015, 11:6991.

29. Fomin YuD. Physics and Chemistry of Liquids, 2019, DOI: 10.1080/00319104.2019.1698043.

30. Ryzhov VN, Tareyeva EE, Fomin YuD, Tsiok EN. Physics-Uspekhi, 2017, 60:857. DOI: 10.3367/UFNe.2017.06.038161.

31. http://lammps.sandia.gov/.