Two-Stage Melting in Quasi-Two-Dimensional Dissipative Yukawa Systems

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The results of numerical study of physical characteristics (the pair and triplet correlation functions, the isothermal compressibility, the heat capacities, and the diffusion constants) are presented for quasi-2D dissipative Yukawa systems. The specific features of these characteristics (reflecting the two-stage melting scenario) are investigated.

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The problems associated with the phase transitions in systems of interacting particles are of significant interest in various fields of science and technology. Two-dimensional (2D) melting is of theoretical and practical interest and is quality different from the liquid-to-solid phase transitions in three-dimensional (3D) systems. According to the Kosterlitz-Thouless-Halperin-Nelson-Young (KTHNY) theory based on unbinding of topological defects (dislocations and disclinations), the 2D system supports two ordered solid phases with the same packing symmetry [1–3]. In the first, both positional order and bond orientation have long range. The second (so-called hexatic phase) has the short-range positional order and the quasi-long-range bond orientation. Thus the transitions from the solid to the liquid occur with the formation of the intermediate hexatic phase. Compelling evidences for KTHNY theory in the systems with the different interparticle potentials have been presented in a set of experimental and numerical works, for example, for electrons on the surface of helium [4], for polymer colloids [5,6], and for magnetic bubbles in thin films [7,8]. One of results of these works is the fact that a point of liquid-to-hexatic phase transitions for systems analyzed has been observed near the point of hexatic-to-solid phase transitions. The difference between these points is not more than 3%–5% [4–8].

The main problem involved in studies of phase transitions in nonideal systems is associated with the absence of an analytical theory of liquid that could explain its thermodynamic properties, give the equations of state, describe heat and mass transfer, and so on. In the case of isotropic pair interparticle interactions $\phi \equiv \phi(l)$ the physical properties of simple (single atomic) liquids, such, as the pressures, P, the energy densities U, are fully determined by the pair correlation function $g(l)$ [9–11]. If both equations of state [*thermal* $P(T, n, \phi, g)$, and *calorific* $U(T, n, \phi, g)$ equations] are known, any thermodynamic characteristics can be obtained from general thermodynamic relationships. (Here l is the interparticle distance, and T, and n are the particle temperature, and density, respectively).

Dusty plasma (consisting of electrons, ions, neutral gas, and solid macroparticles of micron sizes) is a good experimental model for studying of phase transitions in nonideal systems, because, owing to their size, dust particles may be video filmed, which significantly simplifies the use of direct diagnostic methods [12–19]. Dusty plasma is ubiquitous in nature (in space, in planetary atmospheres, etc.) and often appears in a number of technological processes (for example, in processing of semiconductors). It is customary to assume that dust particles in a plasma interact with one another through the screened Coulomb potential (Yukawa type), $\phi = (eZ)^2 \exp(-l/\lambda)/l$, where λ is the screening radius, e is the electron charge, and Z is the dust charge. This type of potential is also often used for a description of interparticle interaction in medical industry, in physics of polymers, etc. [10,11]. Most experimental investigations of dusty plasma are performed in the weakly ionized plasma of gas discharges, where a dissipation of dust energy due to dust-neutral collisions can effect considerably the physical properties of systems. The nonemitting dust particles immersed in gas discharge plasma acquire negative electric charges ($|Z| \sim 10^3$–$10^5 e$) and can form the 3D or quasi-2D strongly coupled dust structures (similar to liquid or crystal). The quasi-2D dust structures, which consist of from 1 to ~10 dust layers, are typical for the plasma of radio frequency (rf) discharge [13–19]. The melting of dust structures in rf discharge arises often through the collective excitation of the rotational domain motion [17–19]; it is quite similar to the qualitative feature of 2D melting described by the KTHNY theory.

Quantitative information on the phase state of 2D systems (including criteria for phase transitions) can be obtained from analysis of the positional, or bond orientation order using the different power, or exponential approximations for spatial reducing of peaks of pair correlations function $g(l)$ or bound orientational function $g_6(l)$ [5–7,17,18]. So, for example, a power law [$g^s \propto (l/l_p)^{-\eta}$ with $\eta < 1/3$] for the slop g^s of $g(l)$ was assumed in [7] for a "perfect" (without defects) crystal, and an exponential approach $g^s \propto \exp(-\mu l)$ for the hexatic phase ($\mu = \mu_h \equiv$ const), and for the liquid ($\mu > \mu_h$). Nevertheless, an arbitrary way for the choosing of parameters for approximating functions makes these approaches hard to use as criteria for both liquid-to-hexatic, and hexatic-to-solid phase transitions [20]. A set of approaches for analysis of

the phase state of 2D systems is based on a study of dynamic behavior of particle correlation [for example, on the time dependencies for the bond-angular order parameter, $G_\Theta(t)$, or for the Lindemann parameter] [8,19,21]. It should be noted that a study of topological phase transitions is often made difficult by the fact that various small perturbations (for example, friction forces, spatial inhomogeneities, or a presence of finite boundaries) imparts some quasi-2D properties to the analyzed systems that makes the special qualitative and quantitative features of 2D phase transitions hard to recognize. So, for example, difficulties with the identification of the intermediate hexatic phase were observed with the numerical analyses of $G_\Theta(t)$ function for pure dispersive (without friction) 2D Yukawa systems [21]. The numerical study of strongly dissipative (colloidal) 2D Yukawa systems shown also that the $g_6(l)$ functions do not have power reducing with increasing of interparticle distance (predicted by KTHNY theory) [22]. Additionally, the point of liquid-to-solid phase transition (crystallization point) in these systems has been observed for the essentially lower temperatures [22] than a point of liquid-to-hexatic transition predicted in [21].

The additional possibilities for detailed experimental investigations of two-stage melting in dissipative 2D-Yukawa systems give the results presented in this Letter. Here we present the results of numerical study of physical properties (the pair and triplet correlation functions, the isothermal compressibility, the heat capacities, and the diffusion constants) for nonideal dissipative systems of macroparticles forming a monolayer for conditions close to that of dusty plasma experiments in rf discharges. The calculations were performed by the Langevin molecular dynamic method for particles in the field of gravity [balanced by linear electric field $\boldsymbol{E}(z) = \beta z$, where β is the value of gradient of $\boldsymbol{E}$] with periodic boundary conditions in two other directions ($\boldsymbol{x}$ and $\boldsymbol{y}$) for $N_p = 225$ independent macroparticles with the cut off of the pair potential equal to $8l_p$ and $\kappa \equiv l_p/\lambda = 1, 2, 4, 6$ [here $l_p = (N_p/S)^{1/2}$ is the mean interparticle distance, and S is the square of the simulated cell]. The simulation technique is detailed in [20,23,24].

The calculations were performed for various values of the effective dimensionless parameters introduced by analogy to 3D-Yukawa systems, namely, the effective coupling parameter, $\Gamma^* = \Gamma(1 + \kappa + \kappa^2/2)\exp(-\kappa)$, and the scaling factor, $\xi = \omega^*/\nu_{\rm fr}$, where $\omega^* = eZ[2(1 + \kappa + \kappa^2/2) \times \exp(-\kappa)/\pi m)l_p^{-3}]^{1/2}$ [23,24]. Here $\Gamma = (eZ)^2/(Tl_p)$ is the Coulomb coupling parameter, $\nu_{\rm fr}$ is the friction coefficient, m is the particle mass, and T is its temperature in energy units. The effective coupling parameter Γ^* was changed from 1 to 150, and the scaling factor ξ was varied from 0.04 to 4, typical for conditions of complex plasma experiments. The calculations show that the order in the system of macroparticles is practically independent on the friction ($\nu_{\rm fr}$) and on the value of the gradient of the electric field (β) and it is determined by Γ^* for weakly correlated systems ($\Gamma^* \sim 10$), as well as for stronger coupled structures up to their crystallization point, where formation of hexagonal type lattice occurs for all cases investigated. [Notice, that in the 3D-Yukawa liquids with $\kappa < 6$, the value of Γ^* also fully defines the form of the pair function $g(l)$ up to the crystallization point $\Gamma^* = \Gamma^*_{c(3D)} \approx 102$ of system, where the body centered cubic lattice is formed [23,24]]. The functions $g(l)$ are shown in Fig. 1 for different parameters of simulated systems. Some little dependence of numerical data on the ξ value has been observed close to the phase transition region only. The first maximum $g_{\max}$ of pair functions $g(l)$ and the ratio of $g_{\max}$ to the first minimum $g_{\min}$ of $g(l \neq 0)$ versus Γ^* are shown in Fig. 2. These values were averaged for varied parameters (Z, κ, $\nu_{\rm fr}$, β) under study. The numerical errors of presented data (the deviations of the $g_{\max}$ and $g_{\max}/g_{\min}$ values for different Z, κ, $\nu_{\rm fr}$, β) are less than 3%. Notice, that our results are in agreement with that for strongly dissipative ($\xi \to 0$) 2D-colloidal systems presented in Ref. [22]. One can easily see, that the $g_{\max}$, and $g_{\max}/g_{\min}$ values have two singular points. First (for $\Gamma^* \sim 65$–72) is the inflection point; it may be related to a critical point of liquid-to-hexatic phase transition. This point is close to the point of phase transition observed in dispersive Yukawa systems for $\Gamma^* \approx 74$ [21]. The second singular point (small jumps of the $g_{\max}$, and $g_{\max}/g_{\min}$ values) is observed for $\Gamma^* \sim 102$–110, and it may be related to the crystallization point of analyzed 2D systems in a solid with the hexagonal lattice (the hexatic-to-solid phase transition).

As additional illustrations of two-stage melting, we present the cross sections of the triplet correlation functions $g_3 = g_3(l_{12}, l_{23}, l_{31})$ [13] ($l_{ij} = |\mathbf{l}_i - \mathbf{l}_j|$) for a fixed value of l_{12} equal to the most probable interparticle spacing $l_{\max}(l_{12} = l_{\max})$ for different Γ^* [see Figs. 3(a) and 3(c)]. In order to represent these functions in a form convenient for comparison, they were normalized to the maximum of g_3: black corresponds to unity, and white corresponds to $g_3 = 0$. The calculation reveals that the pronounced maxima of

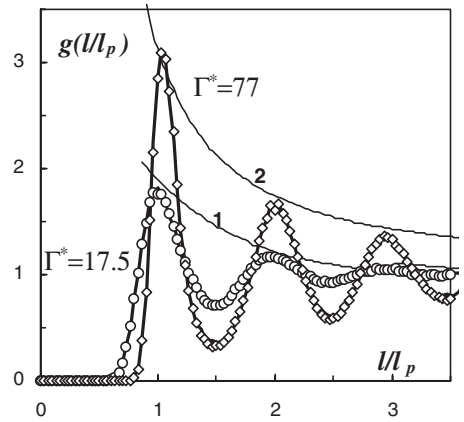

FIG. 1. Functions $g(l/l_p)$ for quasi 2D systems with different Γ^* for: (lines) $\kappa = 2$, $\xi \approx 3$; (○) $\kappa = 4$, $\xi \approx 0.12$; (◇) $\kappa = 6$, $\xi \approx 0.48$. The g^s functions with different μ: (1)1.4; (2) 0.6.

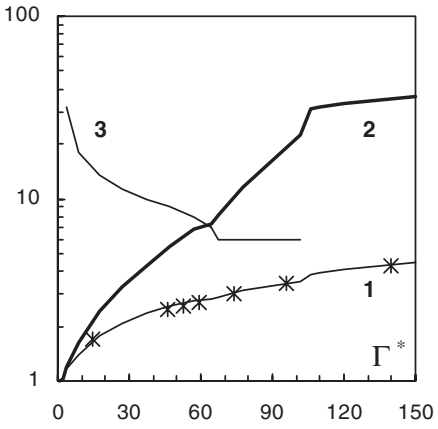

FIG. 2. Maximum g_{max} (1) and the ratio g_{max}/g_{min} (2) vs Γ^* for quasi 2D- dissipative systems; (*) the g_{max} values for 2D-colloidal systems [22]; (3) the values of 10μ.

g_3 in the system arise with the emergence of such maxima for a pair function, $g(l)$ [13]. As Γ^* increases to ~70, the maxima of g_3 increase and, at $\Gamma^* > 70$, the emergence of regular hexagonal clusters of particles is registered. With following increasing of Γ^*, the magnitude of the maxima located at distances $l \approx l_{max}$ grows, new maxima arise at sites of the hexagonal lattice, and a formation of the "perfect" crystal occurs for $\Gamma^* > 102$–110.

The obtained correlation functions $g(l)$, together with given Yukawa potential $\phi(l)$, were used to analyze the thermodynamic properties of the structures under study, namely, to calculate $P(T, n, \phi, g)$, $U(T, n, \phi, g)$, and also their derivatives such as the normalized isothermal compressibility, $\chi_T = T/(\partial P/\partial n)_T$, and the value of heat capacity $C_V = (\partial U/\partial T)_V$. The illustrations of calculated values of C_V and χ_T are presented in Fig. 4 for different parameters ξ. The calculations show that the C_V values are practically independent on the screening parameter (κ) for $\kappa \geq 1.5$ with $\Gamma^* > 10$. In doing so, the value of C_V is close to 2 for the high parameters Γ (with $\Gamma \gg \kappa$), i.e., to the theoretical value of the heat capacity for the 2D crystal of charged points. In the case of $1.5 \leq \kappa \leq 5$ with $\Gamma^* > 10$, the normalized compressibility, χ_T, outside of phase transition regions is in a good agreement with the function $\chi_T = [\Gamma\{-1.5 + 0.09367\kappa\} + 1 + 2\pi\Gamma/\kappa]^{-1}$ proposed for its approximation in [21]. It is easy to see the jumps of the values of C_V, and χ_T in the regions with $\Gamma^* \sim 65$–72 and $\Gamma^* \sim 102$–110. The first of the two singular points may be related to the liquid-to-hexatic phase transitions, the second is the point of formation of the perfect hexagonal crystal. Thus we can assume that the point of hexatic-to-solid phase transitions is observed far from the liquid-to-hexatic transitions. (The difference between these singular points is ~30%–40%). Note also that the decreasing of ξ leads to the shift of the point of hexatic-to-solid transitions to the region of higher Γ^* [see Fig. 4].

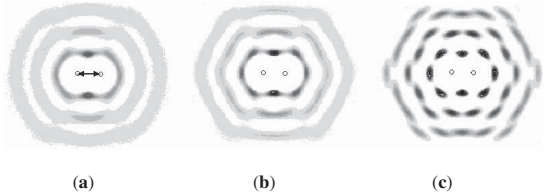

FIG. 3. The cross sections g_3 for Γ^*: (a) 35, (b) 76, (c) 105.

Transport constants (diffusion, viscosity, etc.) are fundamental parameters that reflect the thermodynamic state of systems. In diffusion measurements the ratio of mean square displacement $\langle \Delta l^2 \rangle$ to the observation time t is usually calculated. The coefficient of thermal diffusion $D = \lim_{t\to\infty} D(t)$ of particles in the 2D system can be obtained from $D(t) = \langle\langle \Delta l^2 \rangle_N \rangle_t / 4t$, where $\Delta l = |\boldsymbol{l}(t) - \boldsymbol{l}(0)|$ is the displacement of an isolated particle from its initial position $\boldsymbol{l}(0)$ at the time t, and the angle brackets $\langle\rangle$ denote the ensemble (N) and time (t) average (the average is for all time intervals t). With $t \to \infty$, the $D(t)$ function tends to its constant value D, which corresponds to the standard diffusion rate. Normalized diffusion coefficients $D^* = D(\nu_{fr} + \omega^*)m_p/T_p$ (introduced by analogy with 3D systems [23]) vs. Γ^* are shown in Fig. 5 for various parameters ξ and κ. Here we can also see two singular points: the point of inflection of $D^*(\Gamma^*)$ for $\Gamma^* \sim 70$ (the liquid-to-hexatic transitions); and the point of abrupt change of $D^*(\Gamma^*)$ with $\Gamma^* \sim 102$–110, where the diffusion coefficients $D \to 0$, and the analyzed systems transform into a solid with the hexagonal lattice. Comparison of calculations of D^* in strongly dissipative ($\xi \to 0$) 2D-colloidal systems [22] with our data is shown in Fig. 5. Thus we can conclude that the D^* function for the 2D Yukawa systems is fully determined by the coupling parameter Γ^* for weakly correlated as well as for strongly coupled systems. But we have some difference between solutions of motion equations for the case of weakly dissipative ($\xi > 0.3$) and weakly dispersive ($\xi < 0.25$) systems with $\Gamma^* > 70$.

We have to note that the abrupt changes of diffusion coefficients ($D \to 0$ for $\Gamma^* \sim 102$–110) is the well-known criterion for the crystallization point of systems [22,23]. To

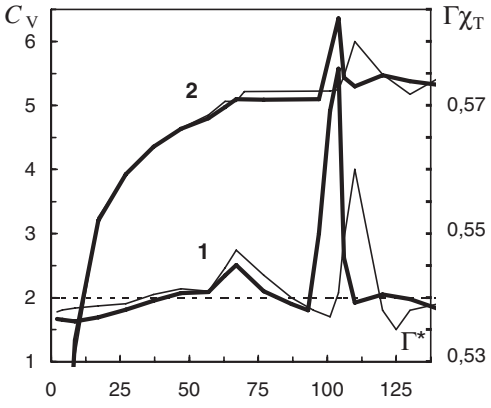

FIG. 4. Function of C_V (1) and $\Gamma\chi_T$ (2) vs Γ^* for $\kappa = 2$ and different ξ: (fine lines) −0.23; (deep lines) −1.86.

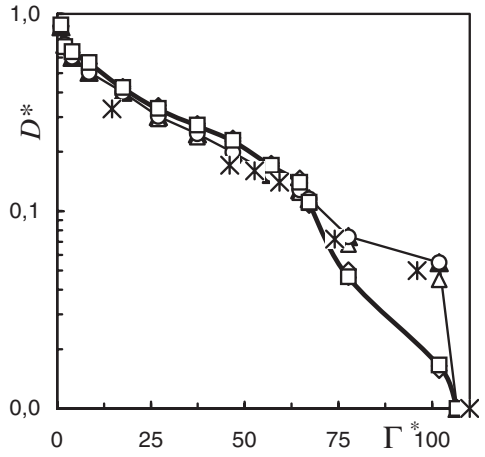

FIG. 5. The D^* values vs Γ^* for different κ: (dark symbols) 4; (transparent symbols) 2, and for different ξ: (◇,◆) 1.86; (□,■) 0.93; (○,●) 0.23; (△,▲) 0.12. Solid lines are the averaged data of D^* calculations for $\xi < 0.25$ (thin line) and for $\xi > 0.3$ (thick line); (*) are the D^* values for 2D-colloidal systems [22].

verify the detected point of liquid-to-hexatic transitions ($\Gamma^* \sim 65$–72), we have considered the often used criterion that establishes the exponential reducing [$g^s \propto \exp(-\mu l)$ with $\mu = \mu_h \equiv$ const] for the peaks of the pair functions, $g(l)$, in the hexatic phase (for the liquid $\mu > \mu_h$; see above). An illustration of reducing pair correlation in simulated systems together with the approximation of the $g(l)$ slope (obtained by the best fitting) by functions $g^s = 1 + (g_{\max} - 1)\exp\{-\mu(l - l_{\max})/l_p\}$ is presented in Fig. 1; here $l_{\max}$ is the $g_{\max}$ position. Coefficients μ of exponential approximations are shown in Fig. 2. We can see that the $\mu = \mu_h \cong 0.6$ is constant for Γ^* from ~70 to ~100, and the value of μ increases visibly with $\Gamma^* > 70$.

Under conditions of our calculations we have simulated a single monolayer of macroparticles. The formation of dust monolayers identical to simulated ones is often occurs in real experiments with dusty plasma in rf discharge [17–19]. In this case a presence of other forces (like the thermoforetic, or ion drag ones) in a sheath area of rf discharge is practically not influenced on structural properties of dust system; these forces may lead to a displacement of dust monolayer in vertical direction. The time fluctuations of dust charges in real experiments may lead to an increasing of their temperature (that can be registered by measurements) [25,26], but in the case of a uniform dust monolayer this process will not change the critical values of the effective coupling parameter in the points of phase transitions.

To conclude, here we have studied the physical properties of quasi 2D-dissipative Yukawa structures. We have introduced generalized dimensionless parameters responsible for the particle correlations, Γ^*, in these systems. The numerical simulations presented here shown that the physical properties of systems under study have two singular points. The first (for $\Gamma^* \sim 65$–72) is related to the liquid-to-hexatic phase transitions, the second ($\Gamma^* \sim 102$–110) is the point of hexatic-to-solid phase transitions. The presented results can be useful for detailed experimental investigations of two-stage melting in dissipative 2D-Yukawa systems.

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