Criteria of Phase Transitions in a Complex Plasma

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New empirical rules for different phase transitions (including the melting of cubic lattices and the transitions between body-centered-cubic and face-centered-cubic structures) are proposed. The arrangements of charged macroparticles in a complex "dusty" plasma are numerically investigated for the conditions of laboratory experiments on weakly ionized gas discharges.

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Complex plasmas containing large (compared with the sizes of electrons and ions), highly charged colloidal particles ("dust") recently attracted wide attention [1–4]. In the majority of laboratory experiments the grains were immersed in a weakly ionized plasma; the combined effect of interactions of grains between themselves as well as with the ambient plasma led to the formation of structures exhibiting liquidlike behavior [1], as well as crystals [1,2], clouds and voids [3], and clusters [4].

The simplest model for the three-dimensional (3D) particle interaction takes into account plasma screening and therefore the electrostatic interaction potential is of the Yukawa type

$$\phi_D = \frac{eZ}{l}\exp\left(-\frac{l}{\lambda}\right), \tag{1}$$

where eZ is the particle charge, λ is the plasma screening length, and l is the interparticle distance. It should be noted that the Yukawa-type approximation might be unsuitable for the vertical direction in those laboratory experiments where grains levitate in the sheath region with strong plasma flows forming the wake potential [5]. Nevertheless, the potential of the Yukawa type (1) can be used with very good accuracy in the horizontal direction [6], and it is especially important for the analysis of dust dynamics in a complex plasma under microgravity conditions where grains can levitate in the plasma bulk.

The nonideality is usually characterized by the coupling parameter $\Gamma = (eZ)^2/dT$ which is the ratio of the Coulomb potential energy of the particle interaction to the kinetic energy of their thermal motion (here, $d = n^{-1/3}$ is the mean intergrain distance, n is the particle number density, and the temperature T is in energy units). It is also well known that phase transitions in Yukawa systems are determined by two dimensionless parameters: Γ and $\kappa = d/\lambda$. The extensive numerical studies [7–14] demonstrate that in a Coulomb system of particles the short-range order appears for $\Gamma \gg 1$, with the critical value $\Gamma_m \approx 106$ on the melting line [10–13] [for complex plasmas, the assumption of plasma screening (1) leads to larger Γ_m]. The studies [14,15] suggest that the condition of the constant (normalized) nonideality parameter $\Gamma^* = (1 + \kappa + \kappa^2/2)\exp(-\kappa)\Gamma$ (namely, $\Gamma^*_m \cong 106$) can be used as the melting criterion for the body-centered-cubic (bcc) lattice. However, the functional dependence relating Γ and κ with the critical value $\Gamma_m = f(\Gamma, \kappa)$ is presently unknown for the transitions of face-centered-cubic (fcc) lattice into the liquid as well as for the transitions between the bcc and fcc structures. Some authors suggest various linear approximations of numerical data for different parts of the phase diagram [7,9]; these approximations usually appear as a result of the best mathematical fit, though sometimes being not fully justified physically. In this Letter, we propose the criteria of phase transitions in the Yukawa system by employing simulation data [7,9,13] obtained for systems without dissipation, as well as on the basis of new original simulations of a 3D Yukawa dissipative system, with parameters close to those in experiments on laboratory weakly ionized gas-discharge plasmas.

There are various phenomenological criteria for phase transitions in the Yukawa system used for a complex plasma. The most popular is the Lindemann criterion stating that the melting occurs when the ratio of the root-mean-square displacement Δ_0 of a particle from its equilibrium position to the average interparticle distance d achieves 0.15. Since in numerical simulations the displacement $\Delta = \sqrt{2}\,\Delta_0$ of a particle from the center of mass is usually computed, the ratio $\delta_c = \Delta/d$ on the melting line should be expected about $\sim$0.21 (for the majority of real solids $\Delta/d \sim 0.2$–0.25 at the melting point). However, various numerical simulations give for the Lindemann parameter the range from 0.16–0.19 for fcc lattices to 0.18–0.2 for bcc structures. These numbers, less than 0.21, may be related to the insufficient number of particles N_p in the modeled systems; we note that $\Delta/d \to 0.2$ with the increase of N_p for the melting of the both types of lattices [9]. Another popular criterion, proposed by Hansen and Verlet [16], defines the value of the first maximum S_1 of the structure factor in the liquid state to be less than 2.85. These numbers can also vary (from 2.5 to 3.2) for different simulations and strongly depend on the definition of the structure factor in the systems with a finite number of particles.

We obtain the condition, analogous to the Lindemann criterion, with the assumption that the average volume

of thermal fluctuations $V_{tf} \sim (\alpha\Delta)^3$ for the bcc lattice should not exceed $(1 - \pi\sqrt{2}/8)V \approx 0.32V$, where $\alpha = (4\pi/3)^{1/3}$, $V = n^{-1} \equiv (\alpha a_{W\text{-}S})^3$, and $a_{W\text{-}S} = (4\pi n/3)^{-1/3}$ is the Wigner-Seitz radius. For a stable fcc structure to exist, we have $V_{tf} < (1 - \pi\sqrt{2}/6)V \approx 0.26V$ [17]. Accounting for the possibility of counterdisplacements of particles, $V_{tf} \cong (2\alpha\Delta)^3$ (the factor 2), we find that the value of the ratio Δ/d must either exceed 0.211 ($\Delta_0/d \geq 0.15$) to melt the bcc structure or 0.198 ($\Delta_0/d \geq 0.14$) to melt the fcc lattice. The criterion for the transition between bcc and fcc structures can then be obtained with the assumption that for the change of the bcc symmetry of the lattice, the interparticle distances should exceed λ (the intergrain interaction is in this case similar to that of "hard spheres" when the formation of fcc structures is possible [18]). Thus, we have the following expression for the line of transition between the bcc and fcc structures:

$$2(1 - \pi\sqrt{2}/6)^{-1/3}\Delta_0 \approx a_{W\text{-}S} - \lambda\,, \qquad (2)$$

where $(a_{W\text{-}S} - \lambda)$ determines the effective size of the region where a displacement of one particle does not significantly influence other particles of the crystal lattice. This assumption is supported by numerical simulations [13] where the fcc structure was not formed when $a_{W\text{-}S} < \lambda$ even for $\Gamma \to \infty$. For the present simulation, the values of δ_c and Δ_0/d for various phase transitions are presented in Table I and Fig. 1; the range of κ between 5.8 and 6.8 defines the region with the triple (bcc-fcc liquid) phase transition.

New empirical rules can be formulated to determine the normalized coupling parameter $\Gamma_n = K_n \exp(-\kappa)\Gamma$ as a value close to a constant C_p at the line of different phase transitions (including the melting of cubic lattices and the transition between the bcc and fcc structures). The normalized coefficient K_n and constant C_p can be obtained from the relationship for the harmonic oscillator:

$$\Delta_0^2 = 3T/m_p\omega_c^2\,, \qquad (3)$$

where m_p is the particle mass and ω_c is the characteristic frequency of particle vibrations in a lattice. The approximation (3) takes place when $T \gg \Theta_D$ (Θ_D is the Debye temperature) and the displacement Δ_0 can be characterized by the frequency ω_c not depending on the temperature. To determine this frequency, the most frequently used are the quasiharmonic [7] and/or the Einstein approximations [7,11] based on the calculations of the oscillation frequency of a particle about its equilibrium position when all other particles are fixed. For both cases, there is no analytical form for ω_c, and the results are usually additionally adjusted by the linear, quadratic, and/or cubical fits of the numerical results for various (sufficiently short) parts of the phase diagrams [7,11].

Earlier, it was demonstrated [12,13,19] that the characteristic frequencies of particle oscillations in complex dust fluids and bcc lattices are proportional to the dust-lattice wave frequencies. Thus the frequency $\omega_c = \omega_{bcc}$ for the bcc lattice can be obtained from the expression $F = (eZ)^2 \exp(-l/\lambda)\,(1 + l/\lambda)l^2$ for the intergrain force assuming that the electric fields of all particles except the nearest ones are fully compensated [14]: it is determined by the probability $8/4\pi$ of the intergrain collisions and by the derivative dF/dl at $l = d$ and is given by $\omega_{bcc} = eZ(4n/\pi m_p)\,(1/2)\,(1 + \kappa + \kappa^2/2)^{1/2}\exp(-\kappa/2)$. Substituting this expression into Eq. (3) gives $\Gamma_n = T^*$ and $C_p \cong 3\pi/(2\delta_c^2) \approx 106$ in accordance with [14,15] (here, $\delta_c \equiv \sqrt{2}\,\Delta_0/d = (1 - \pi\sqrt{2}/8)^{1/3}/2\alpha \approx 0.211$ at the melting line of the bcc structure; see Table I).

On the other hand, the assumption that for the fcc structure $\omega_c = \omega_{fcc} \propto dF/dl$ leads to $\Gamma^* = \text{const}$ on the crystallization line for lattices of both types thus contradicting the results of numerical simulations; see Fig. 2. Suitable approximation $\omega_{fcc}^2 \approx 2\alpha^3 n(eZ)^2\exp(-\kappa)\,(\kappa - \alpha)/m_p$ can be obtained for a homogeneous system with the gradient dF_Σ/dl of the sum F_Σ of the electrical forces estimated as $dF_\Sigma/dl \propto n(eZ)^2\exp(-\kappa)\,(\kappa - \alpha)$. Thus, assuming that $\delta_c = (1 - \pi\sqrt{2}/6)^{1/3}/2\alpha$ on the melting line of the fcc lattice, we find from Eq. (3) $\Gamma_n = \Gamma(\kappa - \alpha)\exp(-\kappa)$, $C_p \cong 3/(\alpha^3\delta_c^2) \approx 18.5$ (see Table I), and

$$\Gamma_m^* \approx 18.5(1 + \kappa + \kappa^2/2)/(\kappa - \alpha)\,. \qquad (4)$$

TABLE I. The ratio δ_c of the most probable displacement Δ to the mean interparticle distance d, the factors of the nonideality parameter $\Gamma = (eZ)^2 n^{1/3}/T = C_p[K_n\exp(-\kappa)]^{-1}$ on the lines of various phase transitions, as well as the coefficient C_w for the approximation $\omega_c^2 = C_w n(eZ)^2\exp(-\kappa)$ of the characteristic oscillation frequencies in bcc and fcc lattices. Here, $a = (4\pi/3)^{1/3}$, $\kappa = d/\lambda$.

Phase transition	$\delta_c = \Delta/d$	C_p	K_n	C_w
bcc → liquid	$\frac{(1-\pi\sqrt{2}/8)^{1/3}}{2\alpha}$	$3\pi/(2\delta_c^2)$	$(1 + \kappa + \frac{\kappa^2}{2})$	
	~ 0.211	~ 106		
bcc → fcc		$3\pi/(2 \times 0.27^2)$	$(1 + \kappa + \frac{\kappa^2}{2})\,(1 - \frac{\alpha}{\kappa})^2$	$\frac{4}{\pi}(1 + \kappa + \frac{\kappa^2}{2})$
	$\frac{(1-\pi\sqrt{2}/6)^{1/3}}{\sqrt{2}\,\alpha}\,(1 - \frac{\alpha}{\kappa})$	~ 64		
fcc → bcc	$\sim 0.27(1 - \frac{\alpha}{\kappa})$	$3/(0.27^2\alpha^3)$	$\kappa(1 - \frac{\alpha}{\kappa})^3$	
		~ 9.8		
fcc → liquid	$\frac{(1-\pi\sqrt{2}/6)^{1/3}}{2\alpha}$	$3/(\alpha^3\delta_c^2)$	$\kappa - \alpha$	$2\alpha^3(\kappa - \alpha)$
	~ 0.198	~ 18.5		

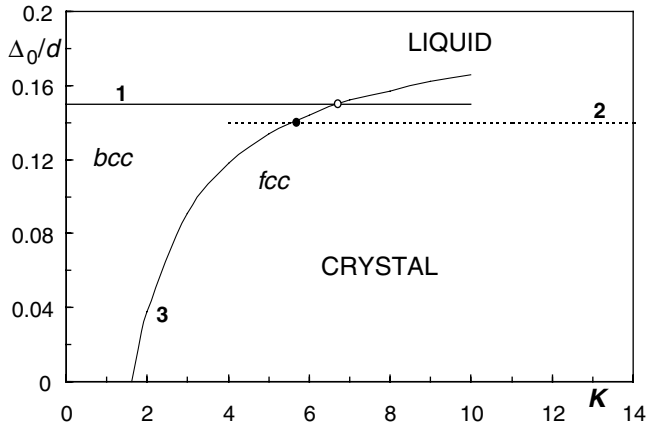

FIG. 1. Dependence of Δ_0/d on κ for various phase transitions: (1) bcc → liquid; (2) fcc → liquid; (3) bcc ↔ fcc: filled circle: $\kappa = 5.8$; circle: $\kappa = 6.8$.

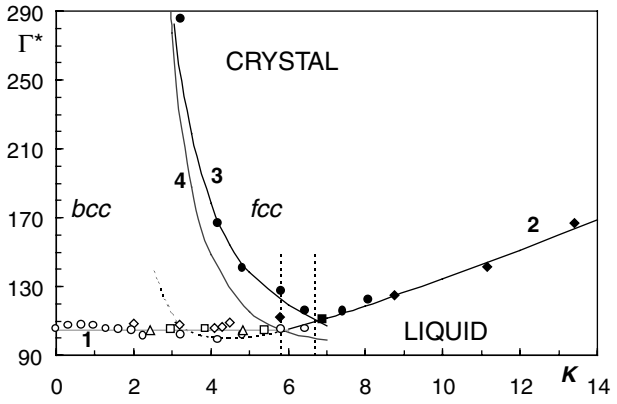

FIG. 2. Dependence of Γ^* on κ for various phase transitions: circles: data of [13]; diamonds [9]; squares [8]; triangles [15] and jump of S_1 for various ξ (filled symbols correspond to the formation of the fcc lattice); (1) $\Gamma^* = 106$ (bcc → liquid); (2) fcc → liquid, Eq. (4); (3) bcc → fcc, Eq. (5); (4) fcc → bcc, Eq. (6).

The use of the modified parameter Γ_m^* (4) allows us to illustrate the behavior of the melting curves for the transitions fcc-lattice liquid and bcc-lattice liquid (Fig. 2). The difference between condition (4) and previous results [9,13] of modeling the fcc-lattice-liquid phase transition does not exceed 2% for $\kappa > 6.8$.

The normalized coupling parameter Γ_n (see Table I) and the modified parameter Γ^* at the line of transition from the bcc to the fcc structure can be obtained from Eqs. (2) and (3) with $\omega_c = \omega_{\rm bcc}$. We have

$$\Gamma^* \approx 64\kappa^2(\kappa - \alpha)^{-2}. \quad (5)$$

Suitability of condition (5) as a criterion for the bcc-fcc transition was checked using data [13]. We obtained that deviation of the calculated values of Γ^* for the bcc-fcc transition from Eq. (5) is within $\pm 2\%$, as illustrated in Fig. 2 (curve 3). Taking into account that the possibility of the reverse transition from the fcc-to-bcc structure is defined by the frequency $\omega_{\rm fcc}$, as a criterion for this transition we can use

$$\Gamma^* \approx 9.8\kappa^2(\kappa - \alpha)^{-3}(1 + \kappa + \kappa^2/2). \quad (6)$$

Note that condition (6) depends on the approximation of the frequency $\omega_{\rm fcc}$ and therefore can be incorrect for small $\kappa \to \alpha$. However, calculations on the basis of (5) and (6) (curves 3 and 4 in Fig. 2) fully determine the region of the triple phase transition ($\kappa = 5.8$–6.8) and agree well with the results obtained for Δ_0/d (see Fig. 1). We note that the difference in the positions of curves described by Eqs. (5) and (6) makes it possible to explain disagreements of the numerical results on the position of the triple point in Refs. [9,13].

Since our determination of the coupling parameters on the lines of phase transitions is based on Eq. (3), their values are independent on the viscosity of the background gas. To examine this assumption, we have calculated the arrangements of grains in a complex plasma for various neutral gas pressures, dust charges and temperatures, and the plasma screening lengths ($\kappa = 2.4$ and $\kappa = 4.8$). The simulation has been performed by the Brownian dynamics method with the stochastic Langevin force taking into account the particle friction with plasma neutrals. To analyze the particle dynamics, we solve 3D equations of motion under the periodic boundary conditions, and the number of independent particles $N_p = 125$; more details are in Ref. [15]. The ratio between the particle interaction and dissipation in the system is defined by the parameter $\xi = \omega_{\rm bcc}/\nu_{fr}$, where the characteristic friction frequency is $\nu_{fr}[{\rm s}^{-1}] \cong C_v P[{\rm Torr}]/(\rho[{\rm g/cm}^3] r[\mu{\rm m}])$; here, r is the particle radius, ρ is the particle density, P is the neutral gas pressure, and C_v is a dimensionless parameter, defined by the nature of the neutral gas, e.g., for argon $C_v \cong 840$, and for neon $C_v \cong 600$. The equilibrium charge [20] of a dust particle can be written as $Z \approx C_z r[\mu{\rm m}] T_e[{\rm eV}]$, where T_e is the electron temperature, and C_z is a parameter defined by plasma components ($C_z \cong 2000$ for the majority of experiments in noble gases). Then for typical experimental conditions ($\rho \cong 4$ g/cm^3, $T_e \cong 1.5$ eV, $C_v \cong 700$, $\kappa \cong 2$), we have $\xi \approx 10^{-3}(4n[{\rm cm}^{-3}]/\pi r[\mu{\rm m}])^{1/2}(P[{\rm Torr}])^{-1}$. If the particle radius is $r = 2$ μm, the number density n is from 10^3 to 10^5 cm^{-3}, and the neutral gas pressure is from 1 to 0.01 Torr, we obtain the range $\xi = 0.04$–6.9.

To analyze the ordering of particles in the modeled system, we use the structure factor $S(q)$. The dependences of the first maxima of S_1 for $S(q)$ as well as the ratios of their positions $q = d_{S1}$ to the position $q_1 = 2\pi(\sqrt{2}\,n)^{1/3}$ of S_1 for the crystal bcc structure versus Γ^* are presented in Fig. 3 for different $\xi = 0.08$, 0.26, 0.79, 2.37, and 7.1. The analysis of these simulations demonstrates that the normalized coupling parameter Γ^* fully determines the particle correlations (formation of the long-range order as well as the short-range ordering) and can be considered

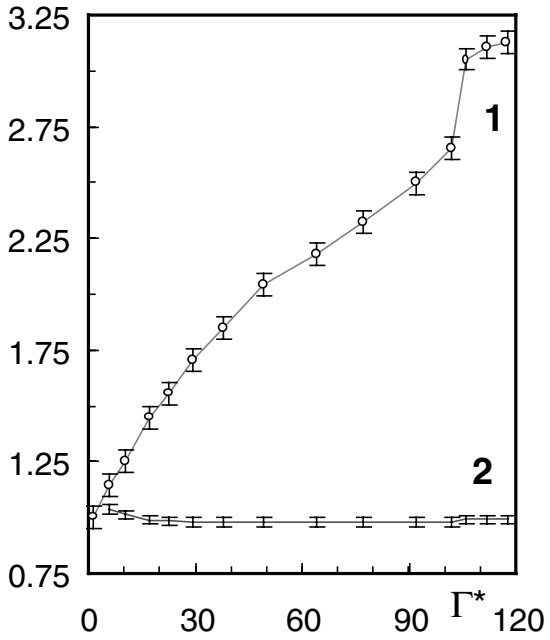

FIG. 3. Dependence on Γ^* of the first maximum of the structure factor S_1 (curve 1) and its position d_{S1}/q_1 (curve 2). We also plot here the ranges of deviations of the computed functions for various values of ξ.

as the parameter responsible for ordering in the dissipative Yukawa system when it changes from $\Gamma^* \sim 1$ up to the crystallization point. For $\kappa < 5.8$, the bcc lattice is formed with the increase of $\Gamma^* \rightarrow \Gamma_m^*$; the abrupt changes of S_1 from 2.65 to 3.1 is observed in the range from $\Gamma_c^* \cong 102$ to $\Gamma_m^* \cong 106$ (Fig. 3). We stress that $\Gamma_{cm}^* \cong 104(\pm 2\%)$ is practically independent of the viscosity of the neutral gas; that is why this criterion is in agreement with results of numerical simulations without dissipation [7,9,13] (see Fig. 2). Deviations of these results from Γ_{cm}^* are within $\pm 5\%$ and can be related to differences in the details of the numerical methods used, as well as by the particular choice of the value Γ either at the melting line or at the crystallization point of the system. We note that the obtained Γ_{cm}^* is in agreement with theoretical results [12], where for the coupling parameter on the line of the phase transition in a Coulomb system the number $105(\pm 3\%)$ was obtained, also agreeing with the results of calculations of the liquid-crystal (crystallization) [11] as well as the crystal-liquid (melting) phase transitions [10].

To conclude, we have proposed new phenomenological criteria for various phase transitions in the Yukawa system of charged macroparticles in a complex plasma. The particle dynamics was studied within the wide range of temperatures and for parameters close to the conditions of laboratory experiments in weakly ionized gas-discharge plasma. The parameter responsible for ordering of grains in the dissipative Yukawa system was obtained. The results (including criteria for phase transitions) are independent on the viscosity of the surrounding gas and can be applied for the analysis of particle dynamics in binary colloidal systems of various types where the Yukawa-type potentials are used extensively — for example, in solutions of viruses or for studies of diffusion-controlled processes in the physics of polymers.

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