

**Universal scaling in complex (dusty) plasmas**O. Vaulina,<sup>1</sup> S. Khrapak,<sup>1,2,\*</sup> and G. Morfill<sup>2</sup><sup>1</sup>*Institute for High Energy Density, RAS, 127412 Moscow, Russia*<sup>2</sup>*Centre for Interdisciplinary Plasma Science, Max-Planck-Institut für Extraterrestrische Physik, D-85740 Garching, Germany*

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The structural and dynamical properties of three-dimensional isotropic complex plasmas are investigated kinetically within the framework of a dissipative Yukawa model. A modified Coulomb coupling parameter is proposed whose value alone determines the location of the complex plasma melting line. This implies that the phase transition has a universal scaling at the kinetic level. In detail, our molecular dynamics investigations show that the system dynamics is universal (but different) in the limits of high as well as low-frictional dissipation, while in the intermediate case it depends considerably on the dissipation rate. Issues such as the influence of the interaction strength on the single particle diffusion constant and the applicability of dynamical criteria for freezing are discussed.

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Complex or dusty plasmas are multicomponent plasmas consisting of charged micron-sized particles, electrons, ions, and neutral atoms or molecules. Usually the particle charge is negative and is determined by the balance of the absorbed electron and ion fluxes. An order of magnitude estimate of the charge carried by an isolated particle is given by equating the grain surface potential (with respect to the surrounding plasma) to the electron thermal energy. For a spherical particle this gives  $Q/e \approx 10^3 a T_e$  electron charges, where  $a$  is the particle radius (in  $\mu\text{m}$ ) and  $T_e$  is electron temperature (in eV).

The interaction between highly charged particles can lead to their structural ordering, e.g., “plasma condensation” and crystallization. This was predicted by Ikezi [1]. Since then different states of complex plasmas ranging from “gaseous plasmas” to “liquid plasmas” and “plasma crystals” were experimentally observed [2–8]. The transitions between different phase states can be easily obtained experimentally by controlled changes in complex plasma parameters [9–11]. Since the dust component may be visualized and analyzed at the kinetic level, complex plasmas are recognized as valuable model systems for the study of phase transitions and other collective processes including transport properties and wave phenomena.

Most theories developed to describe the properties of complex plasmas employ the so called “Yukawa model,” where negatively charged grains are confined within the plasma reactor (due to plasma positive space charge), and interact via isotropic screened Coulomb (or Yukawa) repulsive potentials  $U(r) = Q^2 \exp(-r/\lambda_D)/r$ . The screening is determined by plasma electrons and ions and the screening length is just the corresponding Debye radius  $\lambda_D$ . This model gives a simplified picture of complex plasmas and is unsuited to some experiments, especially when gravity becomes important, as discussed later. However, it has proved useful in providing general predictions in many situations, and hence it may be considered as the basis on which one

can construct more realistic models to represent actual complex plasmas under various conditions.

Besides complex plasmas, particles interacting with a Yukawa potential have been extensively studied in different physical systems ranging from elementary particles to colloidal suspensions. Not surprisingly, phase diagrams of Yukawa systems have received considerable attention. Different numerical methods [usually Monte Carlo or molecular dynamics (MD) simulations] have been employed [12–15].

It is well recognized that the static properties of Yukawa systems are determined by two dimensionless parameters [12–14]. The first one measures the effective system temperature and is defined as  $\tilde{T} = T/m\omega_E^2\Delta^2$ , where  $T$  is a real system temperature,  $m$  is the particle mass,  $\Delta = n^{-1/3}$  characterizes interparticle spacing in terms of their number density  $n$ , and  $\omega_E$  is the Einstein frequency. Since  $\omega_E$  depends on crystal structure, the fcc Einstein frequency is commonly used for reference. The other is the so-called structure parameter  $\kappa = \Delta/\lambda_D$ , which is the ratio of characteristic spacing between particles to the screening length. This choice of parameters comes historically from the theories of colloidal suspensions. On the other hand, the ordering parameter commonly used for complex plasmas is the Coulomb coupling parameter  $\Gamma = Q^2/\Delta T$ , which is the ratio of the Coulomb energy of interaction between neighboring particles to their kinetic energy. This is because the early investigations of plasma crystallization focused on one-component plasmas (OCP), which can be considered as a limiting case  $\lambda_D \rightarrow \infty$  or  $\kappa = 0$  of the Yukawa model. The normalized temperature and the Coulomb coupling parameter are related by  $\tilde{T} = (4\pi\Gamma\omega_E^2/\omega_{pd}^2)^{-1}$ , where  $\omega_{pd} = \sqrt{4\pi Q^2 n/m}$  is the dust plasma frequency. The ratio  $\omega_E/\omega_{pd}$  is a known function of  $\kappa$  [13,16]. In Fig. 1 the phase diagram of Yukawa systems summarizing available numerical results is shown in the  $(\kappa, \Gamma)$  plane. Three phases were found, depending on the values of coupling and structure parameters. For very strong coupling ( $\Gamma > \Gamma_M$ , where subscript  $M$  denotes “melting”) there are solid fcc and bcc phases and a fluid phase for  $\Gamma < \Gamma_M$ . The bcc phase is stable at small  $\kappa$  while fcc is stable at larger  $\kappa$ . The triple point is at  $\kappa = 6.90$  and  $\Gamma = 3.47 \times 10^3$  [15].

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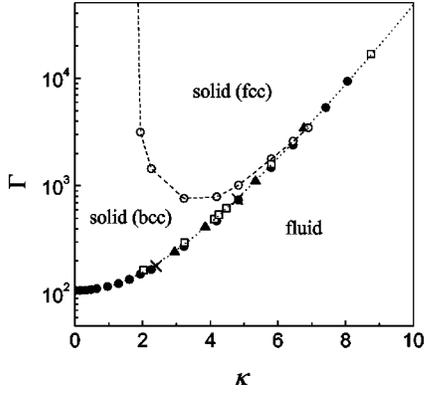


FIG. 1. Phase diagram of Yukawa systems. The symbols are: open circles, bcc-fcc phase boundary data obtained by Hamaguchi *et al.* [15]; solid triangles, open squares, and solid circles, the fluid-solid phase boundary data obtained by Meijer and Frenkel [17], Stevens and Robbins [14], and Hamaguchi *et al.* [15], correspondingly; the crosses correspond to the values of  $\Gamma$ , at which a jump of the diffusion constant occurs in presented calculations (see text). The dashed lines are their fit judged by eye.

Of particular interest for plasma crystal experiments is the form of the melting (crystallization) line  $\Gamma_M = \Gamma_M(\kappa)$  [1,13–18]. Recent results with OCP systems indicate that the crystallization condition is  $\Gamma_{\text{OCP}} \approx 106$  (or 172 if the Wigner-Seitz radius  $(4\pi n/3)^{-1/3}$  is used as the length unit instead of  $\Delta$ ) [19,20]. The corresponding normalized temperature is  $\tilde{T} \approx 0.0022$ . Several assumptions have been made in the literature concerning the behavior of  $\Gamma_M$  in case of nonzero  $\kappa$ . Starting from the OCP limit, Ikezi assumed that one simply has to take screening into account [1]. Then

$$\Gamma_M = \Gamma_{\text{OCP}} \exp(\kappa). \quad (1)$$

Another simple argument is the following: The dimensionless temperature is proportional to the mean squared particle oscillation amplitude in the quasiharmonic approximation and, according to the Lindemann criterion, should be approximately constant along the melting line. As the numerical values of the Einstein frequencies for the bcc and fcc lattices differ by less than 1% in the region where the bcc lattice is stable one may assume

$$\tilde{T}_M \approx \tilde{T}_{\text{OCP}}. \quad (2)$$

Stevens and Robbins [14] used a linear fit to their numerical data to improve this result. They obtained

$$\tilde{T}_M = \tilde{T}_{\text{OCP}}(1 + 0.1\kappa). \quad (3)$$

Finally, Vulina and Khrapak [18] proposed to use the characteristic dust lattice wave (DLW) frequency [21],  $\omega_{\text{DLW}} = \omega_{pd}(1 + \kappa + \kappa^2/2)^{1/2} \exp(-\kappa/2)$  instead of  $\omega_E$  in the determination of  $\tilde{T}$ . Using the Lindemann criterion with this normalization the dependence of  $\Gamma_M$  on  $\kappa$  becomes

$$\Gamma_M = \Gamma_{\text{OCP}} \frac{\exp(\kappa)}{1 + \kappa + \kappa^2/2}. \quad (4)$$

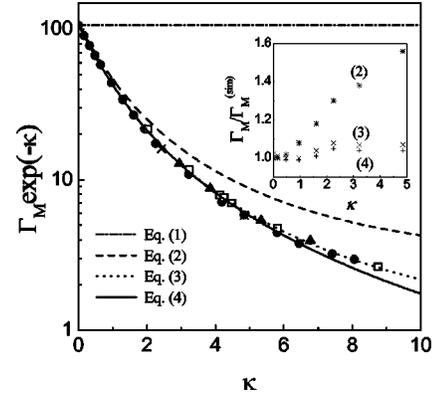


FIG. 2. Melting line of Yukawa systems: Comparison of different analytical approximations with numerical data. The symbols denote the numerical data on the fluid-solid phase boundary and are the same as those used in Fig. 1. The lines correspond to the analytical predictions given by Eqs. (1)–(4). The inset gives the ratio of  $\Gamma_M$  calculated using Eqs. (2)–(4) to simulation data  $\Gamma_M^{(\text{sim})}$  obtained in Ref. [15], in the range of  $\kappa$  relevant for complex plasmas experiments  $-\kappa \lesssim 5$  [Eq. (1) gives too poor an agreement with the simulation and the corresponding data are not shown].

The arguments presented in [18] cannot serve as a rigorous justification of Eq. (4). It is, therefore, considered as an empirical melting criterion.

All these estimates [Eqs. (1)–(4)] give the same (correct) result for  $\kappa=0$ , but have a different dependence of  $\Gamma_M$  on  $\kappa$ . A comparison of these analytical derivations with numerical simulation results is given in Fig. 2. One can see that whilst crystallization condition Eq. (1) is widely used in the literature, it is in poor agreement with simulation results. On the other hand Eqs. (3) and (4) provide good agreement. The functional form of Eq. (4) is simple and gives better agreement with numerical experiments in the range of  $\kappa$  relevant of most complex plasmas experiments ( $\kappa \lesssim 5$ ). Thus, it is useful to introduce a “modified coupling parameter”  $\Gamma^* = \Gamma(1 + \kappa + \kappa^2/2)\exp(-\kappa)$ . The crystallization condition is then  $\Gamma^* > \Gamma_{\text{OCP}}$ . In this way the location of the fluid-solid phase boundary is determined by the value of  $\Gamma^*$  alone.

The motion of dust particles in complex plasmas can be considered as a Brownian motion—modified, however, by interaction between the particles themselves. The question to be answered is, therefore, to what extent does this interaction affect the particle dynamics. Generally speaking the dynamical properties of Yukawa systems are fully determined by three dimensionless parameters as can be clearly seen by normalizing the equations of motion of particles to a dimensionless form. These are  $\Gamma$ ,  $\kappa$ , and an appropriate measure of system dissipativity, which in dimensionless form can be identified as  $\theta = \eta/\omega_d$  (in the following  $\theta$  is called *the dynamical parameter*). Here  $\eta$  represents the damping rate associated with the surrounding medium, and  $\omega_d$  is some characteristic frequency associated with the dust component. In principle, the dust-plasma frequency or the Einstein frequency may be used for normalization, however, for consistency we again use the characteristic DLW frequency for this purpose. It is shown below that this is a particularly convenient normalization.

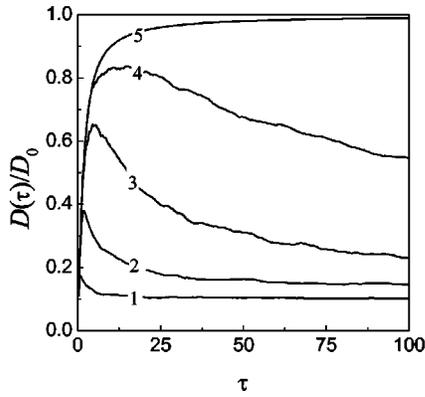


FIG. 3. The ratio  $D(t)/D_0$  in strongly interacting Yukawa systems as a function of dimensionless time for four different values of the dynamical parameter. The simulation is for  $\kappa=4.84$  and  $\Gamma=554.7$  ( $\Gamma^*=77$ ). The values of the dynamical parameter are  $\theta=0.13$  (1), 0.40 (2), 1.2 (3), 3.57 (4). Curve 5 represents the exact solution for an isolated particle (no interaction).

One of the most fundamental quantities characterizing the dynamical behavior of the system is the single particle diffusion coefficient. This coefficient is defined as  $D(t) = \langle [\mathbf{r}(t) - \mathbf{r}(0)]^2 \rangle / 6t$ , where  $\mathbf{r}(t)$  is the time-dependent particle trajectory, and  $\langle \dots \rangle$  denotes ensemble averaging. The diffusion constant is then

$$D_L = \lim_{t \rightarrow \infty} D(t).$$

(The limit  $t \rightarrow \infty$  should be treated with some care, because strictly speaking in a finite vessel the limit is zero.) Due to the interaction between the particles, the value of  $D_L$  is smaller than the bar Brownian diffusion constant for noninteracting particles  $D_0 = T/m\eta$ . In the limiting case of crystalline structure  $D_L$  tends to zero as the displacement of particles located at the lattice sites is limited. Therefore, for dissipative systems the ratio  $D_L/D_0$  appears as a natural quantity reflecting the nature and strength of the interaction potential.

Diffusion in Yukawa systems has been studied before [13,16,22–24]. The problem of self-diffusion in nondissipative Yukawa systems was considered in [13,16,22] (this corresponds to the limit  $\theta=0$  of our approach, where frictional interaction with a background medium is ignored). This approach may be adequate for atomic systems, but is not pertinent for colloidal systems and complex plasmas where viscous damping and random agitation from the medium are important. Note that the quantity  $D_L/D_0$  is meaningless in the limit  $\theta=0$ , hence the normalization  $D^* = D_L/\omega_E \Delta^2$  is used [13,16]. The relation between  $D^*$  and  $D_L/D_0$  can be expressed in terms of  $\Gamma$ ,  $\kappa$ , and  $\theta$ , of course. The interaction with the viscous medium was taken into account in [23,24], and in particular subdiffusive behavior of the time-dependent diffusion coefficient [23] and the value of the diffusion constant at the liquid-solid phase boundary [24] were addressed.

Here we report a systematic evaluation of the diffusion coefficient of strongly coupled dissipative Yukawa systems by means of MD simulations. The equations of motion in-

corporating the interaction between the particles through Yukawa potential and the interaction with the medium were solved numerically in three-dimensional (3D). The interaction with the medium was represented by the Langevin force, consisting of two terms, of which one describes systematic friction, and the other is the random force describing the stochastic action of the medium. The latter is usually associated with individual collisions with neutral atoms or molecules, but in principle can have other origins such as plasma electric field fluctuations, charge fluctuations etc. This random term was modeled by a  $\delta$ -correlated Gaussian white noise whose amplitude determined the system temperature. A cubic simulation box with periodic boundary conditions was used to emulate a large system and to reduce the influence of boundary effects. In order to reduce the computational time, the potential of interaction was cut off at distances exceeding  $3.75\Delta$ . This does not lead to considerable errors at  $\kappa \geq 1$  [13,15]. The diffusion coefficient was determined by appropriate averaging over particle trajectories.

In usual laboratory experiments the ionization fraction is very low,  $\alpha_i \sim 10^{-6} - 10^{-7}$ . Hence friction should be mostly due to dust-neutral collisions and the damping rate is given by the Epstein expression [25]. For typical dust parameters  $a \sim 1 \mu\text{m}$ ,  $m \sim 10^{-11} \text{g}$ ,  $Q/e \sim 10^3$ , and  $n \sim 10^5 \text{cm}^{-3}$ , and for typical bulk plasma parameters  $T_i \sim 0.03 \text{eV}$ ,  $n_i \sim 10^8 - 10^9 \text{cm}^{-3}$ , and  $\lambda_D \sim \lambda_{Di}$ , one obtains the range for  $\Gamma^* \sim 30 - 180$  and  $\kappa \sim 1.5 - 5$ . In the neutral gas pressure range 0.01–0.1 Torr the dynamical parameter is  $\theta \sim 0.04 - 1$ . Based on these estimates, the simulations were performed for two values of structure parameter  $\kappa = 2.42$  and  $\kappa = 4.84$  and for a set of values of dynamical parameter  $\theta = 0.044, 0.13, 0.4, 1.2$ , and 3.6. The coupling parameter was varied over a wide range.

Figure 3 shows the typical time dependence of the ratio  $D(t)/D_0$  for four different values of  $\theta$ . This behavior is compared with the exact analytical solution of a single particle Langevin equation which gives  $D(\tau)/D_0 = 1 - [1 - \exp(-\tau)]/\tau$ , where  $\tau = \eta t$  is dimensionless time. In all cases the particle motion is ballistic at short times  $D(t) \propto t$ . When the interparticle interactions are present, the diffusion coefficient reaches a maximum value at a few  $\tau$  and then decreases monotonically approaching  $D_L$  at large  $\tau$ . Similar intermediate subdiffusive regimes indicating strongly correlated motion were previously reported for nondissipative as well as dissipative Yukawa systems [13,23]. From Fig. 3 it follows that increasing friction enhances the subdiffusive regime, in agreement with [23].

Our main results are summarized in Fig. 4 which shows the behavior of the diffusion constant as a function of the interaction strength. As the coupling increases the diffusion constant decreases. At some point it experiences an abrupt jump—decreases by several orders of magnitude in the very narrow range of  $\Gamma^*$ . This jump takes place at  $102 < \Gamma^* < 105.5$  and is indicated in Fig. 4. This is very close to the value needed for crystallization  $\Gamma_M^* \approx 106$  as discussed above and shown in Fig. 1. Hence, the jump in the diffusion constant marks the first-order phase transition (crystallization).

Figure 4 also shows that the value of  $D_L/D_0$  is completely determined by the parameters  $\Gamma^*$  and  $\theta$  and is not

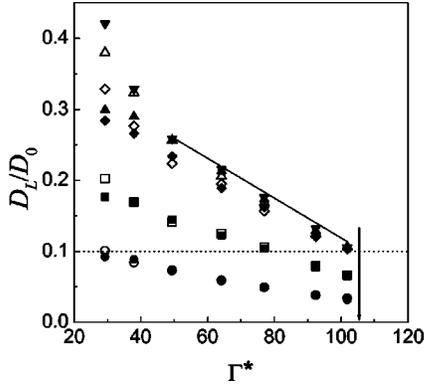


FIG. 4. The ratio  $D_L/D_0$  for strongly interacting Yukawa systems as a function of the modified coupling parameter  $\Gamma^*$  for different values of  $\theta$ . Solid symbols correspond to  $\kappa=2.42$  while open symbols correspond to  $\kappa=4.84$ . The values of the dynamical parameter are  $\theta=0.044$  (circles), 0.13 (squares), 0.4 (diamonds), 1.2 (triangles), 3.6 (inverted triangles) [only shown for  $\kappa=2.42$ ]. The solid line denotes a simple linear approximation of the numerical data in the limit of large  $\theta$  (see text). The dotted line corresponds to  $D_L/D_0=0.1$ . The arrow at  $\Gamma^*=105.5$  marks the point at which the diffusion coefficient has decreased abruptly.

separately dependent on  $\kappa$ . Moreover, if the dissipation rate is not too small, i.e.,  $\theta \gtrsim \theta_{cr} \approx 0.5$ , the ratio of  $D_L/D_0$  in strongly coupled complex plasmas becomes practically independent of  $\theta$  as well. In the range  $50 \leq \Gamma^* < \Gamma_M^*$  we find a simple linear dependence

$$D_L/D_0 = \epsilon(1 - \Gamma^*/\Gamma_M^*) + \delta, \quad (5)$$

with the parameters  $\epsilon \approx 0.3$  and  $\delta \approx 0.1$ . Figure 4 shows that the numerical results tend to this approximate expression (solid line) as  $\theta$  increases.

At lower-dissipation rates ( $\theta \lesssim \theta_{cr}$ ) the diffusion constant becomes dependent on  $\theta$  and the values of  $D_L/D_0$  decrease. It is instructive to compare our results with those obtained in the limiting case of nondissipative Yukawa systems. Recently Ohta and Hamaguchi [16] used the following expression to fit their numerical data:

$$D^* = \alpha(\Gamma_M/\Gamma - 1)^\beta + \gamma, \quad (6)$$

with  $\alpha=0.0132$ ,  $\beta=1$ , and  $\gamma=0.00317$  [16]. The comparison of our numerical simulations with this approximation is shown in Fig. 5. It is clear that with decreasing  $\theta$  the values of  $D^*$  tend towards the fit of Ref. [16]. Therefore, the behavior of the diffusion constant is universal in the limits of high as well as low-system dissipativity, and the distinction is given only by the value of parameter  $\theta$ .

Using the simulation data we can speculate about the applicability of a so-called dynamical criterion for freezing. This simple criterion, close in spirit to the Lindemann criterion, was proposed by Löwen *et al.* [24]. It states that the ratio  $D_L/D_0$  has a universal value of approximately 0.1 along the fluid freezing line and is suggested to be the only freezing criterion that holds simultaneously in three and two dimensions [26]. It was confirmed by Brownian dynamics simulations of highly dissipative Yukawa systems as well as

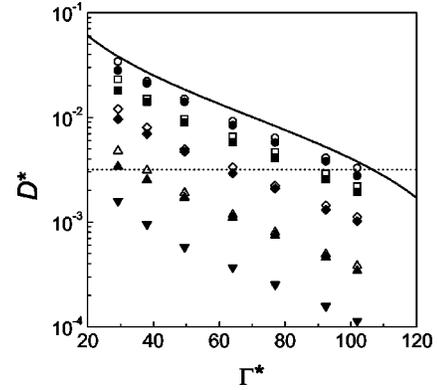


FIG. 5. Normalized diffusion constant  $D^*$  as a function of  $\Gamma^*$ . The symbols are the same as those used in Fig. 4. The solid line corresponds to the scaling proposed by Ohta and Hamaguchi [16] in the limit  $\theta=0$ . The dotted line corresponds to  $D^*=0.00317$ .

by forced Rayleigh scattering experiments on charge-stabilized colloidal suspensions [24]. We have marked the line  $D_L/D_0=0.1$  in Fig. 4. Note that as  $\Gamma^*$  approaches  $\Gamma_M^*$ ,  $D_L/D_0$  tends to this value in the limit of strongly dissipative systems, whereas it is lower for systems with lower dissipation. Thus our numerical simulations support this criterion only for highly dissipative systems  $\theta \gtrsim \theta_{cr}$ . Hence we conclude that the dynamical criterion for freezing is indeed well applicable for (strongly damped) colloids or suspensions, but may fail when applied to complex plasmas (where the background friction is many orders of magnitude smaller than in colloids). Figure 5 shows that one may think of constructing a dynamical criterion for freezing also in a case  $\theta \rightarrow 0$ . The results of Ohta and Hamaguchi suggest that it could be formulated in terms of  $D^*$ , with  $D^* \approx 0.0032$  at the fluid crystallization line. Our present results support this suggestion (see Fig. 5).

Finally, we make some comments on the applicability of Yukawa systems as models of real experimental complex plasmas. The direct application of our results is restricted to the situation of dust structures formed in a bulk isotropic plasma. In most ground-based experiments, however, the particles can be supported against gravity only in a thin boundary layer, e.g., the sheath region above the electrodes. Strong vertical confinement then prevents the formation of large 3D dust structures (see, however, [8]). In addition, the sheath region is characterized by strong anisotropy and supersonic ion flow. Consequently, the interaction between particles is strongly anisotropic: wake effects [27] and particle polarization [28] may be important. The confinement profile may also play some role, especially near the dust cloud boundary.

Even in an isotropic homogeneous plasma the interaction potential may be different from the Yukawa form due to the violation of plasma equilibrium associated with the continuous absorption of electrons and ions on the particle surface. Not only may the long-range asymptotic of the potential be different, but also a long range attractive interaction between a pair of particles—the so-called “shadow force” is possible [29,30]. Nevertheless, there exists some experimental [10,31] and theoretical [32] evidence that a Yukawa type

potential is a reasonable assumption for an isotropic plasma, at least at distances less than a few Debye lengths (which holds for most of the complex plasma experiments). Hence we conclude that Yukawa models are most suited for complex plasmas under microgravity conditions and experiments with fine powders.

Our major results may be formulated as follows. A simple form of the crystallization condition in terms of a modified coupling parameter was introduced  $\Gamma^* > \Gamma_{\text{OCP}}$ . This form is in good agreement with the simulation results of different authors. The diffusion in strongly coupled Yukawa systems was studied by means of MD simulations. It was found that in the case of high dissipation ( $\theta \gtrsim \theta_{\text{cr}}$ ) the ratio  $D_L/D_0$  behaves universally: it depends only on the value of  $\Gamma^*$ . Also, near crystallization  $D_L/D_0 \approx 0.1$ , in agreement with the dynamical criterion for freezing [24]. The critical value of the dynamical parameter was found to be  $\theta_{\text{cr}} \approx 0.5$ . In the

limit of low dissipation,  $\theta \rightarrow 0$ , the behavior of  $D_L/D_0$  is again universal and our results approach to those of Ohta and Hamaguchi for non-dissipative Yukawa system [16]. In the transition region the diffusion constant also depends on  $\theta$  itself and no obvious dynamical criterion for freezing exists.

These results are of fundamental interest and will be useful both for analyzing recent experimental data on particle diffusion and for developing methods of complex plasmas diagnostics. The universal behavior of the single particle diffusion constant, which can be easily determined experimentally, makes this idea rather attractive. Especially, the presented results may be of importance in understanding the structural and dynamical properties of 3D complex plasma clouds in experiments under microgravity conditions.

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