Molecular Dynamics Studies of Solid–Liquid Phase Transition in 2-D Yukawa Systems

Péter Hartmann, Zoltán Donkó, Pradip M. Bakshi, Gabor J. Kalman, and Stamatios Kyrkos

***Abstract*—We present systematic studies aimed at investigating the precise details of solid–liquid phase transition in 2-D classical many-particle systems interacting with the Yukawa potential. This is done by introducing and analyzing a variety of indicators, such as the bond angular order parameter, the angular distribution of the Einstein oscillations, local angular correlations, global positional correlations, and the variation of internal energy in the vicinity of the melting temperature. Our results consequently show rapid changes around $\Gamma = 415$ for $\bar{\kappa} = 2$ of the investigated quantities.**

***Index Terms*—Molecular dynamics (MD) simulation, phase transition, strongly coupled plasma, 2-D Yukawa system.**

I. Introduction

STRONGLY coupled plasma layers can be created in complex (dusty) plasma experiments [1] and charged colloidal suspensions [2]. The dust particles tend to develop crystalline structures at low enough temperatures and a liquidlike behavior at intermediate temperatures. However, it has been shown theoretically that an exact long-range order cannot survive in 1-D and 2-D systems at finite temperatures $T > 0$ [3]–[6]; thus, infinite single crystals do not exist in 2-D.

The interparticle interaction between the dust particles in a single layer can be modeled by a Yukawa-type potential (energy) $\Phi(r) = (Q^2/r)e^{-\kappa r}$, where Q is the charge of one particle. We further assume equal mass and charge for all particles. This model system can fully be parameterized by two dimensionless quantities, namely 1) the Coulomb coupling parameter $\Gamma = \beta(Q^2/a)$ and 2) the screening parameter $\bar{\kappa} = \kappa a$, where $\beta = 1/k_B T$, T is the temperature, $a = (\pi n)^{-1/2}$ is the 2-D Wigner–Seitz radius, and n is the particle number surface density. In the following, we use a as the length unit (e.g., $\bar{r} = r/a$) and $\omega_0 = \sqrt{2\pi Q^2 n/ma}$ as the frequency unit, where m is the mass of a particle.

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A. Molecular Dynamics (MD) Simulation

The numerical simulation is based on the MD method [7], using a rectangular simulation box and periodic boundary conditions. The exponential decay of the Yukawa interaction potential makes it possible to introduce a cutoff radius $R_{\rm cut}$. Only particle pairs separated by less than $R_{\rm cut}$ are taken into account in the force calculation in the solution of Newton's equation of motion. $R_{\rm cut}$ is determined by the screening parameter κ and defined to produce a relative error of $< 10^{-9}$ in the force calculation. At the beginning of the simulations, the particles are situated at triangular lattice sites with random velocities sampled from a Maxwellian distribution. After an initialization period, where the velocities are continuously scaled back to maintain the prescribed average value until the system reaches its stationary state, the particles move without any thermostation. Our computer experiments are performed in this second "measurement" phase of the simulation, where the total energy is conserved (with a relative error of $< 10^{-6}$) and the instantaneous temperature (calculated using the per-particle average kinetic energy $\langle (1/2)mv^2 \rangle = k_B T$) fluctuates around its equilibrium value.

To scan the temperature range of interest, we have carried out identical simulations for several different Γ values. Results are presented in the parameter range of $\Gamma = 390 - 435$ for $\bar{\kappa} = 2$. The number of particles in the simulations is $N = 63\,360$. At this system size, the standard deviation of the statistical fluctuation of the measured Γ value is $\sigma = \pm 0.3\%$.

B. Computed Quantities

To study the nature of solid–liquid phase transition, we have observed a variety of quantities that can be expected to be good indicators of the level of order in the simulated systems. These quantities are given as follows:

1) *Bond-Order Parameter:* Originally introduced by Halperin and Nelson [8] and is given by

$$G_\Theta = \frac{1}{N}\left|\sum_{k=1}^{N}\frac{1}{6}\sum_{m=1}^{6}\exp(i6\Theta_{k,m})\right| \qquad (1)$$

where k runs over all particles of the system and m runs over the neighbors of the kth particle. $\Theta_{k,m}$ is the angle between a predefined (e.g., x) direction and the vector connecting the kth and mth particles. This parameter is constructed to give a "1" for the perfect triangular lattice and a "0" for random configurations.

2) *Einstein Frequencies:* The normal modes of oscillations of a test charge in the presence of a given (static) distribution of charges [9]. To obtain statistical data of the oscillation frequencies and polarizations, we have constructed histograms based on a few hundred temporally uncorrelated particle configurations. For the raw data, one has to calculate the harmonic matrix for every particle for a given configuration, i.e.,

$$H_{\alpha\beta}^{(i)} = \sum_{j\neq i}^{N} \frac{\partial^2 \Phi\left(|\mathbf{r}_i^{\mathrm{eq}} - \mathbf{r}_j|\right)}{\partial r_{i\alpha} \partial r_{i\beta}} \tag{2}$$

where $\mathbf{r}_i^{\mathrm{eq}}$ is the equilibrium position of the ith particle (local minimum of the potential surface), $\Phi(r)$ is the interaction potential, and α and β represent the Cartesian coordinates. The eigenvalues of $H_{\alpha\beta}$ are the squared Einstein frequencies (two for every particle), while the eigenvectors provide the polarization of the oscillation.

3) *Disorder-Related Energy:* A global measure of the level of disorder in the system compared to the perfect lattice state.

The total energy of the system is the sum of the kinetic K and potential V contributions, i.e.,

$$E = K + V = \sum_{i=1}^{N} \frac{1}{2} m v_i^2 + \sum_{i=1}^{N-1} \sum_{j>i}^{N} \Phi\left(|\mathbf{r}_i - \mathbf{r}_j|\right). \tag{3}$$

The "excess" potential energy is $V_{\mathrm{exc}} = V - L$, where L is the ground state energy of the perfect triangular lattice [$L = 0.0365964(Q^2/a)$ per particle for $\bar{\kappa} = 2$, as calculated by numerical lattice summation]. Based on the theorem of equipartition, in the harmonic approximation, $\langle V_{\mathrm{exc}} \rangle = \langle K \rangle$, where $\langle\rangle$ denotes the time and ensemble average value per particle. The harmonic approximation is valid only for very low temperature systems, where the oscillation amplitude of the particles is low enough. Near the melting transition,

$$W = \frac{\langle V_{\mathrm{exc}} \rangle - \langle K \rangle}{\langle K \rangle} \tag{4}$$

is expected to deviate significantly from zero, due to the increasing disorder in the particle distribution and anharmonicity of the particle motions.

4) *Long-Range Pair Correlation:* To measure positional long-range correlations, we have computed the $g(r)$ pair-correlation function (PCF) in the range of $0 \leq \bar{r} \leq \bar{r}_{\max}$ (which is limited by the size of the simulation box and the periodic boundary condition; in our case, $\bar{r}_{\max} = 120$). To quantify the strength of the positional correlations, we have tentatively introduced the quantity

$$G = \int_0^{r_{\max}} \left[g(r) - 1\right]^2 dr. \tag{5}$$

We expect that this quantity (aside from its significant dependence on $r_{\max}$ and the short-range structure) is able to show rapid changes in the long-range tail of the PCF (for large enough $r_{\max}$, the short-range contribution becomes less important).

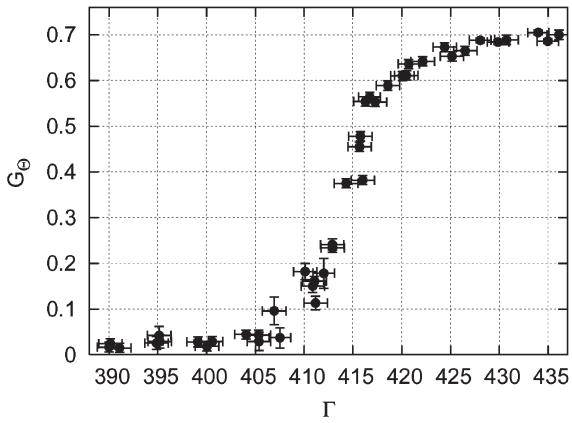

Fig. 1. Bond-order parameter versus Γ for $\bar{\kappa} = 2$.

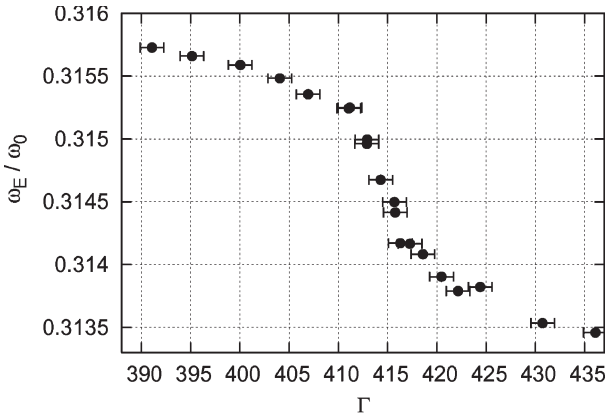

Fig. 2. Average of Einstein frequencies versus Γ for $\bar{\kappa} = 2$.

II. Results of the Simulation

In this section, we present the study of the dependence of the quantities described previously on the measured coupling parameter Γ in the vicinity of the expected solid–liquid phase transition. The error-bars, where available, indicate the standard deviation of the fluctuating quantities.

The bond-order parameter G_Θ (Fig. 1) shows a rapid change from $G_\Theta < 0.05$ to $G_\Theta < 0.6$ in the range of $\Gamma = 407 - 418$ with an inflection point around $\Gamma = 414$.

The average value of the Einstein frequencies [$\omega_E = \langle \omega^2 \rangle^{1/2}$ (Fig. 2)] also shows significant dependence on Γ. In the solid phase, lower frequencies are expected, due to the fact that the triangular lattice is energetically the "best" configuration in 2-D, where the interparticle distances are maximized. Consequently, the average steepness of the potential surfaces around the local equilibrium positions of particles is minimized. The inflection point of the plotted curve is around $\Gamma = 415$.

Fig. 3 shows the distribution of the polarization angle of the first (higher frequency) normal mode in a polar plot for several values of Γ. The polarization of the Einstein oscillations also depends on the underlying structure. The perfect triangular lattice is isotropic to second order in the expansion of

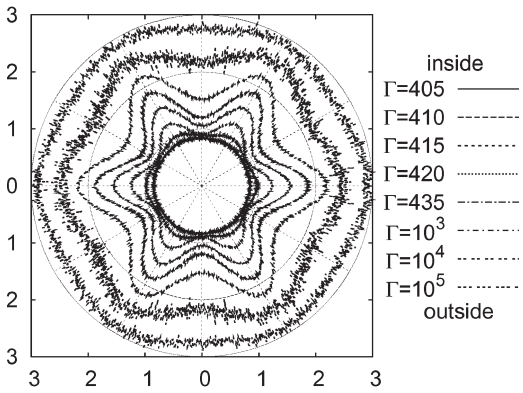

Fig. 3. Distribution of the polarization angle of the higher frequency mode for several values of Γ for $\bar{\kappa} = 2$. The curves are scaled for better visualization, the units are arbitrarily chosen, and there are zero-angle points in the direction of the first neighbor in the lattice (x-axis).

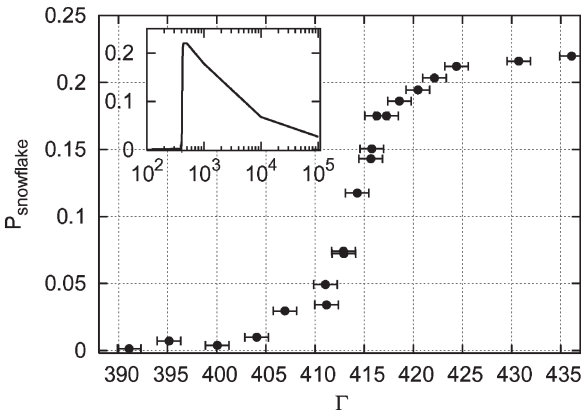

Fig. 4. Modulation strength of the angular distribution of Einstein oscillations versus Γ for $\bar{\kappa} = 2$. The inset shows the slow decay of P for large Γ values.

$H_{\alpha\beta}$ (harmonic approximation; outer curve approximated with $\Gamma = 10^5$). The liquid configuration, on the other hand, is also isotropic, on average, due to the lack of any preferred direction (inner curve with $\Gamma = 405$). At intermediate states (starting just below the freezing temperature), the disordered solid structure introduces strong anisotropy (see also the inset of Fig. 4).

The angular dependence of the distribution of the polarization shows hexagonal symmetry (like snowflakes) and can be approximated in the form $y = h\cos(6\varphi) + y_0$. The modulation strength is therefore $P = h/y_0$ (the so-called "snowflake amplitude"). Fig. 4 shows the dependence of P on Γ. The center of the transition shows up around $\Gamma = 414$, which is similar to the previously introduced quantities.

Fig. 5 shows the disorder-related energy, as introduced in (4). W is expected to reach 0 in the $\Gamma \to \infty$ limit. The center (inflection) of this transition is slightly shifted to higher Γ values around the range of 415–416.

Fig. 6 shows quantity G derived from the $g(r)$ PCF [see (5)]. This quantity exhibits weak Γ dependence in the liquid phase (for more information about the structural properties of the liquid phase, see, e.g., [10] and [11]) and somewhat stronger Γ dependence in the solid phase. These two branches are connected by a steep intermediate region featuring an inflection around $\Gamma = 417$.

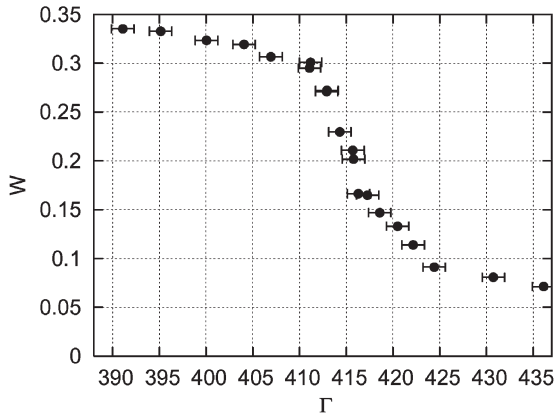

Fig. 5. Disorder-related energy (4) versus Γ for $\bar{\kappa} = 2$.

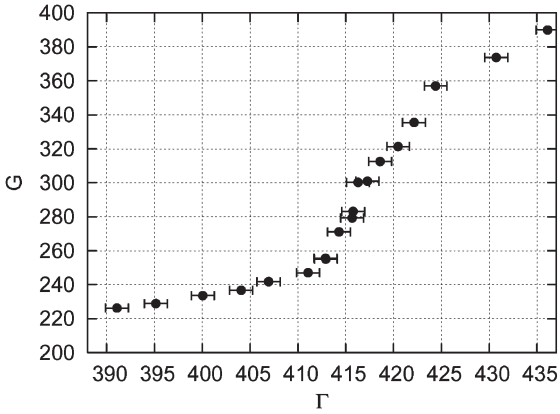

Fig. 6. Long-range pair-correlation strength (5) versus Γ for $\bar{\kappa} = 2$. The units of G are arbitrary, and proper normalization is needed for the final definition of this preliminary quantity.

The particle-number dependence of these results is investigated by applying the same measurements to three different systems with $N = 63\,360$, $N = 15\,840$, and $N = 3960$. Comparative plots are shown for the bond-order parameter in Fig. 7. The standard deviation of the fluctuating quantities (such as the measured Γ parameter) shows strong N dependence, which can be approximated by $\sigma \propto N^{-1/2}$. On the other hand, no significant change in the slope of the curves can be observed in the transition region.

III. Conclusion

We have performed equilibrium analysis of the solid–liquid phase transition in 2-D Yukawa systems. Results are shown for $\bar{\kappa} = 2$ systems. All the quantities studied show a pronounced transition in the $\Gamma = 410 - 420$ interval. The central value ($\Gamma_m = 415$) corresponds to an effective Coulomb coupling parameter ([10], [11]) $\Gamma_m^* = 137.5$ in very good agreement with the widely accepted experimentally obtained value of $\Gamma_m = 137 \pm 15$ [12].

Due to the finite size of the system, we cannot determine whether the continuous changes observed are indicative of the order of the phase transition or are caused by the finite size itself. On the other hand, the behavior in the transition domain according to our present studies does not show significant

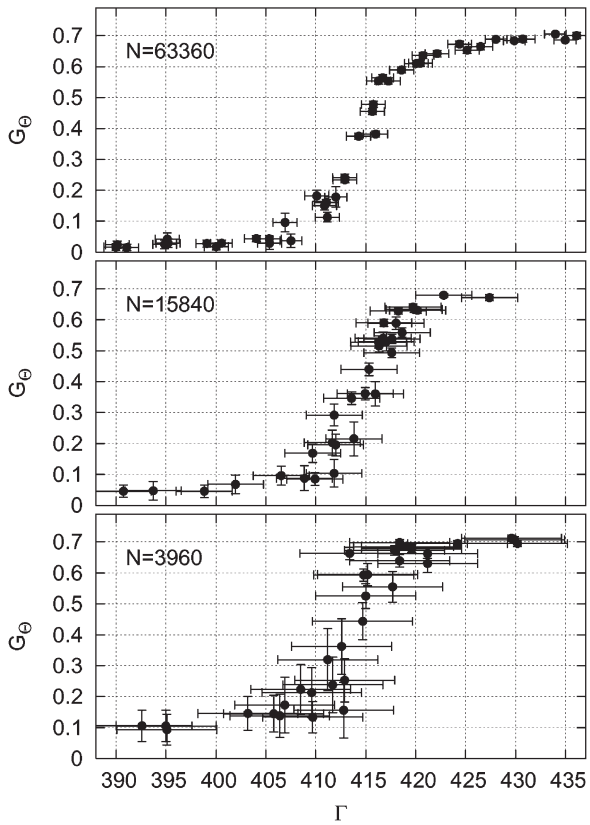

Fig. 7. System-size dependence of the bond-order parameter.

dependence on the number of particles: This makes extrapolation to the thermodynamic limit very uncertain.

Theoretical investigations (see, e.g., [8] and [13]) and some numerical studies of the melting transition in 2-D systems indicate the appearance of two melting stages. According to this theory, as the system is heated, it first transforms from solid to the so-called "hexatic" phase, where the long-range positional order is suppressed, but orientational order still survives. At somewhat higher temperatures, the orientational order also disappears, and the system enters the liquid phase. This issue of the number of distinct phases in relation to the 2-D Coulomb or Yukawa systems has been, for some time, a matter of intense controversy [14]–[18].

Our work provides some additional insight by studying quantities that can be characterized by their degree of dependence on orientational or positional order. In particular, *bond-order parameter* G_Θ is very sensitive to the orientational order but does not really depend on the positions. The same goes for *polarization of the Einstein oscillations* P (the snowflake amplitude). *Disorder-related energy* W and average *Einstein frequency* ω_E are expected to depend on both orientational and positional orders. On the other hand, *long-range pair-correlation strength* G is, by definition, an angularly averaged quantity, so it depends purely on positional order. Approximating the "exact" transition temperature (or Γ_m) with the inflection point of the measured curves, the obtained values show slight differences for the three groups of quantities, namely i) $\Gamma_m(G_\Theta, P) \approx 414$, ii) $\Gamma_m(W, \omega_E) \approx 415{-}416$, and iii) $\Gamma_m(G) \approx 417$. However, we have to add the caveat that, due to the finite (small) system size, the level of fluctuations is too high to allow one to make reliable statements about this issue; with this notwithstanding, the observed trend provides a guideline for further investigations.

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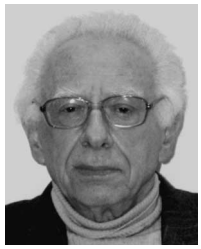

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