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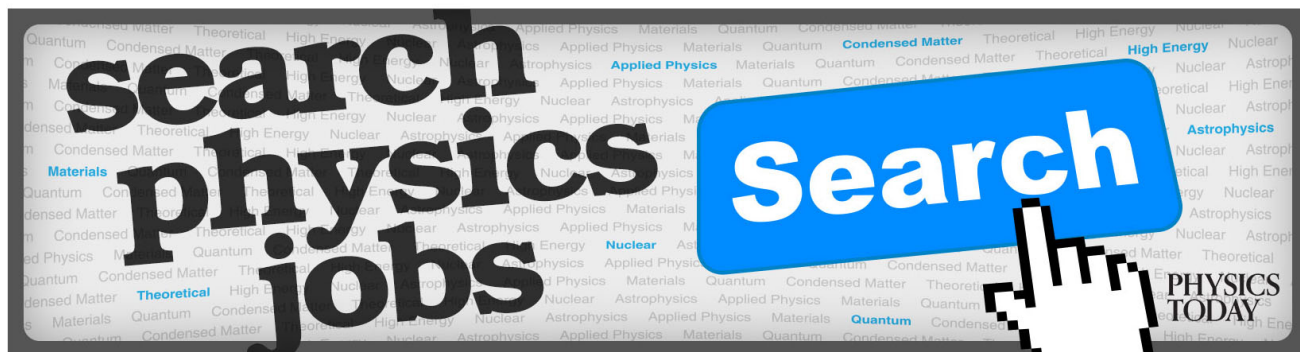
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On the long-waves dispersion in Yukawa systems

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A useful simplification of the quasilocalized charge approximations (QLCA) method to calculate the dispersion relations in strongly coupled Yukawa fluids is discussed. In this simplified version, a simplest possible model radial distribution function, properly related to the thermodynamic properties of the system, is used. The approach demonstrates good agreement with the dispersion relations obtained using the molecular dynamics simulations and the original QLCA in the long-wavelength regime. © 2016 AIP Publishing LLC. [<http://dx.doi.org/10.1063/1.4942169>]

I. INTRODUCTION

The quasilocalized charge approximation (QLCA) was originally proposed by Kalman and Golden¹ as a powerful formalism for the analysis of the dielectric response tensor and collective mode dispersion in strongly coupled Coulomb liquids. The approach is based on a microscopic model in which the charges are quasilocalized on a short-time scale in local potential fluctuations, for a review, see Ref. 2. In last decades, the QLCA approach has been successively applied to various systems of strongly interacting particles to describe wave dispersion relations. In particular, this includes two-dimensional (2D) and three-dimensional (3D) one-component-plasma (OCP),² 2D and 3D Yukawa systems, mainly in the context of complex (dusty) plasmas,^{3–7} classical 2D dipole systems,^{8,9} and 3D dusty plasma with Lennard–Jones-like interactions.¹⁰

Technically, for a given interaction potential, the QLCA approach requires the equilibrium radial distribution function (RDF), $g(r)$, as an input, characterizing the spatial order in the system of particles. The latter can be obtained via various integral equation schemes or via the direct molecular dynamics (MD) or Monte Carlo (MC) simulations. Although, there are no principle difficulties in both these approaches, they remain relatively resource consuming. The purpose of this paper is to demonstrate that to describe the long-wavelength dispersion relations in strongly coupled Yukawa fluids, the accurate knowledge of $g(r)$ is unnecessary. The main effect of strong coupling can be accounted for by using a simple excluded volume consideration. A simplest possible toy $g(r)$ allows us to reproduce the long-wave dispersion curves with a reasonable good accuracy.

In Yukawa systems, which are of some relevance in the context of colloidal suspensions and complex (dusty) plasmas,^{11–13} the particles are interacting via the repulsive potential of the form $V(r) = (Q^2/r) \exp(-r/\lambda)$, where Q is the particle charge, λ is the screening length, and r is the distance between a pair of particles. These systems are conveniently characterized by two dimensionless parameters, which are the coupling parameter $\Gamma = Q^2/aT$, and the screening parameter $\kappa = a/\lambda$. Here, T is the system temperature (in

energy units), n is the particle density, and $a = (4\pi n/3)^{-1/3}$ is the characteristic inter-particle separation (Wigner–Seitz radius). The phase behavior of Yukawa systems is relatively well understood.^{14,15} For $\Gamma e^{-\kappa} \ll 1$, a weakly coupled gaseous regime is realized. As Γ increases, the system shows a transition to the strongly coupled fluid regime. When Γ increases further, the system crystallizes either into body-centered-cubic (bcc) or into the face-centered-cubic (fcc) lattice (bcc is thermodynamically favorable at weak screening, i.e., lower κ). The values of $\Gamma_m(\kappa)$, corresponding to the fluid-crystal transition, have been tabulated;¹⁵ relatively accurate fits are also available.^{16–18} For even higher Γ , the glass transition is predicted, with the glass-transition line almost parallel to the melting line in the extended region of the phase diagram.¹⁹ In this study, we focus on the strongly coupled fluid regime, characterized by $\Gamma \lesssim \Gamma_m$.

II. QUASILOCALIZED CHARGE APPROXIMATION AND ITS SIMPLIFIED VERSION

The dispersion relations in the QLCA approach read

$$\begin{aligned}\omega_L^2 &= \omega_0^2(q) + D_L(q), \\ \omega_T^2 &= D_T(q),\end{aligned}\quad (1)$$

where $q = ka$ is the reduced wave number and the subscripts “L” and “T” stand for longitudinal and transverse modes, respectively. The term $\omega_0(q)$ corresponds to the longitudinal dispersion relation of non-correlated particles (weak-coupling limit)

$$\omega_0^2(q) = \frac{\omega_p^2 q^2}{q^2 + \kappa^2}, \quad (2)$$

where $\omega_p = \sqrt{4\pi Q^2 n/m}$ is the plasma frequency associated with the charged particle component and m is the particle mass. In the context of complex (dusty) plasmas, this mode is known as the dust-acoustic-wave (DAW).²⁰ The respective projections of the QLCA dynamical matrix $D_L(q)$ and $D_T(q)$ are the functions of the equilibrium $g(r)$.^{2,4,7}

$$D_{L/T} = \omega_p^2 \int_0^\infty \frac{dr}{r} [g(r) - 1] \mathcal{K}_{L/T}(qr, \kappa r), \quad (3)$$

with

$$\begin{aligned} \mathcal{K}_L(x, y) = & -e^{-y} \left[\left(2 + 2y + \frac{2}{3}y^2 \right) \right. \\ & \times \left(\frac{\sin x}{x} + 3 \frac{\cos x}{x^2} - 3 \frac{\sin x}{x^3} \right) \\ & \left. + \frac{1}{3}y^2 \left(\frac{\sin x}{x} - 1 \right) \right], \end{aligned} \quad (4)$$

and

$$\begin{aligned} \mathcal{K}_T(x, y) = & e^{-y} \left[\left(1 + y + \frac{1}{3}y^2 \right) \right. \\ & \times \left(\frac{\sin x}{x} + 3 \frac{\cos x}{x^2} - 3 \frac{\sin x}{x^3} \right) \\ & \left. - \frac{1}{3}y^2 \left(\frac{\sin x}{x} - 1 \right) \right]. \end{aligned} \quad (5)$$

The dimensionless distance r in Eq. (3) and throughout the paper is expressed in units of a . When $g(r)$ is known, Eqs. (1)–(5) allow us to calculate the dispersion relations of Yukawa systems in the QLCA approach.

The equilibrium RDF, $g(r)$, is also related to important thermodynamic quantities of the system such as energy and pressure. For pairwise interactions, they can be expressed in terms of the integrals over $g(r)$, which are known as the energy and pressure (or virial) equations.²¹ Since the dispersion relations and thermodynamic properties depend only on the integral of $g(r)$, it is not very unreasonable to presume that if a simple model form is chosen, which describes the thermodynamic properties of the system reasonably well, it will also allow to estimate the dispersion relations of this system. We shall now demonstrate that this is indeed a reasonable assumption, provided the long wavelengths (longer than the mean interparticle separation) are of main interest.

To further pursue the link between dispersion properties and thermodynamics, we chose the most simple step-wise toy model for $g(r)$, i.e., $g(r) = 1$ for $r > R$ and $g(r) = 0$ otherwise. Here, R characterizes an excluded volume around each particle due to strong (repulsive) inter-particle interactions. For this model $g(r)$, the integration in Eq. (3) can be performed analytically. The resulting dispersion relation of the longitudinal mode is

$$\begin{aligned} \omega_L^2 = & \omega_p^2 e^{-R\kappa} \left[(1 + R\kappa) \left(\frac{1}{3} - \frac{2 \cos Rq}{R^2 q^2} + \frac{2 \sin Rq}{R^3 q^3} \right) \right. \\ & \left. - \frac{\kappa^2}{\kappa^2 + q^2} \left(\cos Rq + \frac{\kappa}{q} \sin Rq \right) \right]. \end{aligned} \quad (6)$$

Similarly, for the transverse mode, we get

$$\omega_T^2 = \omega_p^2 e^{-R\kappa} (1 + R\kappa) \left(\frac{1}{3} + \frac{\cos Rq}{R^2 q^2} - \frac{\sin Rq}{R^3 q^3} \right). \quad (7)$$

The remaining step is to find an appropriate model for the dimensionless parameter $R(\kappa, \Gamma)$ and to verify whether the proposed simplification can deliver reasonable results.

Substituting the same step-wise RDF into the energy equation, we easily obtain the excess energy per particle in units of the system temperature

$$u_{\text{ex}} = \frac{2\pi n a^3}{T} \int_0^\infty r^2 V(r) g(r) dr = \frac{3\Gamma}{2\kappa^2} (1 + R\kappa) e^{-R\kappa}. \quad (8)$$

Similarly, for the reduced excess pressure, we get

$$\begin{aligned} p_{\text{ex}} = & -\frac{2\pi n a^3}{3T} \int_0^\infty r^3 V'(r) g(r) dr \\ = & \frac{\Gamma}{2\kappa^2} (3 + 3R\kappa + R^2 \kappa^2) e^{-R\kappa}. \end{aligned} \quad (9)$$

The effective radius of the exclusion sphere, R , can then be obtained from the solution of either Eq. (8) or (9). Naturally, since the toy model for $g(r)$ is used, the result for R somewhat depends on whether the energy or pressure route is used. The corresponding quantities will be referred to as R_u and R_p , respectively.

Thermodynamics of Yukawa systems has been extensively investigated, and the accurate data for u_{ex} and p_{ex} from MC and MD simulations exist (see, for example, Refs. 15, 22, and 23). Various fitting formulas, based on different physical arguments, have also been proposed.^{24–30} In this study, we use simple practical expressions for u_{ex} and p_{ex} applicable in a wide parameter regime characterizing Yukawa fluids, which has been published recently.³¹ These expressions are essentially based on the Rosenfeld–Tarrazona freezing-temperature scaling for the thermal component of the excess internal energy and related thermodynamic quantities for simple fluids with soft repulsive interactions.^{24,25} They demonstrate excellent agreement with the results from numerical simulations in the regime $\kappa \lesssim 5$ and $\Gamma/\Gamma_m \gtrsim 0.1$, which is addressed in this study.

The quantities $R_u(\kappa, \Gamma)$ and $R_p(\kappa, \Gamma)$ evaluated using the expressions for $u_{\text{ex}}(\kappa, \Gamma)$ and $p_{\text{ex}}(\kappa, \Gamma)$ from Ref. 31 exhibit the following properties. For a given pair of κ and Γ , R_u is slightly larger than R_p . Both R_u and R_p demonstrate slow increase as Γ and κ increase. In a relatively wide parameter regime investigated ($1 \leq \kappa \leq 4$ and $0.01 \leq \Gamma/\Gamma_m \leq 1$), the values of R are confined to a relatively narrow range $1 \lesssim R_{u,p} \lesssim 1.3$. An example of calculated dispersion relations using the simple model form of $g(r)$ is shown in Fig. 1. Here, the solid curves correspond to the full QLCA approach with the RDF obtained from direct MD simulations (see below for description), while the dashed (longitudinal mode) and dotted (transverse mode) curves correspond to the proposed simplification employing the simplest model RDF, linked to the thermodynamic properties of the system. Two important observations are as follows: (i) The simplified approach is practically insensitive to whether energy or pressure route is used to determine R , and (ii) The simplified QLCA calculation demonstrates very good agreement with the full QLCA in the regime of sufficiently long wavelength, $q \lesssim 2$ (where the first maximum of the longitudinal wave dispersion occurs).

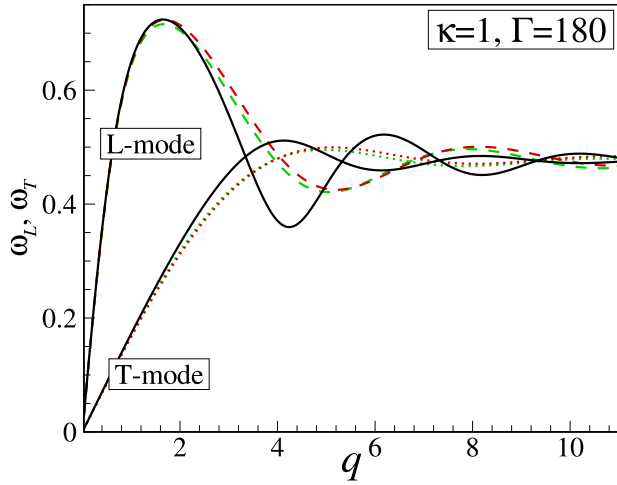


FIG. 1. The dispersion relations of the longitudinal (L-mode) and transverse (T-mode) waves in strongly coupled Yukawa fluid, characterized by $\kappa = 1.0$ and $\Gamma = 180$ (frequency is in units of the plasma frequency ω_p). The solid curves correspond to the conventional QLCA with the radial distribution function obtained via the direct MD simulations. The dashed (L-mode) and dotted (T-mode) curves correspond to the simplified version of QLCA of Eqs. (6) and (7), respectively. Red (green) color corresponds to the pressure (energy) route in determining R ; these curves are almost indistinguishable. The conventional and simplified QLCA shows very good agreement in the long-wavelength regime (for $q \lesssim 2$).

III. DETAILED COMPARISON

Further MD simulations and full QLCA calculations have been performed to verify the main conjecture of this study. Simulations have been performed on graphics processing unit (NVIDIA GTX 960) using the HOOMD-blue software.^{32,33} We used $N = 50653$ Yukawa particles in a cubic box with periodic boundary conditions. The cut-off radius for the potential has been chosen to be $L_{\text{cut}} = 14.5\lambda$. The numerical time step was set to $\simeq 10^{-2}\sqrt{ma^3/Q^2} \sim 10^{-2}\omega_p^{-1}$. Simulations have been performed in the canonical ensemble (NVT) with the Langevin thermostat at a temperature corresponding to the desired target coupling parameter Γ .

The system was first equilibrated for one and a half million time steps, and then, we saved the particle positions and trajectories every 60 time step for 80 000 time steps. The particle current was then calculated

$$\mathbf{J}(\mathbf{k}, t) = \sum_{j=1}^N \mathbf{v}_j(t) \exp(i\mathbf{k} \cdot \mathbf{r}_j(t)),$$

and the Fourier transform in time was performed to obtain the current fluctuation spectra. Moreover, the particle position was saved every 4000 time step for an extra 3×10^6 time steps to extract the accurate RDF.

Simulations have been performed for four pairs of κ and Γ , which are summarized in Table I. These points have been chosen to be located in the strongly coupled fluid state, at approximately the same distance from the melting line, $\Gamma/\Gamma_m = T_m/T \simeq 0.8$. The radial distribution functions obtained in MD simulations are shown in Fig. 2. We observe close similarity of the obtained RDFs. This observation is in line with the isomorph theory put forward recently.^{34,35} Isomorphs are curves in the thermodynamic phase diagram

TABLE I. The longitudinal (c_L) and transverse (c_T) sound velocities of strongly coupled Yukawa fluids evaluated using the QLCA approach (velocities are expressed in units of $\omega_p a$). $c_L^{u,p}$ and $c_T^{u,p}$ denote the longitudinal and transverse sound velocities estimated using the simplified QLCA with the model RDF linked to the thermodynamics via the energy and pressure route, respectively.

κ	Γ	c_L	c_L^u	c_L^p	c_T	c_T^u	c_T^p
0.5	145	1.980	1.979	1.979	0.191	0.193	0.190
1.0	180	0.959	0.953	0.956	0.175	0.174	0.171
2.0	370	0.415	0.403	0.408	0.128	0.122	0.122
3.0	990	0.212	0.202	0.206	0.081	0.076	0.076

along which many properties derived from structure or dynamics are invariant in properly reduced units. The isomorph theory has been developed for liquids, which have strongly correlated fluctuations of their energy and pressure (referred to as Roskilde-simple or just Roskilde systems³⁶). Yukawa fluids belong to this class, and it has been recently demonstrated that the state points characterized by the same T/T_m are approximately isomorphs.³⁷

Using the obtained RDFs, the dispersion of the longitudinal and transverse modes within the QLCA approach has been calculated. The results are presented in Figs. 3 and 4. Here, the color background corresponds to the spectral decomposition of the longitudinal and transverse current fluctuations. The maximum magnitude (red color) marks the approximate location of the collective excitations. The dark curves correspond to the dispersion relations calculated using the conventional (full) QLCA approach. They are in very good agreement with the current fluctuations analysis. The white curves correspond to the simplified QLCA approach discussed in the present work. In the long-wavelength regime ($q \lesssim 3$) shown in Figs. 3 and 4, the agreement with the original QLCA is excellent.

In the long-wavelength limit ($q \rightarrow 0$), both longitudinal and transverse modes exhibit the acoustic dispersion, $\omega_{L/T} \simeq c_{L/T}k$. It should be reminded here that the disappearance of the shear mode at $q \rightarrow 0$ and the existence of the corresponding cutoff wave-vector q_* , which are well known

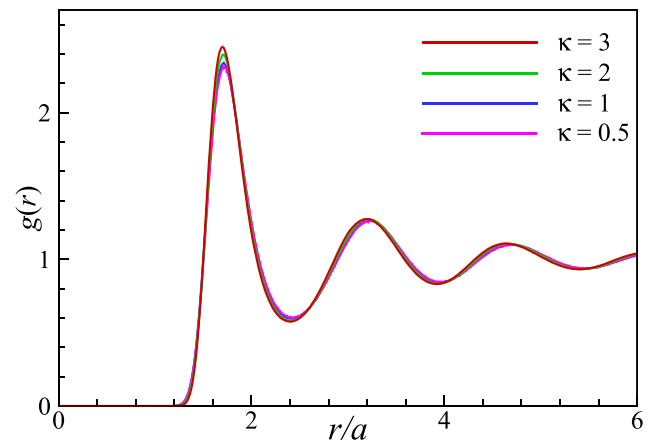


FIG. 2. The radial distribution functions $g(r)$ for four state points of strongly coupled Yukawa fluids, which are listed in Table I. These state points are characterized by approximately the same distance from the melting curve measured in terms of the reduced coupling parameter, $\Gamma/\Gamma_m \simeq 0.8$.

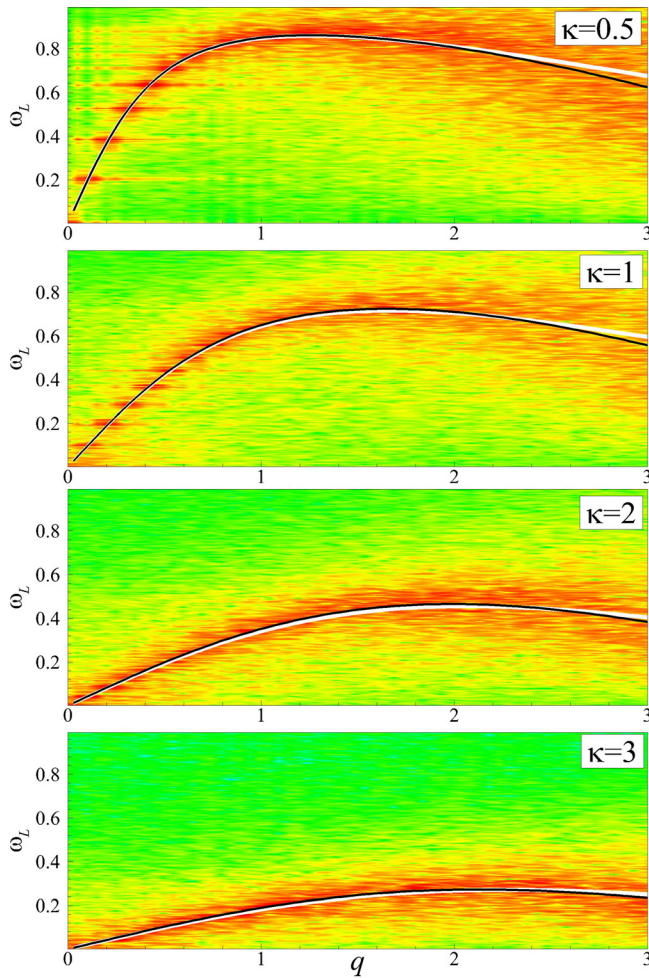


FIG. 3. Dispersion of the longitudinal (plasmon) mode in strongly coupled Yukawa fluids for the (κ, Γ) pairs summarized in Table I. The frequency ω_L is measured in units of the plasma frequency ω_p . The colored background corresponds to the longitudinal current fluctuation spectrum. The dark curves are the results of the full QLCA with $g(r)$ obtained using direct MD simulations. The white curves correspond to the simplified QLCA, Eq. (6), with the energy route to determine R . The vertical scale is chosen the same in the figures to illustrate how the increase in screening (increase in κ) suppresses the wave frequency.

properties of the liquid state, cannot be accounted for within the conventional QLCA, because it does not include damping effects.⁷ Nevertheless, apart from the cutoff, the QLCA shear wave dispersion appears to be nearly parallel to the actual shear wave dispersion curve, and therefore, the QLCA transverse sound velocity remains a meaningful quantity. The longitudinal (c_L) and transverse (c_T) acoustic velocities have been evaluated using the conventional QLCA as well as its simplified version using both the energy (superscript “ u ”) and pressure (superscript “ p ”) routes. The results are summarized in Table I. The overall agreement is very good, and the pressure route is slightly more accurate on average. We have also estimated the thermodynamic longitudinal sound velocity using the conventional fluid approach proposed in Ref. 38. The resulting values are close but slightly lower (several percent deviation) than the QLCA approach yields, as has been already documented.³⁸

In the short-wavelength limit ($q \rightarrow \infty$), both the longitudinal and transverse frequencies approach the common limit,

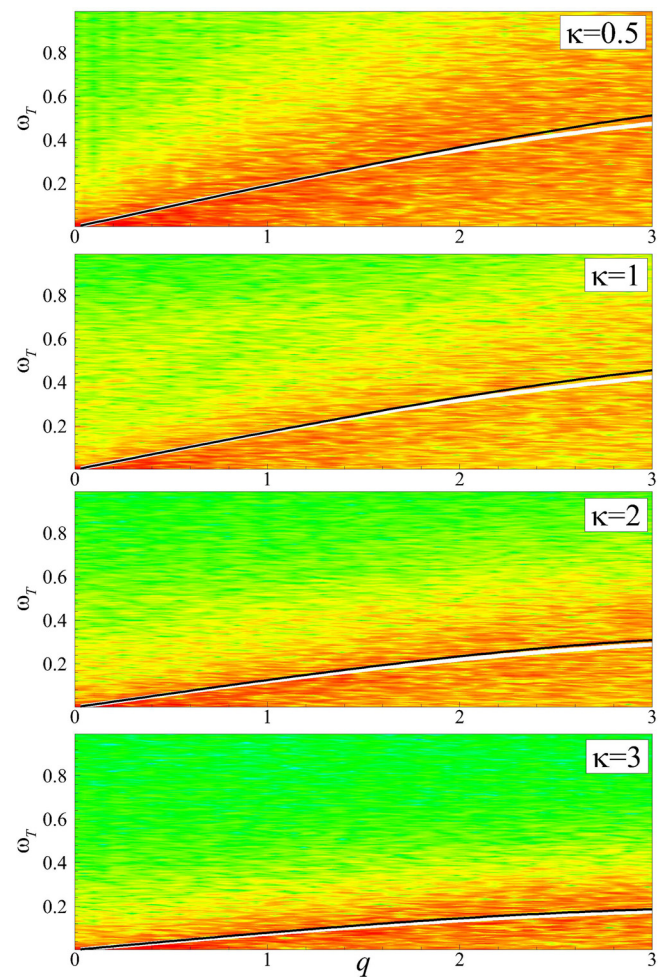


FIG. 4. Same as in Figure 3, but for the transverse (shear) mode.

ω_E , the Einstein frequency, which is the oscillation frequency of a single particle in the fixed environment of other particles (see Fig. 1). For the Yukawa potential, the Einstein frequency is trivially related to the excess internal energy of the system, $\omega_E^2/\omega_p^2 = (2\kappa^2/9\Gamma)u_{ex}$. The same result can be obtained via the energy route of the present simplified QLCA [compare Eqs. (6) and (7) in the $q \rightarrow \infty$ limit with Eq. (8)], indicating that this approach is virtually exact in the short-wavelength limit.

IV. ONE-COMPONENT-PLASMA LIMIT

It is worth to briefly discuss the application of the simplified QLCA to the important limiting case of OCP. This limit corresponds to the unscreened Coulomb interaction between the particles ($\kappa = 0$) and requires the presence of neutralizing background to stabilize the system and ensure finite values for the thermodynamic quantities. The dispersion relations of the simplified QLCA approach can be directly obtained from Eqs. (6) and (7), yielding

$$\omega_L^2 = \omega_p^2 \left(\frac{1}{3} - \frac{2 \cos Rq}{R^2 q^2} + \frac{2 \sin Rq}{R^3 q^3} \right), \quad (10)$$

and

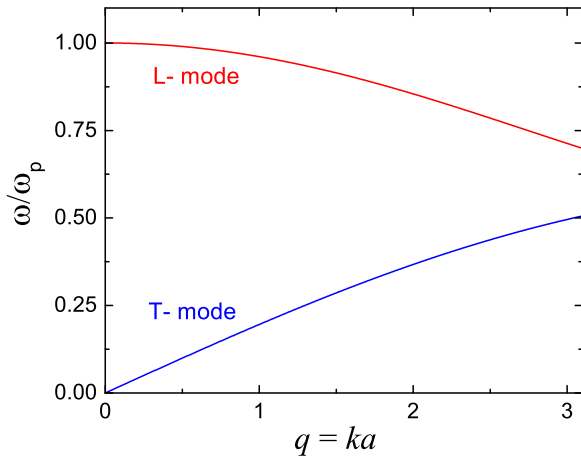


FIG. 5. Longitudinal (plasmon) and transverse (shear) mode dispersion curves of the one-component-plasma model. Eq. (10) for the longitudinal mode and Eq. (11) for the transverse mode are used, with the parameter $R = 1.095$, corresponding to the strongly coupled regime. In this regime, the dispersion curves (with the normalization used) are quasi-independent on the coupling parameter Γ .

$$\omega_T^2 = \omega_p^2 \left(\frac{1}{3} + \frac{\cos Rq}{R^2 q^2} - \frac{\sin Rq}{R^3 q^3} \right). \quad (11)$$

The Kohn sum rule is automatically satisfied, $\omega_L^2 + 2\omega_T^2 = \omega_p^2$. The energy and pressure equations have to be slightly modified due to the presence of the neutralizing background, by substituting $h(r) = g(r) - 1$ instead of $g(r)$. For the Coulomb interaction, the energy and pressure routes give the same result for R in view of the relation $p_{\text{ex}} = \frac{1}{3}u_{\text{ex}}$. The corresponding result is $u_{\text{ex}} = -\frac{3}{4}\Gamma R^2$. At strong coupling, the dominant contribution to the excess internal energy of the OCP model can be approximated, with a good accuracy,^{39,40} as $u_{\text{ex}} \simeq -\frac{9}{10}\Gamma$. Thus, in this strongly coupled regime, the parameter R is practically constant, $R = \sqrt{6/5} \simeq 1.09545$. The corresponding dispersion relations are plotted in Fig. 5, the long-wavelength behavior shown here agrees well with that calculated using the conventional QLCA approach (see, e.g., Fig. 4 from Ref. 2).

In the long-wavelength limit ($q \rightarrow 0$), the longitudinal mode dispersion (10) reduces to

$$\frac{\omega_L^2}{\omega_p^2} \simeq 1 - \frac{2}{30}R^2 q^2 = 1 + \frac{4}{45} \frac{q^2 u_{\text{ex}}}{\Gamma}.$$

For the transverse mode, we get

$$\frac{\omega_T^2}{\omega_p^2} \simeq \frac{1}{30}R^2 q^2 = -\frac{2}{45} \frac{q^2 u_{\text{ex}}}{\Gamma}.$$

These expressions coincide *exactly* with those from the conventional QLCA approach.² In the opposite short-wavelength limit ($q \rightarrow \infty$), the longitudinal and transverse frequencies approach the Einstein frequency, $\omega_E = \omega_p/\sqrt{3}$, which again represents the exact result.

V. CONCLUSION

To summarize, we have proposed a simplified approach to estimate wave dispersion relations in strongly coupled

Yukawa fluids. The approach is based on the QLCA theory and employs the most simple model for the radial distribution function, constructed using excluded volume consideration. Analytic expressions for the longitudinal and transverse wave modes within the simplified QLCA are derived. They demonstrate very good accuracy in the long-wavelength regime and are virtually exact in the short-wavelength limit. The simplified QLCA can be useful when the exact radial distribution functions are not known, but the information about thermodynamics functions (internal energy or pressure) is available. The approach can be easily generalized to Yukawa systems in two dimensions, as well as to other interactions operating in classical systems of strongly coupled particles.

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