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The Roton Minimum: Is it a General Feature of Strongly Correlated Liquids?

G. J. Kalman[1], **S. Kyrkos**[2]*, **K. I. Golden**[3], **P. Hartmann**[1,4], and **Z. Donko**[1,4]

[1] Department of Physics, Boston College, Chestnut Hill, MA 02467, USA
[2] Department of Chemistry & Physics, Le Moyne College, Syracuse, NY 13214, USA
[3] Dept. of Mathematics & Statistics & Dept. of Physics, University of Vermont, Burlington, VT 05405, USA
[4] Research Institute for Solid State Physics & Optics, Hungarian Academy of Sciences, P.O. Box 49, H-1525, Budapest, Hungary

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The roton minimum is a deep minimum in the collective excitation spectrum of the liquid, forming around fairly high k-values. We have discovered, through MD simulations, that this appears to be a general feature of strongly coupled liquids and is ubiquitous in 2D and 3D Yukawa liquids. We suggest that the physical origin of the roton minimum has to be sought in the quasi-localization of particles in a strongly correlated liquid and in the ensuing formation of local microcrystals whose averaged frequency dispersion would show roton minimum-like feature.

The feature that the longitudinal phonon dispersion curve of a dense liquid may exhibit a maxon-roton structure consisting of a sharp maximum followed by a deep minimum was first postulated by Landau [1] and then identified by clear observational evidence for superfluid Helium II [2]. A series of molecular dynamics (MD) simulations [3, 4] have shown (Figure 1) that this phenomenon appears to be a general feature of strongly correlated charged and neutral liquids, in two and three dimensions. (An identification of the underlying basis for this physical phenomenon was anticipated by Fisher [5]). We also have been able to match the predicted roton minimum in a 2D bosonic dipole superfluid [6] with results of classical simulations [7]. Theoretical analysis based on the quasi-localized charge approximation (QLCA) corroborated this picture with the exception that the QLCA generated minimum is not deep enough (Figure 2a) either to match MD simulations (Figure 1) or to satisfy the well-known Feynman upper bound [8]. In Figure 2b, we note the close resemblance between the dispersion curves in the high-temperature classical and low-temperature quantum domains; r_D, the quantum counterpart of the classical coupling parameter Γ_D (defined below), is defined in references [7, 9].

The formation of the observed roton minimum for a strongly correlated liquid is a rather counterintuitive feature of the dispersion curve. Thus, a physical explanation is called for, serving as a basis for an approximation scheme that can rectify the apparent flaw in the QLCA. In this paper, we provide a scenario that relates the existence of the roton minimum in the liquid phase to the features of the phonon dispersion in the crystal lattice. More precisely, the roton minimum can be related to the $\omega = 0$ point at the Brillouin zone boundary. The wave number k-value where this occurs is angle dependent: the lowest k value occurs at 30°.

In order to elucidate this point we study the dynamical behavior of a concrete physical system. We study a model that consists of a collection of spinless electric point dipoles, each of mass m, fixed on a 2D lattice [7, 9]. The repulsive interaction potential is $\phi(r) = \mu^2/r^3$, where μ is the electric dipole strength. The interaction strength is characterized by the coupling parameter $\Gamma_D = \mu^2/a^3 k_B T$ [7], where a is the 2D Wigner-Seitz radius. Note that the 2D dipole system solidifies at $\Gamma_D \approx 70$. Based on the phase diagrams in the classical and quantum domains [10], the closely spaced electron-hole bilayer in its dipole-like excitonic phase may be, in a good approximation, modeled as such a 2D monolayer of interacting point electric dipoles.

In the strongly coupled liquid in the vicinity of the phase boundary, the system may be portrayed as a superposition of randomly oriented microcrystals. In order to obtain the dispersion relation for the liquid based on this

* Corresponding author. E-mail: kyrkoss@lemoyne.edu

model, as outlined in the following pages, we construct the density response function $\chi(k,\omega;\varphi)$ of a microcrystal at a finite temperature, by starting with the lattice response at $T = 0$ and emulating temperature effects through the addition of a phenomenological damping (imaginary frequency ν) term in the response function.

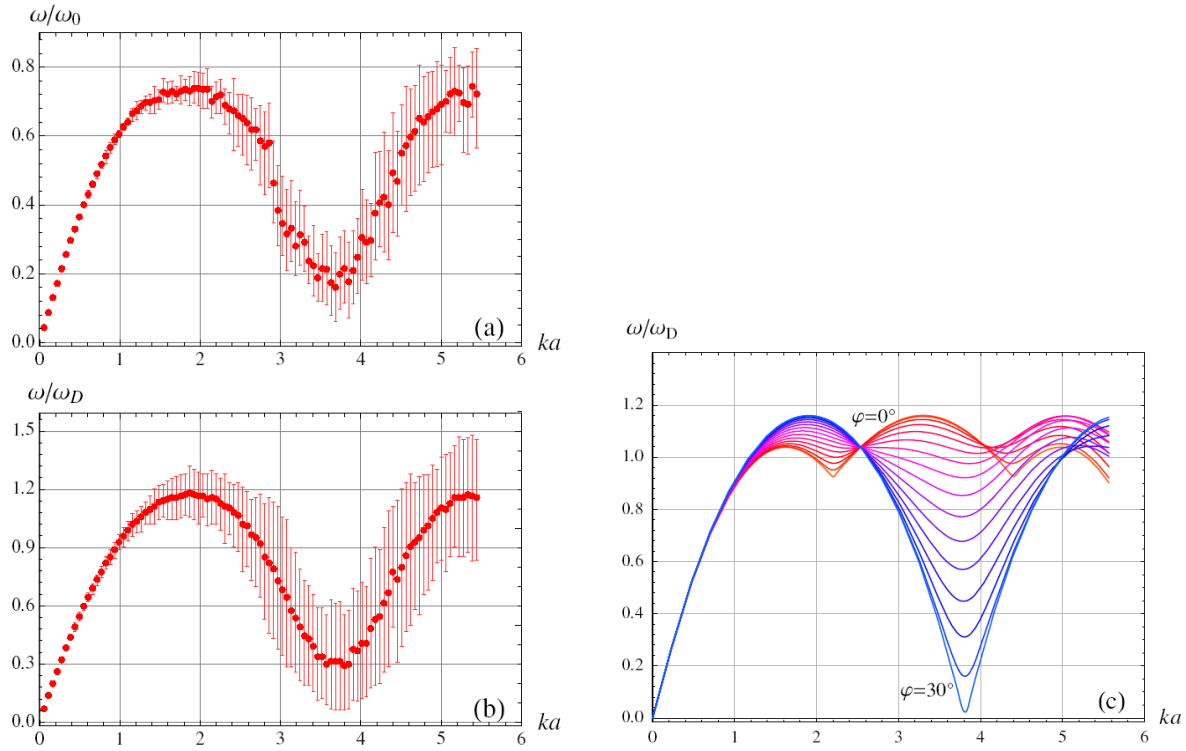

Fig. 1 MD generated longitudinal dispersion functions for (a) a 2D Yukawa system with $\kappa = 1$, $\Gamma = Z^2q^2/ak_BT = 180$ (effective $\Gamma^* = 136$), and $\omega_0^2 = 2\pi q^2 n/ma$, and (b) a 2D dipole system with $\Gamma_D = 60$, $\omega_D^2 = 2\pi n\mu^2/ma^3$. The vertical lines indicate the widths of the spectra. In both cases the minimum is located at the same value of k. (c) The longitudinal phonon dispersion, for $\varphi = 0 - 30^\circ$, for a dipole system. Note the close proximity of the position of the roton minimum in (a) and (b) with the position of $\omega = 0$ for $\varphi = 30^\circ$ at the Brillouin zone boundary.

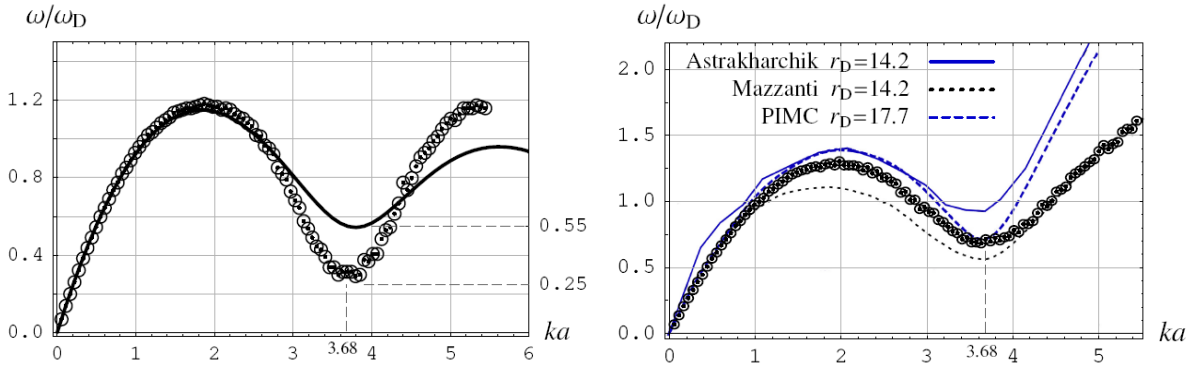

Fig. 2 (a) MD (circles) and QLCA (line) longitudinal dispersion curves for a dipole system, for $\Gamma_D = 60$. (b) Comparison of dispersion curves: MD (classical molecular dynamics) at $\Gamma_D = 15$ (circles); Astrakharchik et al ($T = 0$ Feynman upper bound based on quantum Monte Carlo QMC [6a] simulations) at $r_D = 14.2$; Mazzanti et al [6c] ($T = 0$, presumed lower bound based on QMC simulations at $r_D = 14.2$; and finite-temperature path-integral Monte Carlo (PIMC) simulations [9] at $r_D = 17.7$.

Then, by invoking the classical Fluctuation Dissipation Theorem (FDT), one obtains

$$S(k,\omega;\varphi) = -\frac{1}{\beta n}\,\mathrm{Im}\,\chi(k,\omega;\varphi) \tag{1}$$

The density fluctuation spectrum of the liquid $S(k,\omega)$ can be generated by an angular averaging of the spectra of the microcrystals $S(k,\omega;\varphi)$, with a random orientation of φ. Finally, $S(k,\omega)$ is compared with the same function generated by MD simulations. $\chi(k,\omega;\varphi)$ of the crystal is calculated according to the formula

$$\chi(\mathbf{k},\omega) = \frac{n}{m}\left[\frac{1}{(\omega+i\nu)^2 - \mathbf{C}(\mathbf{k})}\right]_{\mu\nu} k_\mu k_\nu \tag{2}$$

where $C_{\mu\nu}$, the dynamic matrix of the crystal, is

$$\begin{aligned} C_{\mu\nu}(\mathbf{k}) &= -\frac{1}{m}\sum_i [\exp(i\mathbf{k}\cdot\mathbf{r}_i) - 1]\partial_\mu\partial_\nu\varphi_D(r_i) \\ &= \frac{3\mu^2}{m}\sum_i [\exp(i\mathbf{k}\cdot\mathbf{r}_i) - 1]\frac{1}{r_i^5}\left[\delta_{\mu\nu} - 5\frac{r_{i\mu}r_{i\nu}}{r_i^2}\right] \end{aligned} \tag{3}$$

$S(k,\omega;\varphi)$ then follows from Equations (1), (2) and (3):

$$\begin{aligned} S(k,\omega;\varphi) =& \frac{1}{m\beta}2\omega\nu k^2 \\ &\times \frac{[(C_{12}^2 + (C_{22}-\omega^2)^2]\cos^2[\varphi] - C_{12}[C_{11}+C_{22}-2\omega^2]\sin[2\varphi] + [C_{12}^2 + (C_{11}-\omega^2)^2]\sin^2[\varphi])}{[\omega^2 - \Omega_L^2(k,\varphi)]^2[\omega^2 - \Omega_T^2(k,\varphi)]^2 + \omega^2\nu^2\left\{2[\omega^2 - \Omega_L^2(k,\varphi)][\omega^2 - \Omega_T^2(k,\varphi)] + [\Omega_L^2(k,\varphi) - \Omega_T^2(k,\varphi)]^2\right\} + \omega^4\nu^4} \end{aligned} \tag{4}$$

Ω_L^2 and Ω_T^2 are the eigenvalues of the $\mathbf{C}(\mathbf{k})$ matrix. ν values in the range 0.1 to 1.0 were chosen for the calculations. Figure 3 portrays $S(k,\omega;\varphi)$ at a k near the minimum of the dispersion function for a few representative φ values and for $\nu = 0.1$. The $\nu = 0.1$ value has been selected to give the best agreement with our MD simulations. Having obtained $S(k,\omega;\varphi)$ for a microcrystal, we perform an averaging over the φ angles, by summation over a one degree step, in the range 0 to 30 degrees. The results are displayed in Figure 4b. Similar calculations have been carried out for a 2D Yukawa system and the results, portrayed in Figure 4a, are similar to the dipole system and show the universality of the effect. The main peak closely corresponds to the position of the roton minimum in the dispersion curve of Figure 2a. Note the small peak to the right of the main peak of the curve. Its origin presumably is due to the emergence of a longitudinal component in the second ("transverse") mode in the phonon spectrum for a general direction of propagation [11].

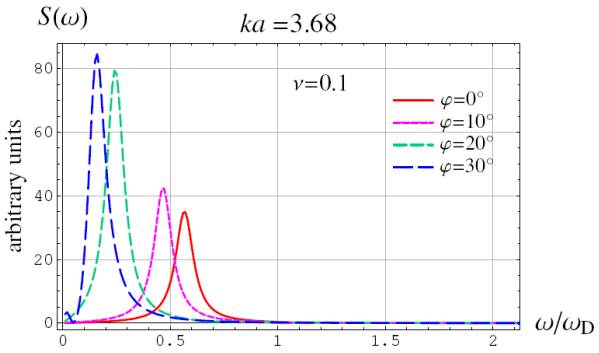

Fig. 3 $S(\omega)$ for a dipole system, at a k value near the minimum of the dispersion function, for some representative angles φ.

A comparison with molecular dynamics simulations of the liquid $S(k,\omega)$ reveals a good qualitative and quantitative agreement (Figure 5). It also shows a substantial lowering of the minimum frequency value compared with the QLCA result. The differences in the tail of the plot are probably due to free (uncaged) particles that are not accounted for in the lattice model. The peaks near $\omega = 0$ are probably due to the approximations introduced by generating the average by a finite sum of discrete values.

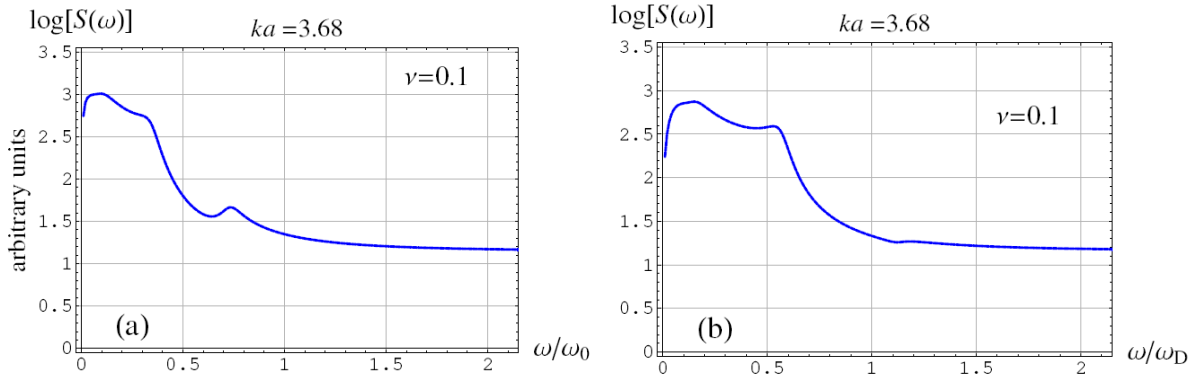

Fig. 4 Angle-averaged $S(\omega)$ at a k value near the minimum of the dispersion function, (a) for a 2D Yukawa system, and (b) for a dipole system. "log" is used to express the base-10 logarithm function.

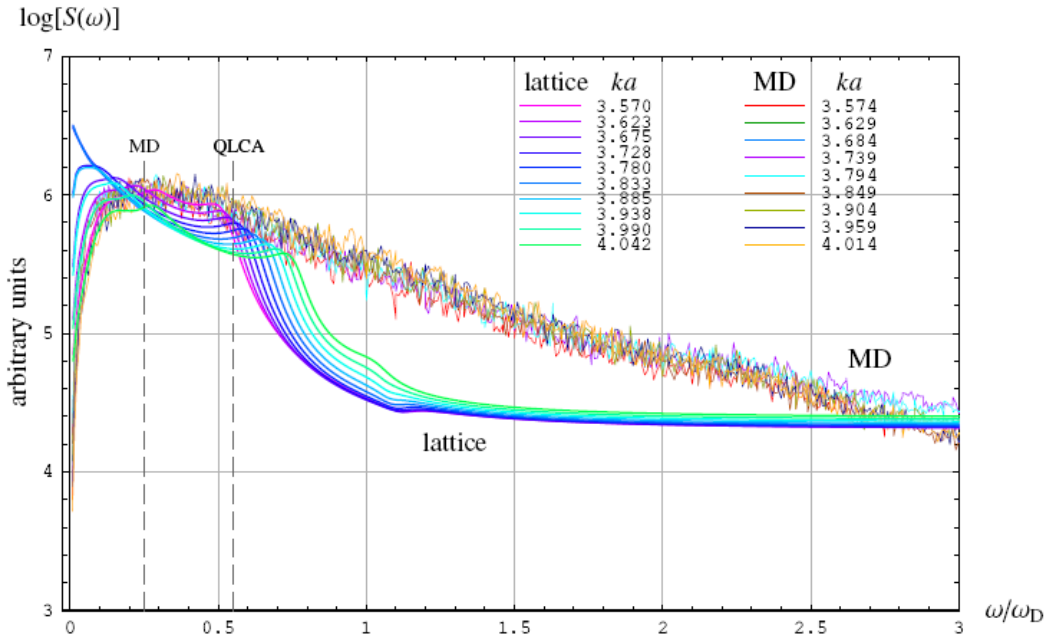

Fig. 5 Comparison of MD generated $S(\omega)$ with results of this work, for some representative k values near the minimum of the dispersion function. The location of the peak predicted by QLCA is also shown; the location of the peaks predicted by MD and by the lattice calculation are close, while the QLCA peak is somewhat off. At $\omega = 1$, k values increase from bottom to top.

In summary, the sharp roton minimum in the dispersion curve in strongly coupled 2D dipole and Yukawa liquids is attributed to the underlying structure of randomly oriented microcrystals. This observation is translated into a phenomenological theory for $S(k, \omega)$ that faithfully emulates the qualitative and quantitative features of its behavior in the vicinity of the roton minimum. In particular, the low value of the roton minimum frequency, missed by the QLCA, is reproduced. The essence of the method is that the collective excitations are calculated from the average of the anisotropic fluctuations, rather than from the fluctuations of the average system, as it is done in the QLCA. We expect that the combination of this method with the QLCA will open the way to account for the local anisotropy of the liquid within the framework of a more sophisticated QLCA model.

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