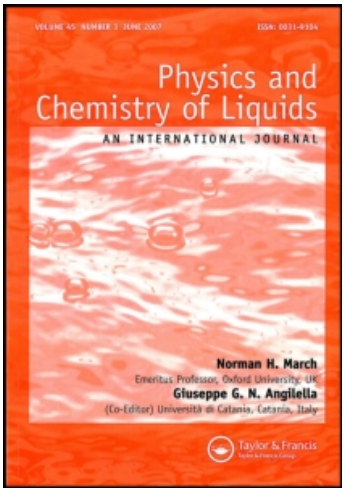

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Melting Criteria for Classical and Quantal Wigner Crystals

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Letter

Melting Criteria for Classical and Quantal Wigner Crystals

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Liquid structure and potential energy considerations lead to an approximate correspondence between the strongly coupled quantal electron plasma near the Wigner transition and its classical counterpart. However no such correspondence exists between the coupling strengths for the two freezing transitions. Electron tunnelling must therefore be important in determining the quantal Wigner transition, a possible quantal picture being proposed by analogy with Zener's idea of classical 'ring' diffusion.

After a great deal of early work on the crystallization of a quantal electron plasma, at a critical coupling strength $a/a_0 = r_{sc}$ say,[1] a measuring the mean interelectronic spacing Ceperley and Alder[2] have established by quantal computer simulation that $r_{sc} \simeq 80$. In this Letter, we shall compare and contrast this strongly coupled quantal assembly near the Wigner transition with its classical counterpart. In the classical one-component plasma (OCP), the coupling strength is conventionally measured by

$$\Gamma = e^2/ak_B T. \tag{1}$$

Following the pioneering work of Brush *et al.*,[3] Hansen[4] and later workers[5] have established that the classical OCP freezes when Γ reaches a value, Γ_c say, of about 180.

As a useful starting point, we have noticed a marked qualitative similarity between the electron pair correlation function $g(r)$ of the strongly coupled electron liquid at $r_s \equiv a/a_0$ equal to 100, as calculated by Lantto and

Siemens[6] and shown in Figure 1a and the pair function of the classical OCP[7] at $\Gamma = 10$, given also in Figure 1b. To press this similarity and thereby to relate the two coupling strengths in Figures 1a and 1b, we consider the Fourier transform of $g(r) - 1$, that is the liquid structure factor $S(k)$. Then, in the long wavelength limit the f-sum rule allows one to write the exact result[8]

$$\lim_{k\to 0} S(k) = \tfrac{1}{2}\hbar\omega_p k^2/4\pi ne^2 \tag{2}$$

for the quantal system, ω_p being the electron plasma frequency and n the electron density $3/4\pi a^3$. The corresponding result for the classical plasma is

$$\lim_{k\to 0} S(k) = k_B T k^2/4\pi ne^2. \tag{3}$$

Thus, this immediately suggests a correspondence between classical and quantal plasmas

$$k_B T \leftrightarrow \tfrac{1}{2}\hbar\omega_p, \tag{4}$$

which is equivalent to

$$\Gamma \leftrightarrow (4r_s/3)^{1/2}. \tag{5}$$

This correspondence, for $r_s = 100$ in the example shown in Figure 1a, yields $\Gamma \simeq 12$, which is satisfactory in comparison with Figure 1b.

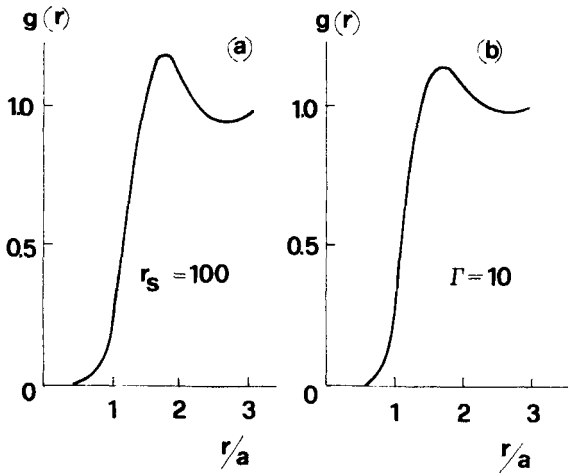

FIGURE 1 The radial pair distribution function $g(r)$ versus r/a in the quantal electron plasma at $r_s = 100$ ((a), from Ref. 6) and in the classical plasma at $\Gamma = 10$ ((b), from Ref. 7).

From $g(r)$, the potential energy per electron is determined by the well known formula

$$U/N = \tfrac{1}{2}ne^2 \int \mathrm{d}\mathbf{r}[g(r) - 1]/r. \tag{6}$$

For classical and quantal systems, using the results of DeWitt *et al.*[5] and of Ceperley and Alder[2] respectively, we find again that the correspondence given in Eq. (5) gives similar values for the potential energy per particle, when measured in units of e^2/a. Thus, although the correspondence in (5) above was derived from a long wavelength argument, it turns out still to apply semiquantitatively to liquid structure and potential energy.

However, when we turn to compare the coupling strengths for melting of classical and quantal Wigner crystals, a wholly different situation obtains. Whereas the critical r_s value for the quantal case, as already mentioned, is about 80, the critical Γ for the classical OCP is about 180. The conclusion from these numbers is that from liquid structure arguments the quantal crystal is more stable than its classical counterpart. The remaining discussion below is therefore focussed on how one can reconcile these apparently quite different melting points, at least in a qualitative manner.

It is helpful to consider first the Lindemann criterion for the melting of the quantal Wigner crystal. Following early work by Pines and Nozières[9] and by Coldwell-Horsfall and Maradudin,[10] extensive phonon calculations on the Wigner lattice were reported by Kugler,[11] and then utilized in the Lindemann criterion. His conclusion was that due to disruption by vibrational energy, the Wigner crystal ought to melt at an r_s value which is an order of magnitude greater than the Ceperley and Alder finding.[2] Though precise numerical values are difficult to obtain, the main point for our present discussion is that the kinetic energy of the localized electrons in a purely vibrational model must be too large, reduction clearly being essential to further stabilize the quantal crystal as required by the Ceperley–Alder results.

What is needed, therefore, is a mechanism whereby electron delocalization can occur, but which still retains the basic structural features of the insulating Wigner crystal. Though, as discussed above, structure determines the potential energy, the virial theorem links potential with kinetic energy. The model we advocate is to retain the insulating character of the Wigner crystal by allowing electron rotational degrees of freedom to play a role, through a mechanism somewhat analogous to that proposed long ago by Zener.[12] His idea was to consider a ring of atoms undergoing classical diffusive motions in unison. Of course, his process was a thermally activated one, and, to our knowledge, the activation energy is sufficiently high in classical crystals so far examined that diffusion proceeds dominantly through

other mechanisms. However, in our case, such rotational motions can occur by quantum mechanical tunnelling at $T = 0$. This mechanism will delocalize electron wave functions from the localized Wigner gaussian orbitals,[1] and, equally important, will retain the insulating $T = 0$ character of the Wigner crystal by the use only of closed loops in the delocalized process. In making the above model quantitative, it is important to consider carefully electron spin, the elementary picture of the Wigner crystal being one of a Néel antiferromagnet, with upward spin electrons on the sites of one simple cubic lattice and downward spins on the other interpenetrating simple cubic lattice.

In summary, we have shown that the correspondence between a strongly coupled quantal electron liquid near the Wigner transition and the classical OCP, embodied in the relation (5) above, is borne out semiquantitatively for (a) liquid structure and (b) potential energy. However, such a correspondence cannot relate melting criteria in the two cases. We have therefore proposed a model in which electrons are delocalized by 'ring diffusion'. This must have the implication that, as the Wigner crystal transition is approached from $r_s > r_{sc}$, the Néel-type antiferromagnetic array of electron spins on a body-centred cubic lattice described above must be relaxed: according to our proposal by rotational interchange of electrons with opposite spins.

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