Is the Quantum Melting of a Polaron Wigner Crystal an Insulator-to-Superconductor Transition?

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On examining the stability of a Wigner Crystal (WC) in an ionic dielectric, two competitive effects due to Polaron formation are found to be important: (i) the screening of the Coulomb forces which destabilizes the crystal, compensated by (ii) the increase of the carrier mass (polaron mass). The quantum melting of the Polaron Wigner Crystal (PWC) is examined. By calculating the quantum fluctuations of both the electrons and the polarization, we show that there is a competition between the dissociation of the Polarons at the insulator-to-metal transition (IMT), and a melting towards a polaron liquid. We find that at strong coupling ($\alpha > \alpha^*$), a liquid state of polarons cannot exist, and the IMT is driven by polaron dissociation. Next, we show that the dipolar interactions between localized polarons are responsible for a phonon instability of the PWC as the density increases. This provides a new mechanism for the IMT in doped dielectrics. Examining the sign of the dielectric constant of the PWC, we conjecture that such an instability could yield an Insulator-to-Superconductor transition.

1. INTRODUCTION.

The possibility of having a Wigner crystallization of large polarons in the cuprates was first proposed in 1989 by Remova and Shapiro \[1\]. This problem was independently studied by us on purely theoretical grounds \[2,3\] in the framework of the Fröhlich model, regardless of the particular structure of the cuprates. Up to now, the many-polaron problem was only studied in the high density (metallic) limit \[4,5\]. Recently, it was found \[6\] that a CDW instability can occur for sufficiently low densities. Our approach relies on the opposite low density limit, which we believe to be relevant for doped ionic dielectrics.

Let us consider a continuous polar medium characterized by three parameters: the static dielectric constant $\varepsilon_s$, the high frequency dielectric constant $\varepsilon_\infty$, and the longitudinal optical phonon frequency $\omega_{LO}$. Reasonable estimates in the cuprates (e.g. in La$_2$CuO$_4$ \[7\]) give $\varepsilon_s \approx 30$, $\varepsilon_\infty \approx 5$ and $\omega_{LO} \approx 20-40\text{meV}$. Importantly, the static dielectric constant is almost isotropic in all directions of space \[8\]. This experimental fact means that the Coulomb forces and the phonons carrying the polarization field act in the same way in all directions. On doping such a polar material, the optical phonons dress and screen the doping charges (this is the well-known polaron effect). The strength of the electron-phonon coupling is given by the dimensionless constant $\alpha = (m^*/2\hbar^2\omega_{LO})^{1/2}e^2/\varepsilon$, where $1/\varepsilon = 1/\varepsilon_\infty - 1/\varepsilon_s$ and $m^*$ is the band mass. Due to their layered structure, the carrier masses in the cuprates are highly anisotropic (in particular, no coherent motion takes place along the $c$-axis). In our model, this situation can be qualitatively described by introducing a second parameter $m_c \gg m^*$, where $m^*$ now refers to the Cu-O planes only. One usually estimates $m^* \approx 1 - 2m_e \ (m_e$ being the bare electron mass), so that the e-ph coupling in the $ab$-planes is roughly $\alpha \approx 4 - 5$.

As is well established, two polarons repel at distance $d$ as $1/\varepsilon_s d$, provided that the dielectric constants satisfy $\varepsilon_\infty/\varepsilon_s > \approx 0.1 \[8\]. Therefore, at
low density and neglecting the details of chemical doping, the crystallized state is necessarily the ground-state of the many-polaron problem. This basic result has several important consequences:

(i) At zero temperature, as the density increases, there is a competition between the quantum melting of the PWC towards a degenerate polaron liquid, and the dissociation (ionization) of the polarons themselves.

(ii) At large values of the e-ph coupling \( \alpha \), the melting towards a polaron liquid is not possible, and it is the polaron dissociation that drives the transition \( \tilde{\varepsilon}_L \).

(iii) Taking into account the dipolar interactions between polarons, it can be shown that the PWC has an intrinsic phonon instability above a given critical density \( n_c \), which is closely related to the polaron dissociation.

(iv) Such an instability has an optical signature: the peak due to the existence of polarons in the optical conductivity, which is centered at a definite frequency at very low density, is progressively shifted towards lower values as the transition is approached, and eventually saturates at \( \omega_{LO} \). This softening has been experimentally observed in Nd\(_{2-x}\)Ce\(_x\)CuO\(_{4-y}\).

(v) The longitudinal dielectric constant \( \varepsilon(k,\omega) \) of the PWC is negative on a large region of \( (k,\omega) \). In particular, \( \varepsilon(k,0)<0 \). As already noted in \( \tilde{\varepsilon}_L \), the repulsive Coulomb interactions between free charges liberated above the critical doping would be overscreened by the high frequency vibrations of the localized PWC.

2. POLARON DISSOCIATION: A NEW QUANTUM CRITICAL POINT.

Let us focus on the quantum melting (at zero temperature) of the PWC. When the e-ph coupling vanishes, we are left with an ordinary WC of electrons. As the density increases, the quantum fluctuations of the localized particles increase and when they reach a certain magnitude, the crystal melts towards a liquid state of electrons. The effect of the host lattice is negligible in that case. In the opposite limit \( \omega_{LO} \to 0 \), the e-ph coupling \( \alpha \) becomes infinite. Each electron in this limit is self-trapped in a polarization potential well which is frozen. As it is well-known, the latter has a coulombic nature and behaves as \( 1/\varepsilon r \) at large distance. Since the polarization cannot move, the polarons, i.e. the electron plus the polarization, cannot go towards a liquid state. The IMT is thus driven by the dissociation of the polarons and the screening of the polarization potentials by the liberated electrons. We are in a situation which closely resembles the usual Mott transition. For finite values of \( \omega_{LO} \), we have applied the Lindemann criterion to the PWC’s melting, as was already done for the usual WC by Nozières et al. \([10]\). The main difference here is that one must take into account the composite nature of the polarons. To be able to delocalize the polarons towards a liquid state, the quantum fluctuations for each polaron moving as a whole, i.e. the motion of each electron together with its surrounding polarization, must become large in comparison with \( R_s \). On the contrary, if the relative fluctuations of the electron with respect to the polarization become large, the polarons break apart (dissociation). The Feynman treatment for the polaron \([11]\), which relies on path integral calculations, yields a natural way to evaluate such quantities: since the polarization field in Feynman’s approach is replaced by a rigid particle with coordinate \( X \) and mass \( M \), one easily calculates both the quantum fluctuations of the center of mass \( R = (m^* x + MX)/(m^* + M) \) and of the relative coordinate \( r = x - X \) (\( x \) is the electron coordinate). This gives two different Lindemann criteria, which correspond to two competing melting scenarios: (i) \( \langle \delta R^2 \rangle^{1/2}/R_s > 1/4 \) for the melting towards a liquid state of polarons; (ii) \( \langle \delta r^2 \rangle^{1/2}/R_s > 1/4 \) for the polaron dissociation.

The frequencies of the two degrees of freedom \( r \) and \( R \), respectively \( \omega_{int} \) and \( \omega_{ext} \), were calculated in ref.\([3]\), together with the ratios (i) and (ii) for different \( \alpha \). From the basic observation that a polaron is a bound state of an electron plus a phonon cloud, it was pointed out that the frequency of vibration of the polaron as a whole is physically limited by the phonon frequency \( \omega_{ext} \leq \omega_{LO} \). In other words, at strong electron-phonon coupling (or equivalently at small \( \omega_{LO} \)), the lattice polarization cannot dynamically follow the increase in
kinetic energy induced by the doping, and the quantum fluctuations are transferred to the internal degree of freedom, breaking the polaron apart. For that reason, the polarons are expected to dissociate at the transition for sufficiently high $\alpha (\alpha > \alpha^* \approx 7.5)$, rather than melting to a polaron liquid. The calculated critical density for $\alpha > \alpha^*$ is $n_c \approx 10^{19} - 10^{20} cm^{-3}$ for $m^* = 1 - 2 m_e$.

Importantly, we have also calculated the same quantities for an anisotropic PWC ($m_c \gg m^* = m_{ab}$), as described in the introduction. In that case the critical coupling for polaron dissociation is $\alpha^* \approx 3$, and the critical density is higher than in the isotropic case: $n_c \approx 10^{20} - 10^{21}$ for $m^* = 1 - 2 m_e$. The rather moderate values of $\alpha^*$ found in the anisotropic case suggest that polaron dissociation should be taken into account in the description of the IMT in the cuprates.

3. THE INSTABILITY AND ITS OPTICAL SIGNATURE.

The spectrum of the low-lying excitations (phonons) of the PWC can also be calculated according to the Feynman model. Basically, one can write down an effective Lagrangian for the PWC \cite{23}, where each polaron is replaced by a two-particle unit $(x_i, X_i)$ localized on a Bravais lattice site $R_i$. The Lagrangian contains two kinds of interactions: (i) local $(x_i, X_i)$ terms (polaron effect, plus the average local field of the remaining electrons); (ii) long-range dipolar interactions, both instantaneous $(x_i, x_j)$ and retarded $(X_i, x_j)$, which are responsible for the dispersion in the vibrational spectrum.

According to this semi-classical treatment, the phonon spectrum of the PWC is given by:

$$\Omega(k, \lambda) = \frac{1}{2} \left\{ \omega_{pol}^2 + \omega_W^2(k, \lambda) / \varepsilon_{\infty} \right\} \pm \sqrt{\left[ \omega_{pol}^2 + \omega_W^2(k, \lambda) \right]^2 - 4 \omega_{LO}^2 \omega_W^2(k, \lambda) / \varepsilon_{\infty}}$$

(1)

where $\omega_W(k, \lambda)$ are the eigenfrequencies of the usual WC \cite{22}, labeled by the index $\lambda$ (two transverse acoustical branches and one longitudinal optical). These satisfy the Kohn sum rule: $\sum_3 \omega_W(k, \lambda) = \omega_p^2$, $\omega_p^2 = 4 \pi n e^2 / m^*$ is the usual electron plasma frequency, $n$ being the doping density). The PWC has thus 6 branches: three with the ‘+’ sign in (1), that we call $\Omega_+$, all optical and which correspond to the vibrations of the ‘internal’ degrees of freedom (the relative motion of the polarization with respect to the electrons); three branches with the ‘-’ sign, that we call $\Omega_-$, corresponding to the vibrations of the polarons as a whole (electrons + polarization oscillating in phase). Two of these branches are transverse acoustical, as in the case of the ordinary WC. The general aspect of the spectrum is sketched on Fig.1. In eq. (1), we have introduced an important quantity:

$$\omega_{pol}^2 = (M/m^* + 1) \omega_{LO}^2 - \omega_p^2 / 3 \varepsilon$$

(2)

which corresponds to the transverse collective modes of the ‘internal’ degree of freedom at $k = 0$. Its evolution as a function of the density is sketched on Fig.2. We have proved elsewhere \cite{3} that such excitations can only be stable provided $\omega_{pol} > \omega_{LO}$. Beyond this limit, i.e. at densities higher than a certain $n_{c1}$, the PWC is unstable with respect to polaron dissociation.

Since $\omega_{pol}$ is the frequency of a transverse mode at $k = 0$, it corresponds to a peak – the polaron peak – in the optical conductivity. As the density increases, $\omega_{pol}$ decreases until $n_{c1}$
is reached, and eventually saturates at $\omega_{pol} \approx \omega_{LO}$. This behaviour has indeed been observed in Nd$_{2-x}$Ce$_x$CuO$_{4-y}$ [1].

4. OVERSCREENING OF THE COULOMB FORCE.

The (longitudinal) dielectric constant $\varepsilon_L(k, \omega)$ can also be calculated. It is given by:

$$\frac{1}{\varepsilon_L(k, \omega)} = \frac{1}{\varepsilon_{ph}(\omega)} \left[ 1 - \frac{\omega_p^2}{\varepsilon_{ph}(\omega)} \times \sum_{\lambda} \frac{(k \cdot e k \lambda)^2}{k^2} \frac{\omega_{LO}^2 - \omega^2}{\omega_{LO}^2 - \omega^2} \right]$$

where $\varepsilon(\omega) = \varepsilon_{\infty}(\omega_{LO}^2 - \omega^2)/(\omega_{TO}^2 - \omega^2)$, is the dielectric constant of the medium, with as usual $\omega_{LO}^2/\omega_{TO}^2 = \varepsilon_s/\varepsilon_{\infty}$. From eq. (3), it is easy to extract the static dielectric constant, which is just proportional to the static dielectric constant of the usual WC calculated by Bagchi [12]:

$$\frac{1}{\varepsilon_L(k, 0)} = \frac{1}{\varepsilon_s} \left[ 1 - \frac{\omega_p^2}{k^2} \sum_{\lambda} \frac{(k \cdot e k \lambda)^2}{\omega_{W}^2(k, \lambda)} \right].$$

Owing to the Kohn sum rule, one easily shows that $\varepsilon(k, 0) < 0$ for any $k > 0$. As it was discussed quite generally by Dolgov et al. [13], this does not necessarily imply an instability: it simply means that the Coulomb repulsion between two test charges in such a system is overscreened by the vibrations of the WC. When one approaches the phonon instability, the negativity of the dielectric constant is extended on a large region of $(k, \omega)$. From this result, we conjecture that beyond the IMT, the moving liberated charges can condense in a superconducting state owing to the overscreening of their Coulomb repulsion by the remaining localized PWC. This can work only up to a second critical density $n_{c2}$ at which the localized polarons themselves are completely screened and destroyed. In some sense, we have some kind of staggered Mott-like transition which begins at $n_{c1}$ and ends at $n_{c2}$. For $n < n_{c1}$ the doping charges form polarons which crystallize. They all disappear for $n > n_{c2}$, while for $n_{c1} < n < n_{c2}$, there could be a separation between localized polarons and condensed charges, the glue being the long-range Coulomb force mediated by the vibrations of the PWC.

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