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Polarization catastrophe in the polaronic Wigner crystal

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Abstract. We consider a three dimensional Wigner crystal of electrons lying in a host ionic dielectric. Owing to their interaction with the lattice polarization, each localized electron forms a polaron. We study the collective excitations of such a polaronic Wigner crystal at zero temperature, taking into account the quantum fluctuations of the polarization within the Feynman harmonic approximation. We show that, contrary to the ordinary electron crystal, the system undergoes a polarization catastrophe when the density is increased. An optical signature of this instability is derived, whose trend agrees with the experiments carried out in Nd-based cuprates.

PACS. 71.30.+h Metal-insulator transitions and other electronic transitions – 71.38.-k Polarons and electron-phonon interactions – 71.45.-d Collective effects

1 Introduction

Let us consider a Wigner crystal (WC) of electrons – a state where the carriers form an ordered array in order to minimize the repulsive Coulomb energy – and put it into a polar dielectric – a material which can respond to the electron motion by displacing the positive and negative ions. If the density of charges added to the dielectric is low, each carrier independently forms a *polaron*, *i.e.* it is surrounded by a polarization cloud due to the interaction with the longitudinal optical phonon modes of the host medium. In the opposite high density limit, dynamical screening will prevent both the localization of charges due to the Coulomb repulsion and the formation of polaronic bound states, thus leading to a weakly interacting liquid. The phase transition occurring upon varying the density between these two extreme limits has been analysed in reference [1] starting from the crystallized state. The resulting picture allows to distinguish between two qualitatively different situations. While in the weak and intermediate electron-phonon coupling regimes, the polaron Wigner crystal (PWC) can melt towards a polaron liquid, this is prevented when the polar coupling is strong: in that case the polarization cloud is virtually frozen and polarons tend to *dissociate* as the system becomes metallic.

The scope of this work is to go beyond the mean-field approach presented in reference [1], with particular attention to the collective modes arising from the long-range interactions between electrons in the crystallized phase. The main result of our study is that in the strong electron-phonon coupling regime, where polarons have a well-defined internal structure, the dipolar electron-electron interactions can lead to very anomalous dielectric properties, and eventually to an instability at high enough density, characterized by a softening of a long wavelength transverse collective mode. An equivalent result was already obtained in previous works assuming a static polarization potential [2], *i.e.* neglecting the quantum fluctuations of the polarization. Similar indications have also been given by other authors [3] starting from a classical liquid state. All these cases are practical realizations of the very general phenomenon of a "dielectric catastrophe" [4], occurring when local dipoles oscillating with a given restoring force (the electrons inside the polarization potential-wells) interact through the long-range Coulomb forces, which tend to soften the transverse restoring potential. Such instability, which can be traced back to the local Lorentz field, has an experimental signature: the frequency of the polaron peak in the optical conductivity decreases with doping, a trend that has indeed been observed in underdoped $Nd_{2-x}Ce_xCuO_{4-y}$ (NCCO) below the insulator-to-metal transition [5].

The paper is organized as follows. The main features of the model, which was thoroughly introduced in reference [1], are summarized in Section 2. In Section 3, we calculate the phonon dispersion of the PWC on the basis of Feynman's harmonic trial model for the polarons. In Section 4, we evaluate the effect of the vibrations of the PWC by calculating the dielectric constant $\varepsilon(\mathbf{k},\omega)$ as a function of the density. We show that it is *negative* on a large region of $\mathbf{k}$ and ω when the system approaches the instability. In conclusion, we propose a scenario where free carriers liberated by the dissociation of polarons could coexist with the residual localized polarons in a "mixed" phase beyond the instability, and speculate about a novel

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pairing mechanism where the collective vibrations of the PWC or, equivalently, the "overscreening" related to the negativity of the dielectric constant $\varepsilon(k,\omega)$, acts as a glue for the Cooper pairs.

2 Model and approximations

The model for the crystallized state has been thoroughly defined in reference [1], equations (1–5). Electrons with band mass m^* at a density $n = (4\pi/3)^{-1} R_s^{-3}$ interact directly through the long-range Coulomb forces. These are immersed in a polar background characterized by the longitudinal phonon frequency ω_{LO} and the effective dielectric constant $\tilde{\varepsilon}$. The polar medium responds to the electron motion through a Fröhlich type interaction characterized by a coupling constant $\alpha = \left(m^*/2\hbar^3\omega_{LO}\right)^{1/2} e^2/\tilde{\varepsilon}$, which can lead to polaron formation. Once the degrees of freedom of the polarization field are integrated out, the resulting action for the electrons can be expanded assuming small displacements $\mathbf{u}_i$ relative to the equilibrium positions. To quadratic order, this reads:

$$S\left(\{\mathbf{u}_i\}\right) = \sum_i S_i + \frac{1}{2}\sum_{i\neq j} S_{ij} \tag{1}$$

with

$$S_i = -\beta\frac{9e^2}{10\varepsilon_s R_s} - \int_0^\beta \left[\frac{m^*}{2}\dot{u}_i^2(\tau) + \frac{m^*}{2}\frac{\omega_W^2}{\varepsilon_s}u_i^2(\tau)\right]\mathrm{d}\tau + \frac{\omega_{LO}e^2}{4\tilde{\varepsilon}}\int_0^\beta\int_0^\beta \frac{G_{\omega_{LO}}(\beta,\tau-\sigma)}{|\mathbf{u}_i(\tau)-\mathbf{u}_i(\sigma)|}\mathrm{d}\tau\mathrm{d}\sigma \tag{2}$$

and

$$S_{ij} = -\frac{e^2}{\varepsilon_\infty}\sum_{\alpha\gamma}\int_0^\beta u_i^\alpha(\tau)\Lambda_{ij}^{\alpha\gamma}u_j^\gamma(\tau)\mathrm{d}\tau + \frac{\omega_{LO}e^2}{2\tilde{\varepsilon}}\sum_{\alpha\gamma}\int_0^\beta\int_0^\beta u_i^\alpha(\tau)\Lambda_{ij}^{\alpha\gamma}u_j^\gamma(\sigma)G_{\omega_{LO}}(\beta,\tau-\sigma)\mathrm{d}\tau\mathrm{d}\sigma \tag{3}$$

where the indices $\alpha,\gamma = (x,y,z)$ denote the Cartesian coordinates. The dipolar matrix elements are defined as:

$$\Lambda_{ij}^{\alpha\gamma} = \frac{\delta_{\alpha\gamma}R_{ij}^2 - 3R_{ij}^\alpha R_{ij}^\gamma}{R_{ij}^5}\,. \tag{4}$$

We have introduced the phonon propagator $G_\omega(\beta,\tau-\sigma) = (\bar{n}+1)\mathrm{e}^{-\omega|\tau-\sigma|} + \bar{n}\mathrm{e}^{\omega|\tau-\sigma|}$, together with $\bar{n} = (\mathrm{e}^{\beta\omega}-1)^{-1}$. The frequency ω_W is related to the plasma frequency by the relation $\omega_W^2 = \omega_p^2/3 = e^2/m^*R_s^3$. β is the inverse temperature, and the units are such that $\hbar = k_B = 1$.

Expressions (1–3) constitute the basic model for the polaron Wigner crystal (PWC). It is based essentially on the following approximations: (i) anharmonic terms in the expansion in $\{\mathbf{u}_i\}$ are not considered; (ii) exchange effects between electrons are neglected, and so is the possible (weak) magnetic ordering of the crystal. For the main purposes of our studies, *i.e.* the crystal stability, all these approximations are well justified in the range of densities that we consider, as was discussed in detail in reference [1].

The local part of the electron action has been extensively studied in reference [1], by isolating neutral entities containing only one electron – the so-called Wigner spheres – which are non-interacting at mean-field level. In this approach, all the correlation effects are carried by a local harmonic potential (the second term in square brackets in Eq. (2)), which merely adds to the polaron problem (the additional constant term is the electrostatic energy coming from the jellium approximation). The last term in equation (2) is the self-induced interaction mediated by the polarization, which is responsible for polaron formation. It is not solvable, but it can be treated successfully within the Feynman variational approach [6] by introducing a quadratic trial action S_0 of the form

$$S_0 = -\frac{m^*}{2}\int \dot{u}^2\mathrm{d}\tau - \frac{m^*}{2}\frac{\omega_W^2}{\varepsilon_s}\int u^2\mathrm{d}\tau - \frac{Kw}{8}\int\int |\mathbf{u}(t)-\mathbf{u}(s)|^2 G_w(\beta,\tau-\sigma)\mathrm{d}\tau\,\mathrm{d}\sigma, \tag{5}$$

(we have dropped the site index, since at this stage all the sites are equivalent). It can be seen that expression (5) results from the path-integration over $\mathbf{X}$ of the following Lagrangian:

$$L_0 = \frac{m^*}{2}\dot{u}^2 + \frac{M}{2}\dot{X}^2 - \frac{K}{2}\left(\mathbf{u}-\mathbf{X}\right)^2 - \frac{m^*}{2}\frac{\omega_W^2}{\varepsilon_s}u^2 \tag{6}$$

where $w^2 = K/M$. In equation (6), K and w are variational parameters to be adjusted in order to minimize the free energy. This method of approximation, not only yields the best known analytical results for the polaron energy, but it also gives a very good insight into the physics of the polaron problem: when the variational parameters K and w are properly adjusted, the model (6) is a good schematization of the true polaron, especially at low temperatures (roughly at $T < \omega_{LO}$, where the polarization cloud is sufficiently "rigid"). Basically, the actual polarization of the polaron is replaced by an auxiliary particle with coordinate $\mathbf{X}$ and mass M, which reduces the many-body polaron problem to a two-body problem.

Let us now consider the non-local part (3) for $i \neq j$. These terms, which represent dipole-dipole interactions between different electrons, become important beyond the mean-field approximation described above, and they are responsible for the dipersion of the vibrating collective modes. The first term in equation (3) is instantaneous, and it comes out from the direct Coulomb electron-electron repulsion. The second term is a retarded dipole-dipole interaction which, as the polaron term in (2), is mediated by the polarization. Its physical meaning is that an electron i, when it moves, feels the polarization field created by electron j. It is important to emphasize that, unlike the polaron term in (2), this retarded dipole-dipole interaction

vanishes with ω_{LO} (*i.e.* $\alpha \to \infty$): when the polarization is completely frozen, only the instantaneous dipole-dipole interaction remains at work. This particular limit, which is very instructive, has been already studied in reference [2]. Here, we consider the general case $\omega_{LO} \neq 0$, where the retarded dipole-dipole interactions cannot be neglected.

2.1 Effective quadratic model

In the same spirit as the Feynman approach for the single polaron, we introduce an effective Lagrangian:

$$L_{\mathit{eff}} = \sum_i \frac{m^*}{2}\dot{u_i}^2 + \frac{M}{2}\dot{X_i}^2 - \frac{K}{2}\left(\mathbf{u}_i - \mathbf{X}_i\right)^2 - \frac{m^*}{2}\frac{\omega_W^2}{\varepsilon_s}u_i^2 - \frac{1}{2}\sum_{i\neq j}\mathbf{u}_i\hat{A}_{ij}\mathbf{u}_j - \sum_{i\neq j}\mathbf{u}_i\hat{B}_{ij}\mathbf{X}_j \quad (7)$$

where the $\hat{A}_{ij}$ and $\hat{B}_{ij}$ are 3×3 matrices in coordinate space. This model can be thought of as a generalization of the Lorentz lattice of dipoles, which includes retarded dipolar interactions through the auxiliary particles $\mathbf{X}_i$.

The effective action for the electrons is obtained after path-integration on the coordinates $\mathbf{X}_i$:

$$\begin{aligned} S_{\mathit{eff}} = &-\frac{m^*}{2}\sum_i \int \mathrm{d}\tau[\dot{u}_i^2(\tau) - (K/m^* + \omega_W^2/\varepsilon_s)u_i^2(\tau)] \\ &-\frac{1}{2}\sum_{i\neq j}\int \mathbf{u}_i(\tau)\hat{A}_{ij}\mathbf{u}_j(\tau)\mathrm{d}\tau \\ &+\frac{1}{4Mw}\iint \mathrm{d}\tau\mathrm{d}\sigma G_w(\beta,\tau-\sigma)\times \\ &\times\left\{\sum_i \mathbf{u}_i(\tau)\left[K^2 - 2K\hat{B}_{ii} + \sum_l \hat{B}_{li}\hat{B}_{li}\right]\mathbf{u}_i(\sigma)\right. \\ &\left.\sum_{i\neq j}\mathbf{u}_i(\tau)\left[-2K\hat{B}_{ij} + \sum_l \hat{B}_{li}\hat{B}_{lj}\right]\mathbf{u}_j(\sigma)\right\}. \end{aligned} \quad (8)$$

At this stage, the most rigorous way to proceed would be to self-consistently solve the full Feynman variational problem given by:

$$E = \min_{K,w}\left[E_0 + \lim_{\beta\to\infty}\langle S - S_{\mathit{eff}}\rangle\right], \quad (9)$$

where E_0 is the zero-point energy of all the eigenmodes of (7) [7]. However, we can get more insight into the problem by making use of two additional approximations:

(i) We restrict to a single variational parameter, by putting $w = \omega_{LO}$ in the expression (8). As was pointed out in reference [1], this has very little effect on the results when dealing with moderate to large e-ph couplings ($\alpha >\sim 5$). From now on, we will mainly focus on this case, which is to our opinion the most interesting one.

(ii) For the variational parameter K, we use the results obtained in the framework of the mean-field approximation (see Ref. [1]). The underlying idea is that of a perturbation expansion around the mean-field result, so that the first correction is calculated by taking the zeroth order as a good starting point. At low enough density, the dipolar terms are indeed a small perturbation, since they explicitely involve the fluctuations $\mathbf{u}_i$ of the electrons around their equilibrium positions, which are small in this limit. More quantitatively, in the case of the ordinary WC, it was shown that the inclusion of dipolar interactions changes the total energy by no more than 5–10 percent [8].

Of course, the previous argument no longer applies when approaching the critical density, where one can expect some quantitative deviations from the predicted phonon spectrum. However, our results should be at least qualitatively correct in all the insulating phase.

Within this framework, there is no need to perform new path-integrations: the matrices $\hat{A}_{ij}$ and $\hat{B}_{ij}$ are chosen in order that all the dipole-dipole terms of $S - S_{\mathit{eff}}$ in equation (9), vanish term by term. This leads to the following set of equations:

$$\hat{A}_{ij} = \frac{e^2}{\varepsilon_\infty}\hat{\Lambda}_{ij} \quad ; \quad i\neq j \quad (10)$$

$$-2K\hat{B}_{ii} + \sum_l \hat{B}_{li}\hat{B}_{li} = 0 \quad (11)$$

$$-2K\hat{B}_{ij} + \sum_l \hat{B}_{li}\hat{B}_{lj} = K\frac{e^2}{\tilde{\varepsilon}}\hat{\Lambda}_{ij} \quad ; \quad i\neq j. \quad (12)$$

The first equation straightforwardly defines the matrix $\hat{A}_{ij}$ of the instantaneous dipole-dipole interactions, and the set of equations for the retarded terms $\hat{B}_{ij}$ will be solved in the next section. As a result, we are able to calculate the phonon spectrum of the effective Lagrangian (7), which in the same manner as (6) for the single polaron problem, mimics the collective excitations of the PWC.

3 Phonon spectrum and instability

In this section, we derive the phonon spectrum of the Lagrangian (7), as was done for the usual Wigner crystal and Lorentz lattice of dipoles in references [8–10].

First, the WC of electrons is recovered from equation (7) by putting the e-ph coupling to zero, with $\varepsilon_s \to \varepsilon_\infty$. All the terms involving $\{\mathbf{X}_i\}$ thus vanish, and we obtain [10]:

$$L_W = \sum_i \frac{m^*}{2}\dot{u_i}^2 - \frac{m^*}{2}\frac{\omega_W^2}{\varepsilon_\infty}u_i^2 - \frac{1}{2}\sum_{i\neq j,\mu\nu}\mathbf{u}_i\hat{A}_{ij}\mathbf{u}_j \quad (13)$$

with the matrices $\hat{A}_{ij}$ for $i\neq j$ given by equation (10). The potential term of (13) can be included in the definition of the $\hat{A}$'s by putting $\hat{A}_{ii} = (m^*\omega_W^2/\varepsilon_\infty)\hat{I}$, with $\hat{I}$ the identity matrix. We can now diagonalize the coupled equations of electron motion

$$m^*\ddot{\mathbf{u}}_i(t) - \sum_j \hat{A}_{ij}\mathbf{u}_j(t) = 0 \quad (14)$$

by introducing eigenmodes of the form:

$$\mathbf{u}_i(t) = u_{\mathbf{k}\lambda}\epsilon_{\mathbf{k}\lambda}\mathrm{e}^{\mathrm{i}\left(\mathbf{k}\cdot\mathbf{R}_{ij} - \frac{\omega(\mathbf{k},\lambda)}{\sqrt{\varepsilon_\infty}}t\right)} \tag{15}$$

where $\epsilon_{\mathbf{k}\lambda}$ is the polarization vector of the mode $(\mathbf{k},\lambda)$, $\mathbf{k}$ being a wavevector in the Brillouin zone, and λ the index of the (three) phonon branches. If we define the Fourier transform as

$$\hat{A}_{\mathbf{k}} = \frac{1}{N}\sum_j \hat{A}_{ij}\mathrm{e}^{\mathrm{i}\mathbf{k}\cdot\mathbf{R}_{ij}} \tag{16}$$

then the eigenfrequencies fulfill:

$$\epsilon_{\mathbf{k}\lambda}N\hat{A}_{\mathbf{k}}\epsilon_{\mathbf{k}\lambda'} = m^*\frac{\omega^2(\mathbf{k},\lambda)}{\varepsilon_\infty}\delta_{\lambda\lambda'}\cdot \tag{17}$$

For a given Bravais lattice, the matrix elements of $\hat{A}_{\mathbf{k}}$ are given by [9]:

$$A^{\mu\nu}_{\mathbf{k}} = k^\mu k^\nu V_{\mathbf{k}} + \sum_{\mathbf{K}\neq 0} V_{\mathbf{k}+\mathbf{K}}(k^\nu + K^\nu)(k^\mu + K^\mu) - \sum_{\mathbf{K}\neq 0} K^\mu K^\nu V_{\mathbf{K}} \tag{18}$$

where $V_k = 4\pi e^2/k^2\varepsilon_\infty$ is the Fourier transform of the Coulomb potential, with $V_{k=0}$ set to zero for charge neutrality, and $\mathbf{K}$ a reciprocal lattice vector. From (17) and (18) it is easy to prove the following result:

$$\sum_\lambda \frac{\omega(\mathbf{k},\lambda)^2}{\varepsilon_\infty} = 3\omega_W^2/\varepsilon_\infty = \omega_p^2/\varepsilon_\infty \tag{19}$$

which is known as the Kohn sum rule [9,10]. The phonon spectrum of the WC contains two acoustical transverse modes (t) and one optical longitudinal mode (ℓ), which satisfies $\omega(0,\ell) = \omega_p/\sqrt{\varepsilon_\infty}$.

Let us now calculate the Fourier transform $\hat{B}_{\mathbf{k}}$ of the matrices $\hat{B}_{ij}$, defined in the same way as $\hat{A}_{\mathbf{k}}$ in equation (16). Multiplying equation (12) by $\exp(\mathrm{i}\mathbf{k}\cdot\mathbf{R}_{ij})$, summing over j, then making use of (11), one gets:

$$-2KN\hat{B}_{\mathbf{k}} + N\hat{B}_{\mathbf{k}}N\hat{B}_{\mathbf{k}} = K\frac{e^2}{\tilde{\varepsilon}}N\hat{\Lambda}_{\mathbf{k}} - \frac{m^*\omega_W^2}{\tilde{\varepsilon}}\hat{I}\cdot \tag{20}$$

The above equation can be solved in the same basis $\{\epsilon_{\mathbf{k},\lambda}\}$ which diagonalizes $\hat{\Lambda}_{\mathbf{k}}$ and $\hat{A}_{\mathbf{k}}$. Therefore, one obtains $\epsilon_{\mathbf{k},\lambda}N\hat{B}_{\mathbf{k}}\epsilon_{\mathbf{k},\lambda'} = g(\mathbf{k},\lambda)\delta_{\lambda,\lambda'}$ where:

$$g(\mathbf{k},\lambda) = K\left[1 - \sqrt{1 + \frac{1}{\tilde{\varepsilon}}\frac{m^*}{M}\frac{\omega^2(\mathbf{k},\lambda) - \omega_W^2}{\omega_{LO}^2}}\right]\cdot \tag{21}$$

3.1 Phonon spectrum

We can now determine the whole phonon spectrum of the PWC by diagonalizing the Lagrangian (7). Let us introduce the Fourier transforms

$$\mathbf{u}_i = \frac{1}{\sqrt{N}}\sum_{\mathbf{k}\lambda} u_{\mathbf{k}\lambda}\epsilon_{\mathbf{k}\lambda}\mathrm{e}^{-\mathrm{i}\mathbf{k}\cdot\mathbf{R}_i}$$
$$\mathbf{X}_i = \frac{1}{\sqrt{N}}\sum_{\mathbf{k}\lambda} X_{\mathbf{k}\lambda}\epsilon_{\mathbf{k}\lambda}\mathrm{e}^{-\mathrm{i}\mathbf{k}\cdot\mathbf{R}_i} \tag{22}$$

then the equations of motion become

$$\ddot{u}_{\mathbf{k}\lambda} = \left[\frac{\omega_W^2}{\tilde{\varepsilon}} - \frac{\omega^2(\mathbf{k}\lambda)}{\varepsilon_\infty} - \frac{K}{m^*}\right]u_{\mathbf{k}\lambda} + \frac{[K - g(\mathbf{k},\lambda)]}{m^*}X_{\mathbf{k}\lambda}$$
$$\ddot{X}_{\mathbf{k}\lambda} = \frac{[K - g(\mathbf{k}\lambda)]}{M}u_{\mathbf{k}\lambda} - \frac{K}{M}X_{\mathbf{k}\lambda}. \tag{23}$$

For each wavevector $\mathbf{k}$ and polarization λ, the solution of the secular equation gives the eigenfrequencies $\Omega(\mathbf{k},\lambda)$ of the PWC in terms of the eigenfrequencies $\omega(\mathbf{k},\lambda)$ (formula (18)) of the WC of electrons:

$$\Omega^2(\mathbf{k},\lambda) = \frac{1}{2}\Bigg\{\omega_{pol}^2 + \frac{\omega^2(\mathbf{k},\lambda)}{\varepsilon_\infty} \pm\sqrt{\left[\omega_{pol}^2 + \frac{\omega^2(\mathbf{k},\lambda)}{\varepsilon_\infty}\right]^2 - 4\omega_{LO}^2\frac{\omega^2(\mathbf{k},\lambda)}{\varepsilon_s}}\Bigg\} \tag{24}$$

where we have defined

$$\omega_{pol}^2 = \left(\frac{M}{m^*} + 1\right)\omega_{LO}^2 - \frac{\omega_W^2}{\tilde{\varepsilon}}\cdot \tag{25}$$

In the following, we shall denote by $\Omega_{int}(\mathbf{k},\lambda)$ $[\Omega_{ext}(\mathbf{k},\lambda)]$ the solution with the $+$ $[-]$ sign. By looking at the equations of motion (23), one can verify that in the limit of low k, these frequencies are associated respectively with the out-of-phase and in-phase vibrations of the electrons relative to the polarization. Interestingly enough, within our approximation scheme, the polaron effect now enters in the frequency spectrum through the parameter ω_{pol} alone. Moreover, the parameter ω_{pol} is identified as the frequency of the transverse mode $\Omega_{int}(\mathbf{k},t)$ at $k = 0$.

It is instructive to work out the solutions of equation (24) in the low density limit, where ω_W is small compared to the internal frequency of an isolated polaron. In this case we obtain

$$\Omega_{ext}^2(\mathbf{k},\lambda) \simeq \frac{\omega^2(\mathbf{k},\lambda)/\varepsilon_s}{M_P/m^*} \tag{26}$$

$$\Omega_{int}^2(\mathbf{k},\lambda) \simeq \omega_{pol}^2 + \frac{\omega^2(\mathbf{k},\lambda)}{\varepsilon_\infty}. \tag{27}$$

Those frequencies are sketched in Figure 1. At low density, there is a clear separation of energy scales between:

i) the low frequency "external" modes with $\Omega_{ext} \ll \omega_{LO}$, which correspond to a Wigner crystal of particles with mass M_P instead of m^*. At such low frequencies, the polarons behave as rigid particles and the internal electronic motion can be neglected. Furthermore, all the polarization cloud participates in the screening of the dipolar interactions, so that the eigenfrequencies are reduced by a factor ε_s.

ii) the high frequency "internal" modes with $\Omega_{int} \gg \omega_{LO}$, which correspond to a Lorentz lattice of dipoles with characteristic frequency ω_{pol}, which was extensively studied in reference [2]. In this case, the electron vibrations are very fast and the appropriate dielectric constant is ε_∞.

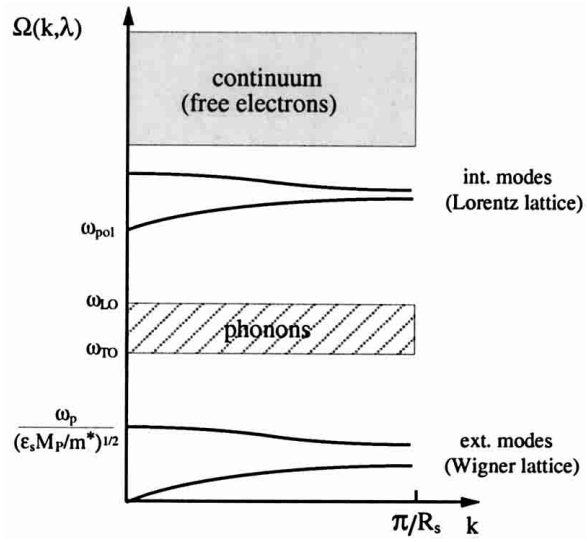

Fig. 1. The excitation spectrum of the PWC.

Such separation of energy scales is strictly valid only at very low densities. At densities such that ω_{pol} approaches ω_{LO}, *i.e.* close to the phonon instability (see below), the full expression (24) must be used to determine the eigenfrequencies.

The treatment of this section provides us with an estimate of the low-lying excitations of the PWC. At this stage, it is useful to comment on a few points concerning our approximations. First of all, above some threshold frequency higher than ω_{pol}, the calculated eigenmodes will eventually merge into a continuum of states, corresponding to free electrons ionized from both the polarization potential and the periodic potential of the Wigner crystal itself. In addition, more bound states can appear below such threshold, if there are metastable interstitial states with long enough lifetimes. As far as the Feynman approximation is concerned, we know from the single polaron results [12] that the interpretation of the eigenfrequency of the quadratic model as representative of the excitation spectrum of the real system is qualitatively correct in the strong coupling limit, where the optical Franck-Condon transition is very sharp (indeed, the polarizability of a system with a definite quantum transition has exactly the same form as for a classical harmonic oscillator). At intermediate values of α, however, the transitions inside the polarization potential-well are considerably broadened. In this case, the calculated eigenfrequencies should still be of significance, provided that we associate to them a finite linewidth Γ, related to the so-called Huang-Rhys factor.

3.2 Instability of the polaron system

Contrary to the ordinary WC of electrons, the PWC can undergo a *phonon instability* when the density reaches a critical value. This can be most clearly seen by writing the action (8) as a sum over the quadratic actions of the independent eigenmodes $u_{\mathbf{k}\lambda}$, $S_{\mathit{eff}} = \sum_{\mathbf{k}\lambda} S_{\mathbf{k}\lambda}$, with

$$S_{\mathbf{k}\lambda} = -\frac{m^*}{2}\int |\dot{u}_{\mathbf{k}\lambda}(\tau)|^2 \mathrm{d}\tau - \frac{m^*\omega^2(\mathbf{k},\lambda)}{\varepsilon_s}\int |u_{\mathbf{k}\lambda}(\tau)|^2 \mathrm{d}\tau - \frac{K(\mathbf{k},\lambda)\omega_{LO}}{8}\int\int |u_{\mathbf{k}\lambda}(\tau) - u_{\mathbf{k}\lambda}(\sigma)|^2 G_{\beta,\omega_{LO}}\,\mathrm{d}\tau\,\mathrm{d}\sigma. \tag{28}$$

We see that each of the $S_{\mathbf{k}\lambda}$ has the same form as the mean-field action (5), but the coefficient of the polaron term (the retarded interaction of $u_{\mathbf{k}\lambda}$ with itself) is now

$$K(\mathbf{k},\lambda) = K\left(1 + \frac{1}{\tilde{\varepsilon}}\frac{m^*}{M}\frac{\omega^2(\mathbf{k},\lambda) - \omega_W^2}{\omega_{LO}^2}\right).$$

Both $\omega^2(\mathbf{k},\lambda)$ and ω_W^2 are proportional to the density, so that for $n \to 0$ we recover $K(\mathbf{k},\lambda) = K$. However, upon increasing the density, the restoring force $K(\mathbf{k},\lambda)$ can become negative, and the system is unstable. The first instability occurs due to the long wavelength transverse modes for which $\omega(k=0,t) = 0$. It is thus *the long wavelength transverse vibrations of the electrons relative to their polarization cloud which cause the instability*, a phenomenon which is reminiscent of the phenomenological dissociation criterion introduced in reference [1].

In terms of the parameter ω_{pol}, we see that $K(k=0,t)$ vanishes (*i.e.* the system is on the verge of the instability) when

$$\omega_{pol} = \omega_{LO}. \tag{29}$$

This instability condition is the correct generalization of the condition $\omega_{pol} = 0$ of reference [2] to the case of a finite phonon frequency. Let us stress once again that the phonon instability occurs at $k = 0$, where the properties of the system are independent of the details of the lattice ordering, provided that the system remains isotropic. As a consequence, the instability criterion (29) also applies to disordered insulating polaron phases, which can be realized in real solids where the doping ions act as impurity potentials, thus driving the particles away from their configurations on a Bravais lattice to form a Wigner glass. The above criterion should also hold in the case of a polaron liquid, provided that the motion of the polarons is slow compared to the vibrations of the electrons inside their potential wells [11].

In Table 1 we have reported the instability parameter $r_s^{(inst)}$ as deduced from criterion (29), compared to the values for polaron dissociation and crystal melting obtained in reference [1]. In the weak coupling limit, the retarded interactions can be neglected and the PWC tends to a WC of electrons, which has no phonon instability [10]. Correspondingly, $r_s^{(inst)}$ vanishes (the critical density diverges) as $\alpha \to 0$. In the opposite strong coupling limit, where $\omega_{LO} \to 0$, we recover the results of reference [2]. Generally speaking, in all the range $\alpha > \alpha^*$, the results for $r_s^{(inst)}$ agree within a factor of two with the critical r_s for polaron dissociation, which confirms that there is an intimate connection between the instability of the long

Table 1. The density parameter $r_s^{(inst)}$ for the phonon instability, compared with the critical r_s for crystal melting and polaron dissociation in the mean field approximation (see Ref. [1]). Parameters are $\varepsilon_\infty = 5$, $\varepsilon_s = 30$ and $m^* = 2m_e$. The critical densities can be obtained from the relation $n = 1.6 \times 10^{24}/r_s^3$ cm^{-3}.

α	$r_s^{(melt)}$	$r_s^{(dis)}$	$r_s^{(inst)}$
0	960	-	-
3	537	-	19
7	52	-	18
10	-	32	16
100	-	31	15

wavelength transverse phonons and the mechanism of polaron dissociation studied in previous references. On the contrary, for $\alpha < \alpha^*$, the crystal melts due to the quantum fluctuations of the polarons before the occurrence of the phonon instability, and the latter is never realized in practice.

Figure 2 shows the evolution of ω_{pol} *vs.* the density in an intermediate situation, $\alpha = 7$. The behaviour is very similar to the case $\omega_{LO} = 0$ presented in reference [2], where it was shown that the frequency of the collective mode is gradually reduced due to the effects of the local Lorentz field (see also Refs. [3,13]).

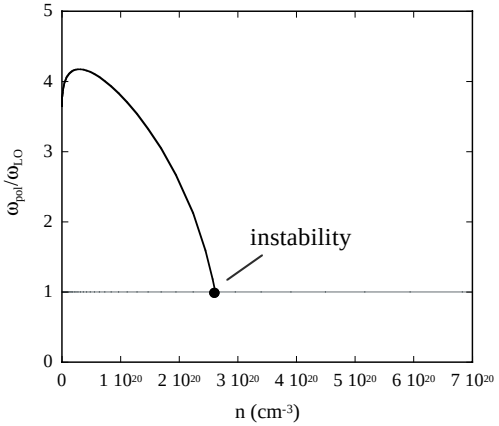

Fig. 2. Softening of the collective frequency ω_{pol} with density. The instability occurs when $\omega_{pol} = \omega_{LO}$. Parameters are $\alpha = 7$, $\varepsilon_\infty = 5$, $\varepsilon_s = 30$ and $m^* = 2m_e$.

4 Longitudinal dielectric constant and optical response

In this section, we derive the screening properties and the absorption spectrum of the PWC in the harmonic approximation. The determination of the dielectric constant of the model (7) follows the lines given in reference [10], and the optical conductivity is deduced from the $k = 0$ limit of this quantity.

4.1 Dielectric constant

The dielectric constant is defined as the response to a perturbing external charge, that we take of the form

$$\rho_{ext}(\mathbf{r}, t) = \rho_0 \mathrm{e}^{\mathrm{i}(\mathbf{k}\cdot\mathbf{r} - \omega t)}. \tag{30}$$

Taking into account the screening of the host polar lattice, which we schematize as $\varepsilon_{ph}(\omega) = \varepsilon_\infty(\omega^2 - \omega_{LO}^2)/(\omega^2 - \omega_{TO}^2)$, ω_{LO} and ω_{TO} being respectively the longitudinal and transverse optical phonon frequency, the force acting on an electron at site i can be written as

$$\mathbf{F}_i(t) = -e\mathbf{E}_{ext}(\mathbf{R_i}, \mathbf{t}) = \mathrm{i}\frac{4\pi\mathbf{k}}{k^2}\frac{e\rho_0}{\varepsilon_{ph}(\omega)}\mathrm{e}^{\mathrm{i}(\mathbf{k}\cdot\mathbf{r} - \omega t)}\,. \tag{31}$$

Introducing this driving force into the equations of motion (23) and solving for the induced displacement $u_{\mathbf{k}\lambda}$ we obtain

$$u_{\mathbf{k}\lambda} = \frac{\mathrm{i}4\pi e\rho_0}{\varepsilon_{ph}(\omega)}\frac{(\mathbf{k}\cdot\epsilon_{\mathbf{k}\lambda})^2}{k^2}\frac{\omega_{LO}^2 - \omega^2}{[\Omega_{int}^2(\mathbf{k},\lambda) - \omega^2]\,[\Omega_{ext}^2(\mathbf{k},\lambda) - \omega^2]}.$$

The longitudinal dielectric constant can be obtained by comparing the total charge associated to the displacement $u_{\mathbf{k}\lambda}$ and the perturbing charge distribution, namely $\varepsilon_L(\mathbf{k},\omega) = \rho_{ext}/\rho_{tot}$ (for more details, see Ref. [10,2]). The result for the PWC reads

$$\frac{1}{\varepsilon_L(\mathbf{k},\omega)} = \frac{1}{\varepsilon_{ph}(\omega)}\left\{1 + \frac{\omega_p^2}{\varepsilon_\infty}\sum_\lambda \frac{(\mathbf{k}\cdot\epsilon_{\mathbf{k}\lambda})^2}{k^2} \times \frac{\omega^2 - \omega_{TO}^2}{[\Omega_{int}^2(\mathbf{k},\lambda) - \omega^2]\,[\Omega_{ext}^2(\mathbf{k},\lambda) - \omega^2]}\right\}. \tag{32}$$

This expression can be simplified in the following limits:

4.1.1 Long wavelengths (k = 0)

Since in this limit $\mathbf{k}\cdot\epsilon_{\mathbf{k}t} = 0$, the transverse modes do not enter in the sum over λ, and the dielectric constant reduces to

$$\varepsilon_L(0,\omega) = \varepsilon_{ph}(\omega)\frac{\left[\omega^2 - \Omega_{int}^2(0,\ell)\right]\left[\omega^2 - \Omega_{ext}^2(0,\ell)\right]}{\omega^2\left[\omega^2 - \omega_{pol}^2\right]}\,. \tag{33}$$

This result is sketched in Figure 3. In this limit, the transverse and longitudinal dielectric constants become equal, and the zeros and poles of (33) correspond respectively to the longitudinal and transverse collective modes.

The pole at $\omega = 0$ corresponds to a transverse sliding mode of the PWC, which is equivalent to the sliding mode of a CDW [14]. This mode is expected to be current-carrying when the PWC is not pinned, as is the case in our jellium model, leading to an infinite d.c. conductivity. In real materials, however, this excitation can be shifted to higher frequencies due to the pinning by impurities and commensurability with the host lattice. As a consequence, the infinite d.c. conductivity is destroyed and the infinite (negative) dielectric constant at $\omega = 0$ is replaced by a large (positive) value below some threshold

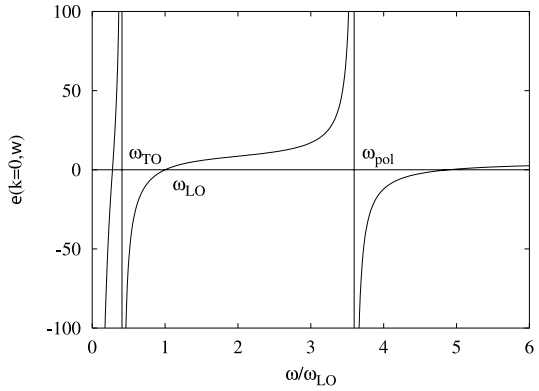

Fig. 3. Dielectric function of the PWC at $k = 0$, with the same parameter set as in Figure 2 at $n = 5 \times 10^{19}$ cm^{-3}. From the Lyddane-Sachs-Teller relation, the transverse phonon frequency is $\omega_{TO} = 0.41\omega_{LO}$.

frequency Δ_{pin} characteristic of the pinning process (this effect can be introduced in our model in a phenomenological way by taking the frequency $\Omega_{ext}(\mathbf{k}, \omega)$ of the transverse mode to be equal to Δ_{pin} at low k).

4.1.2 Static limit ($\omega = 0$)

In this limit, equation (33) becomes

$$\frac{1}{\varepsilon_L(\mathbf{k}, 0)} = \frac{1}{\varepsilon_s}\left\{1 - \frac{\omega_p^2}{k^2}\sum_{\lambda}\frac{(\mathbf{k}\cdot\epsilon_{\mathbf{k}\lambda})^2}{\omega^2(\mathbf{k},\lambda)}\right\}. \qquad (34)$$

By comparing this expression with the result of reference [10], we see that the static response of the PWC in the jellium model is identical to the ordinary WC, with an enhancement ε_s due to the host phonon screening.

4.1.3 Sign of the dielectric constant

By taking a simple model for the phonon dispersion of the WC of electrons, we can study the sign of the longitudinal dielectric constant of the PWC in the $(\mathbf{k}, \omega)$ space. The model consists of two transverse acoustical branches $\omega(\mathbf{k}, t) = ak$ and one longitudinal optical branch $\omega(\mathbf{k}, l) = \sqrt{\omega_p^2 - 2a^2k^2}$, in order to fulfill the Kohn sum rule. An estimate of the parameter a (the velocity of sound) can be obtained by arguing that the value of the transverse frequency at the end of the Brillouin zone ($k_0 = \pi/R_s$), must be equal to some fraction of the plasma frequency (we shall take $a = 0.1\omega_p/k_0$). Substituting these expressions into equation (24) we get the eigenfrequencies of the PWC, that we use in equation (32) to determine the dielectric constant as a function of the density. For simplicity, we restrict $\mathbf{k}$ to vary along one of the principal axes of the crystal, where by symmetry the transverse polarisation vectors obey $\epsilon_{\mathbf{k}t} \cdot \mathbf{k} = 0$.

As can be seen in Figure 4, the regions where $\varepsilon_L < 0$ expand with increasing density. When one approaches the critical density, the dielectric constant is negative in all the region below the frequency $\Omega_{int}(k, \ell) \simeq \omega_p/\sqrt{\varepsilon_\infty}$.

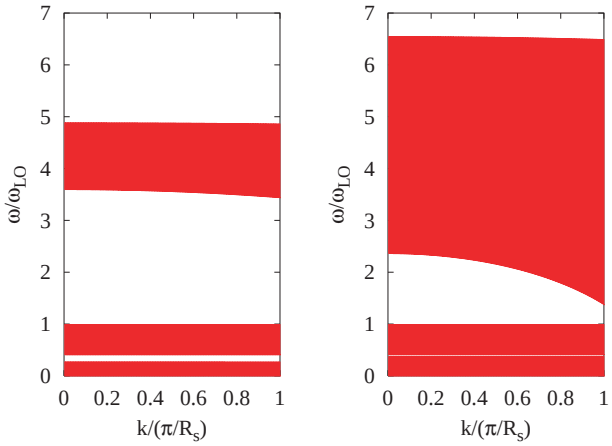

Fig. 4. Regions in the (k, ω) plane where the dielectric constant is negative, which are located between the zeros and the poles of ε_L. Left panel: $n = 5 \times 10^{19}$ cm^{-3}; right panel: $n = 1.7 \times 10^{20}$ cm^{-3}. Parameters are the same as in Figure 1.

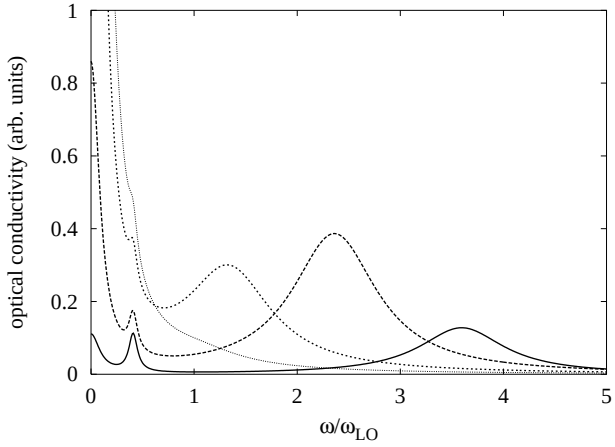

Fig. 5. Optical conductivity of the PWC at various densities: $n = 0.5$, 1.7, 2.3 and 2.4×10^{20} cm^{-3}, with the same parameter set as in Figure 2 (the instability occurs at $n^{(inst)} = 2.7\times10^{20}$ cm^{-3}). We have introduced the phenomenological linewidths $\Gamma_{pol} = \omega_{LO}$, $\Gamma_0 = \Gamma_{ph} = 0.1\omega_{LO}$. Note the shift of the polaron peak, and the transfer of spectral weight taking place between the feature at ω_{pol} and the zero frequency peak. When the polaron lattice is pinned by impurities or by commensurability effects, the $\omega = 0$ sliding mode moves to a finite frequency $\omega = \Delta_{pin}$.

4.2 Optical conductivity

The real part of the optical conductivity is related to the imaginary part of the transverse dielectric constant by

$$\sigma(\omega) = \frac{\omega}{4\pi}\mathrm{Im}\ \varepsilon(k = 0, \omega)\,. \qquad (35)$$

We remark that expression (33) is perfectly real, and therefore it can only lead to delta-like absorption peaks located at its poles $\omega = 0$ and $\omega = \omega_{pol}$. As was stated in the previous section, at finite values of the e-ph coupling, the collective excitations have an intrinsic lifetime which cannot be obtained within the quadratic model (7). We

thus introduce phenomenological linewidths by putting $\omega \to \omega + \mathrm{i}\Gamma$. Substituting the values of $\Omega_{int}(0,\lambda)$ and $\Omega_{ext}(0,\lambda)$ given by equation (24), we obtain

$$\sigma(\omega) = \sigma_{ph}(\omega) + \sigma_{pol}(\omega) + \sigma_0(\omega) \tag{36}$$

where the first term is the response from the phonons of the host medium, the second term comes from the collective transverse excitation at ω_{pol}, and the last term is the contribution from the sliding mode centered at $\omega = 0$. As was stated in the previous section, if the PWC is pinned due to disorder or lattice commensurability, a gap opens in the optical conductivity, and the zero-frequency peak is shifted to $\omega = \Delta_{pin}$. Therefore, the optical response of the polaron Wigner crystal is characterized by the emergence of two collective-mode peaks, a polaron peak located above the host phonon frequency and a sliding (or pinning) peak around or below the phonon frequency.

Assuming that the above contributions are well separated from one another (*i.e.* that the peaks do not overlap), these take the form of Lorentz peaks:

$$\sigma_{ph}(\omega) = \frac{\varepsilon_s - \varepsilon_\infty}{4\pi}\omega_{TO}^2 \frac{\Gamma_{ph}/4}{(\omega-\omega_{TO})^2 + \Gamma_{ph}^2/4} \tag{37}$$

$$\sigma_{pol}(\omega) = \left(1 - \frac{\omega_{LO}^2}{\omega_{pol}^2}\right)\frac{\omega_p^2}{4\pi}\frac{\Gamma/4}{(\omega-\omega_{pol})^2 + \Gamma^2/4} \tag{38}$$

$$\sigma_0(\omega) = \left(\frac{\omega_{LO}^2}{\omega_{pol}^2}\right)\frac{\omega_p^2}{4\pi}\frac{\Gamma_0}{\omega^2 + \Gamma_0^2}. \tag{39}$$

The total spectral weight associated with the absorption of the PWC is $\omega_p^2/8$, and it obeys the conductivity sum rule. At low density, if $\alpha > \alpha^*$, one typically has $\omega_{pol} \gg \omega_{LO}$ and almost all the spectral weight is carried by the polaron peak in $\sigma_{pol}(\omega)$, as can be seen from equation (38). As the density is increased, some spectral weight is transferred to the sliding mode at $\omega = 0$ ($\omega = \Delta_{pin}$ in the pinned case), until the polaron peak eventually disappears at the instability point.

5 Conclusion

In this paper, we have analyzed the melting of a polaron Wigner crystal in the strong electron-phonon coupling limit, taking into account the dipole-dipole interactions between localized electrons, *i.e.* going beyond the mean field approach developed in reference [1]. We have shown that the system undergoes a phonon instability due to the effect of the Lorentz local field, which confirms the polaron dissociation scenario introduced previously. The instability has an optical signature: the polaron peak in the optical conductivity *shifts towards low frequencies* as the doping density is increased. This red-shift should be accompanied by the rise of a collective peak in the far infrared, whenever the polaron Wigner crystal is pinned by disorder or by commensurability effects. This complex behaviour has been experimentally observed in NCCO [5], which suggests that *both* the polarons and the unscreened long-range Coulomb interactions should play an important role in the high Tc compounds. We suggest that the same behaviour could be observed in other superconducting materials.

We have calculated the dielectric constant of the PWC, and shown that it is negative on a large region of $(\mathbf{k},\omega)$. This indicates the possibility of reaching a superconducting phase, if carriers of different nature – localized polarons and free electrons – can coexist close to the instability density. In such a regime, the itinerant electrons would be paired through the high frequency internal vibrations of the polaron lattice, which would act as the phonons in the ordinary BCS theory. The large collective frequencies of the polaron crystal would naturally lead to a high superconducting transition temperature. Eventually, at higher doping levels, the polaron clouds are completely screened by the free electrons, so that the mixed phase (and the superconducting phase) should disappear in favor of a more conventional metal [18].

It should be kept in mind that our results are based on the continuous Fröhlich model in the low density regime and at strong electron-phonon coupling, which misses many essential physical features of the cuprates. Indeed, we are not considering the spin degrees of freedom, although these are certainly responsible for most of the anomalous properties of the normal state. We are not taking into account the formation of stripes, although the practical realization of the mixed phase conjectured above could well involve such form of microscopic phase segregation. On the other hand, the inclusion of other ingredients, such as the anisotropy of electronic bands in the cuprates, or the discretization of the lattice, would increase the tendency to localization and are therefore expected to push the system towards the present "polarization catastrophe" scenario.

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