

Optical properties of an interacting large polaron gas

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Abstract. The normal state conductivity, $\sigma(\omega)$, of a system of interacting large polarons is calculated within the Random Phase approximation and some numerical results are presented. The behaviour of the optical absorption as a function of the charge carrier density and of the temperature is analyzed for different values of the electron-phonon coupling constant. It is shown that $\sigma(\omega)$ exhibits features similar to those observed in the infrared spectra of the cuprates.

PACS. 71.38.+i Polarons and electron-phonon interactions

1 Introduction

The infrared absorption of large polarons is a well studied problem [1]. However, a large part of the studies has been focused mainly on the very low density regime (non interacting polaron limit) which is suitable for not heavily doped polar semiconductors and ionic insulators. In this regime the most accurate approach has been discussed by Devreese *et al.* [2] starting from the expression of the impedance function derived by Feynmann [3]. More recently the interest for this problem has been renewed in connection with the infrared absorption of cuprates in the normal phase [4,5]. In these materials the interaction among polarons can be important. It is, therefore, of interest to consider the effect of the Coulomb long range interaction on the large polaron absorption. To our knowledge this effect has not been yet considered within the Devreese-Feynman approach to the polaron absorption.

The aim of this paper is to calculate the conductivity $\sigma(\omega)$ of a large polaron gas within the random phase approximation (RPA) by using, for the lowest order polarization insertion, the propagator of the Feynman polaron model [6,7]. We find that it is possible to identify in $\sigma(\omega)$ different contributions with features common to some structures of the infrared spectra of cuprates. In particular we observe a significant displacement of spectral weight from higher to lower frequencies when the polaron density increases.

2 The model

We consider a system made of band electrons in the effective mass approximation interacting with non dispersive LO phonons and repelling each other through the

Coulomb potential screened by the background high frequency dielectric constant ϵ_∞ [8]. The electron-phonon (e-ph) interaction is assumed to be:

$$H_{e-ph} = \sum_{\mathbf{p}\mathbf{q}} M_q c_{\mathbf{p}+\mathbf{q}\sigma}^\dagger c_{\mathbf{p}\sigma} (a_{\mathbf{q}} + a_{-\mathbf{q}}^\dagger) \quad (1)$$

where

$$M_q = -i\hbar\omega_0 \frac{R_p^{1/2}}{q} \left(\frac{4\pi\alpha}{V} \right)^{1/2} \quad (2)$$

is the Fröhlich e-ph matrix element [9].

In the above expressions $c_{\mathbf{p}\sigma}$ ($c_{\mathbf{p}\sigma}^\dagger$) and $a_{\mathbf{q}}$ ($a_{\mathbf{q}}^\dagger$) indicate, respectively, the annihilation (creation) operators for electrons and phonons, V is the system's volume, α is the Fröhlich e-ph coupling constant, $R_p = (\hbar/(2m\omega_0))^{1/2}$ is the polaron radius and ω_0 is the LO phonon frequency.

The normal state conductivity of this system, $\sigma(\omega)$, is related to the retarded form of the current-current correlation function $\Pi(\omega)$ by the Kubo formula [10]:

$$\sigma(\omega) = \frac{i}{\omega} \left(\frac{ne^2}{m} + \Pi(\omega) \right) \quad (3)$$

where n is the density of charge carriers. In order to estimate the conductivity we follow the approach suggested by Feynman [3] and adopted with success by Devreese *et al.* [2] for the optical absorption of non interacting large polarons. Within this approach the real part of the conductivity is given by:

$$\text{Re}[\sigma(\omega)] = -G(\omega) \frac{\text{Im}\Sigma(\omega)}{(\omega - \text{Re}\Sigma(\omega))^2 + (\text{Im}\Sigma(\omega))^2} \quad (4)$$

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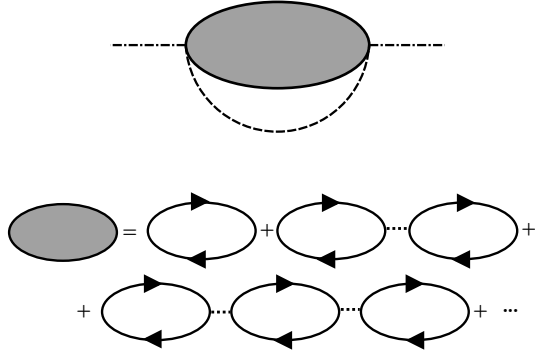


Fig. 1. Diagrammatic representation of the equation (6). The solid line indicates the polaron propagator, the phonon is given by the dashed line, the dotted line describes the Coulomb interaction and the dotted-dashed line represents the incident photon.

where $G(\omega) = ne^2/m$ and $\Sigma(\omega) = \Pi^0(\omega)\omega/G(\omega)$ (for the definition of $\Pi^0(\omega)$ see below). A justification of equation (4) has been given by Feynman [3], while a more general derivation can be obtained by making use of the Mori-Zwanzig [11] projection operator technique, as shown by Devreese *et al.* [12] and Maldague [13]. It is worth to note that the sum rule for the real part of the conductivity:

$$\int_0^\infty d\omega \text{Re}[\sigma(\omega)] = \frac{\pi ne^2}{2m} \quad (5)$$

is satisfied by using equation(4).

To evaluate $\Pi^0(\omega)$ we use the set of diagrams shown in Figure 1, *i.e.*, we consider a R.P.A.-like approximation for polarons interacting each other through the Coulomb potential screened by ϵ_∞ (v_q^∞). Following Mahan [14], we write:

$$\Pi^0(\omega) = \frac{-e^2}{m^2\omega^2} \int \frac{d^3q}{(2\pi)^3} q_\mu^2 [N(\mathbf{q}, \omega) - N(\mathbf{q}, 0)] \quad (6)$$

where

$$N(\mathbf{q}, \omega) = \int_{-\infty}^{\infty} \frac{du}{\pi} \int_{-\infty}^{\infty} \frac{ds}{\pi} \frac{1}{v_q^\infty} \text{Im} \left[\frac{1}{\epsilon(\mathbf{q}, u)} \right] \times \text{Im} \left[\bar{M}_q^2 D(\mathbf{q}, s) \right] \left(\frac{n_B(s) - n_B(u)}{u - s + \omega + i\delta} \right). \quad (7)$$

In the equations (6, 7) $\mu = x, y$ or z , $\epsilon(\mathbf{q}, \omega)$ is the dielectric function, \bar{M}_q is the renormalized e-ph matrix element, $D(\mathbf{q}, \omega)$ is the phonon Green function and $n_B(\omega)$ is the boson occupation number. In our model the dielectric function of the system is assumed to be (see Fig. 1):

$$\epsilon(\mathbf{q}, \omega) = 1 - v_q^\infty P(\mathbf{q}, \omega) \quad (8)$$

where $P(\mathbf{q}, \omega)$, the lowest order polarization insertion, can be expressed in terms of the polaron spectral weight

function $A(\mathbf{q}, \omega)$:

$$P(\mathbf{q}, \omega) = \frac{2}{\hbar^2} \int \frac{d^3p}{(2\pi)^3} \int_{-\infty}^{\infty} \frac{du}{2\pi} \int_{-\infty}^{\infty} \frac{ds}{2\pi} A(\mathbf{p}, s) \times A(\mathbf{p} + \mathbf{q}, u) \left(\frac{n_F(s) - n_F(u)}{\omega + s - u + i\delta} \right). \quad (9)$$

Here $n_F(\omega)$ is the fermion occupation number. For simplicity we shall use in the equation (7) the unperturbed phonon Green function $D^0(\omega) = 2\omega_0/(\omega^2 - \omega_0^2 + i\delta)$ and we shall ignore the frequency dependence of the dielectric function in the expression of the renormalized e-ph matrix element assuming:

$$\bar{M}_q = \frac{M_q}{\epsilon(\mathbf{q}, 0)}. \quad (10)$$

Until this point we have not specified the polaron spectral weight function that appears in the equation (9). It is well known that, from a theoretical point of view, the problem of an electron in a polar crystal which interacts with the longitudinal optical modes of lattice vibrations has not been solved exactly. However, it is universally recognized that of all published theories that of Feynman [6], which uses a variational method based on path integrals, gives the best available results in the entire range of the coupling constant α . For this reason we choose the spectral weight function of the Feynman polaron model [6, 7]:

$$A(q, \omega) = \sum_{l=-\infty}^{\infty} \delta \left[\omega - \left(\frac{\hbar^2 q^2}{2M_T} + lv \right) \right] \exp \left[\frac{l\beta v}{2} \right] \times \exp \left[-\frac{q^2 R}{M_T} (2n_B(v) + 1) \right] 2\pi I_l(z) \quad (11)$$

where $R = (v^2/w^2 - 1)/v$, $M_T = v^2/w^2$, $z = \frac{2q^2 R}{M_T} \times [n_B(v)(n_B(v) + 1)]^{1/2}$ and I_l are the Bessel functions of complex argument. The dimensionless parameters v and w are related to the mass and the elastic constant of the model [6,7], in which the electron is coupled *via* a harmonic force to a fictitious second particle simulating the phonon degrees of freedom. The values of v and w can be obtained by the variational approach described by Feynman [6] and Schultz [7]. In particular, at $T = 0$, $A(q, \omega)$ takes the form:

$$A(q, \omega) = 2\pi \sum_{l=0}^{\infty} \delta \left[\omega - \left(\frac{\hbar^2 q^2}{2M_T} + lv \right) \right] \times e^{-\frac{q^2 R}{M_T}} \left(\frac{q^2 R}{M_T} \right)^l / l! \quad (12)$$

The spectral weight follows a Poissonian distribution and it is maximum for an excitation involving a number l of phonons of order of $l \sim \frac{q^2 R}{M_T}$. We note that, in all the numerical results which will be shown in this paper, the terms with $-6 \leq l \leq 6$ in the equation (11)

and the first six term in the equation (12) are enough to a good convergence of the real and imaginary parts of the correlation function $\Pi^0(\omega)$ in the frequency range of interest ($\omega \leq 10\omega_0$).

We want to note that the approach proposed in this paper restores, when $n \rightarrow 0$, the well known results of the optical absorption of a single polaron [12] and allows to introduce within the R.P.A. approximation the effects of the polaron-polaron interaction. This approach is expected to be valid when the formation of bipolaron states is not favored ($\eta = \epsilon_\infty/\epsilon_0 > 0.01$ or $\eta < 0.01$ and $\alpha < 6 - 7$) [15]. In fact, in this situation the overlap between the wells of two particles can be neglected and the system is well approximated by an interacting large polaron gas. Of course the proposed approach is valid in the density range where we can exclude Wigner-like localization. As discussed by Quemerais *et al.* [16] for coupling constant not larger than $\alpha = 6$ and density $n \geq 10^{18} \text{ cm}^{-3}$ our approach is justified.

3 The results

In Figure 2a is reported the optical absorption per polaron as a function of the frequency for different values of the charge carrier density at $T = 0$. Three different structures appear in the normal state conductivity: a) a zero frequency delta function contribution; b) a strong band starting at $\omega = \omega_0$ that is the overlap of two components: a contribution from the intraband process and a peak due to the polaron transition from the ground state to the first relaxed excited state; c) a smaller band at higher frequency due to the Frank-Condon transition of the polaron. The identification and characterization of these structures stem from the detailed analysis of the polaron absorption in the case of non interacting large polarons [1]. Increasing the charge carrier density, we find that the large polaronic band due to the excitation involving the relaxed states (b contribution) tends to move towards lower frequencies while its intensity decreases in favor of the rise of a Drude-like term around $\omega = 0$ (see Figs. 2a and 2b).

In particular, at $T = 0$ the Drude weight D , *i.e.*, the coefficient of the zero frequency delta function contribution, is determined making use of the sum-rule for the real part of the conductivity (see Eq. (5)). This allows an estimate of the effective polaron mass $M_{\text{eff}}(n)$ as a function of the electron density, being the Drude weight D related to $M_{\text{eff}}(n)$ by the following expression:

$$D = \frac{\pi n e^2}{2M_{\text{eff}}(n)}. \quad (13)$$

In Table 1 the values of $M_{\text{eff}}(n)$ for $\alpha = 5$ and $\alpha = 6$ are reported. It is evident that, increasing the charge carrier density, the screening of the e-ph interaction increases and the polaron mass is reduced, tending to the band mass for large values of n . This behavior is a confirmation of the trend obtained at weaker coupling in a model that includes polaron screening but neglects exchange effects [17].

Table 1. Effective polaron mass, in units of the electron band mass, as a function of the charge carrier density. The value of n_0 is $n = 1.4 \times 10^{-5}$ in terms of R_p^{-3} , R_p being the Fröhlich polaron radius.

	n_0	$10 n_0$	$10^2 n_0$	$10^3 n_0$
$\alpha=5$	3.56	3.07	2.19	1.42
$\alpha=6$	6.21	5.28	3.58	1.94

In Figure 3a we plot the normal state conductivity for larger electron densities at $T/T_D = 0.5$, where $T_D = \hbar\omega_0/k_B$ and k_B is the Boltzmann constant. Increasing the charge density, we find that the optical absorption, in agreement with experimental data in the metallic phase of the cuprates [18–20], is more and more controlled by the Drude-like term and, only for very high electron densities, no signature of the b) contribution is left.

The behavior of the polaron absorption with temperature is also of interest. As shown in Figure 3b, with increasing T there is a transfer of spectral weight towards higher frequencies. Moreover, the intensity of the contributions b) and c) increases with decreasing the temperature saturating at $T/T_D \simeq 0.5$.

4 Discussion

The effects shown are very intriguing in connection of recent measurement of infrared absorption in cuprates [4]. In fact several experiments on the infrared response of the cuprates have pointed out that the optical absorption exhibits features which are common to many families of high- T_c superconductors.

In particular, the infrared normal state conductivity does not diminish with frequency as rapidly as one expects from a simple Drude picture [22]. This behavior has been interpreted in terms of two different models: the anomalous Drude model [23] and the multi-component or Drude-Lorentz modelé [24]. In the former approach, the infrared conductivity has been attributed to a contribution from free charge carriers with an ω -dependent scattering rate (arising, for example, by strong interactions with spin waves [22]). In the Drude-Lorentz approach, instead, one assumes that the absorption is the result of the superposition of different structures which can be identified in the conductivity spectra: 1) a Drude-like peak centered at zero frequency; 2) infrared-active vibrational modes (IRAV) [25]; 3) a broad excitation in the infrared band which is constituted by two different components: one temperature-independent around 0.5 eV ($\sim 4000 \text{ cm}^{-1}$) [19,26] and the other strongly dependent on temperature with a peak around 0.1 eV ($\sim 1000 \text{ cm}^{-1}$) [27,28] (d band); 4) the charge transfer band (CT) in the visible range which is attributed to the charge transfer transitions between O_{2p} and Cu_{3d} states. The Drude, the d band and IRAV contributions depend on the charge density injected by the doping and a significant transfer of spectral weight from the d band and Irav contributions

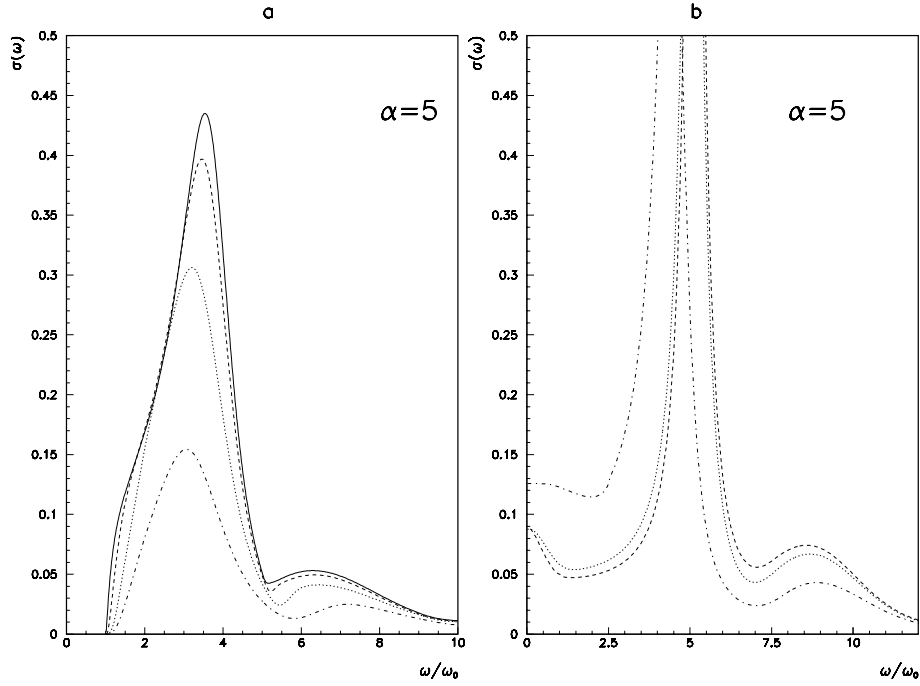


Fig. 2. a) Optical absorption *per* polaron, at $T = 0$, as a function of the frequency for different electron densities: $n = 1.4 \times 10^{-5}$ (solid line), $n = 1.4 \times 10^{-4}$ (dashed line), $n = 1.4 \times 10^{-3}$ (dotted line), $n = 1.4 \times 10^{-2}$ (dotted-dashed line); b) Optical absorption *per* polaron at finite temperature, $T/T_D = 0.5$, for different values of the charge carrier density: $n = 1.4 \times 10^{-4}$ (dashed line), $n = 1.4 \times 10^{-3}$ (dotted line), $n = 1.4 \times 10^{-2}$ (dotted-dashed line). The value of $\epsilon_0/\epsilon_\infty$ is 3.4. The electron density is measured in units of R_p^{-3} , R_p being the Fröhlich polaron radius, and the conductivity $\sigma(\omega)$ is expressed in terms of $ne^2/m\omega_0$. The value of R_p^{-3} is $7 \times 10^{20} \text{ cm}^{-3}$ when $\omega_0 = 30 \text{ meV}$ and $m = m_e$, where m_e is the electron mass.

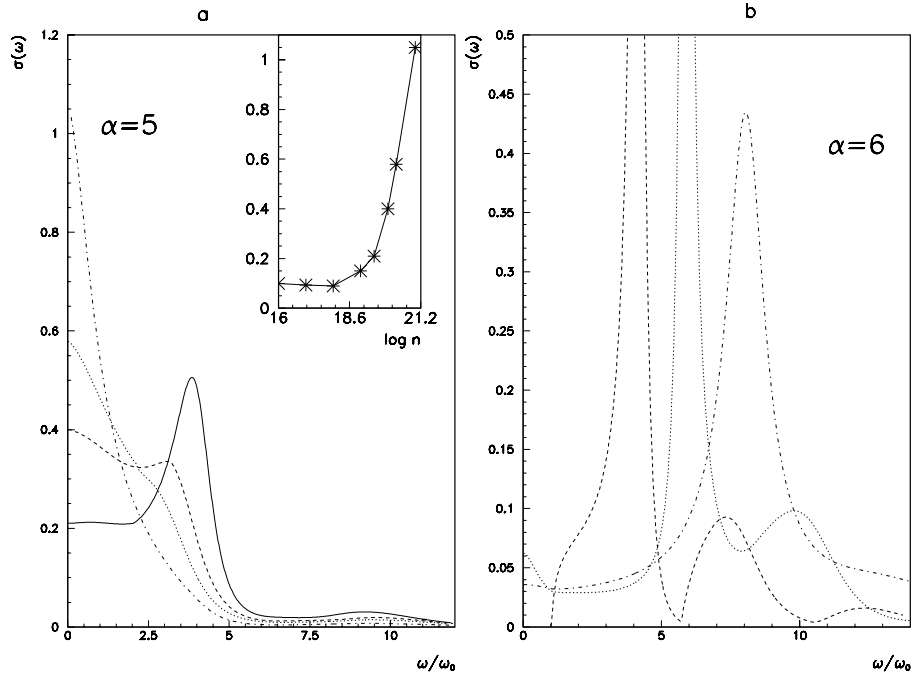


Fig. 3. a) Optical absorption *per* polaron at finite temperature, $T/T_D = 0.5$, for different values of the charge carrier density: $n = 4.5 \times 10^{-2}$ (solid line), $n = 1.4 \times 10^{-1}$ (dashed line), $n = 0.28$ (dotted line), $n = 1.4$ (dotted-dashed line). The electron density is measured in units of R_p^{-3} , R_p being the Fröhlich polaron radius, and the temperature is given in units of $T_D = \hbar\omega_0/k_B$, k_B being the Boltzmann constant. In the inset is plotted the polaron mobility (μ), *i.e.* $\text{Re}\sigma(\omega \rightarrow 0)$, as a function of the charge density for $\omega_0 = 30 \text{ meV}$, $\epsilon_\infty = 3$ and $m = m_e$ where m_e is the electron mass; b) Optical absorption of a single Feynman polaron for different values of T : $T/T_D = 0$ (dashed line), $T/T_D = 0.5$ (dotted line), $T/T_D = 1$ (dotted-dashed line).

to the Drude peak is observed increasing the doping. To the d band and the IRAV, which appear when extra charge are injected into the lattice of a cuprate, it has been assigned a polaronic origin both in electron-doped and hole doped compounds [28,29]. However, there is no general consensus on the type of polarons involved in the absorption. In particular, the 1000 cm^{-1} feature has been attributed to optical transitions involving small [30], large polarons [5,31] or both types of polarons [32]. In any case, as first noted by Bi and Eklund [33] in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_{4+\delta}$, the polaron tend to be small in the dilute limit (small x) whereas in the limit of large x the polaron tend to expand, *i.e.*, the polaron size increases with x tracking a dopant-induced transformation to metallic conductivity.

Finally we note that, recently, Calvani *et al.* [34] have measured the infrared absorption of four different perovskite oxides observing an opposing behaviour between the cuprates and the non cuprates. In particular, in $\text{La}_2\text{NiO}_{4+y}$ and in SrMnO_{4+y} , in which there is a strong evidence of the presence of small polarons, the minimum of the d band deepens at low T by a transfer of spectral weight towards higher energies. On the contrary, in the slightly and heavily doped cuprates at low temperatures the minimum of the d band tends to be filled by a transfer of spectral weight in the opposite direction (see also Li *et al.* [35]) as it happens for a large polaron system previously described.

The above scenario shows that the normal state conductivity of an interacting large polaron system presents features common to the optical absorption of the cuprates [36]. Even if the similarities found are not conclusive in assigning to large polarons the main role in the absorption of cuprates - more accurate calculations are needed to attempt a quantitative comparison between theory and experiments - we believe that the inclusion of long range Coulomb interactions among polarons is essential in understanding whether infrared absorption in cuprates can be assigned to large polarons.

5 Conclusions

In this paper we calculated the normal state conductivity of a large polaron system including the electrostatic polaron-polaron interaction within the R.P.A. approximation. The approach recovers the optical absorption of non interacting Feynman polaron [12] and allows to introduce in a perturbative way the many polaron effects. With increasing charge carrier density, we have found evidence of a transfer of spectral weight of the optical absorption towards lower frequencies: the Drude-like contribution increases as well as the screening effects of the e-ph interaction. Moreover, with increasing temperature, there is a transfer of spectral weight of the infrared absorption towards higher frequencies. Both these behaviours are observed in the infrared spectra of cuprates.

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36. In this paper we calculate the normal state conductivity of a homogeneous three dimensional (3D) system. On the other hand it is well known that almost all of the cuprates show strong anisotropy and, when the electric field is polarized along the conducting planes, the optical response is very close to that of a homogeneous 2D system. In this paper we assume that our 3D results are qualitatively equal to those of a homogeneous 2D system. This assumption is supported by the fact that, for non interacting polarons, a simple scaling relation connects the 2D and the 3D impedance functions (see F.M. Peeters and J.T. Devreese, Phys. Rev. B **36**, 4442 (1987)). The inclusion of the anisotropy is currently under study and it will be discussed in a future work.