# Electron-screening effects on the self-trapping of polarons

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In a polar semiconductor with a number of electrons or holes in the conduction or valence band, the interactions of an external charge with the longitudinal optical phonon are screened by the electron/hole density. Consequently the self-energy depends on the density. We show that, for any value of the Fröhlich electron-phonon coupling constant $\alpha$, an electronic density $n^*$ can be found such that for density $n>n^*$ the Lee polaron theory for the intermediate coupling (not self-trapped polarons) gives self-energies lower than those of the strong coupling (self-trapped polarons). This is due to the static and dynamical screening introduced by the plasmon field on the electron-phonon interaction. This result is confirmed by applying the path-integral technique to the problem and calculating both the self-energy and the renormalized mass. It is found that the density $n^*$ depends on $\alpha$ and $\eta$ (the ratio of the high frequency dielectric constant to the static one). We note that typical charge carrier densities in high $T_c$ superconductors are larger than $n^*$ for a wide range of value of $\alpha$. This indicates that no self-trapped polarons might have a role in high $T_c$ superconductors.

The aim of this work is to show that the peculiar properties of the Fröhlich polaron (self-energy and mass) are strongly modified if a number of free charges screen the electron-electron and the electron-phonon interactions. In particular, a self-trapped polaron (strong coupling regime) becomes untrapped increasing the free charge density and this occurs for any value of the electron-phonon coupling constant.

It is well known that the polaron problem in the Fröhlich scheme[1] can be treated within a variational scheme in the so-called intermediate coupling regime[2] when the electron-phonon interaction

$$\alpha=\frac{1}{2}\,\frac{e^2}{\epsilon_\infty R_p}\,\frac{1-\eta}{\hbar\omega_l}$$

[$e$ is the electron charge, $\omega_l$ the optical longitudinal phonon frequency, $\eta=\epsilon_\infty/\epsilon_0$, $\epsilon_0$ and $\epsilon_\infty$ the static and high frequency dielectric constants, $R_p=(\hbar/2m\omega_l)^{1/2}$ the polaron radius, and $m$ the effective electronic band mass] is less than $\sim 10$ and in the strong coupling regime[3] for higher values of $\alpha$. Furthermore it is also well known that the Feynman path-integral formalism, applied to the polaron problem, gives for the self-energy values lower than those found with the variational methods;[4] in the limits of small and large $\alpha$ the Feynman results tend to the intermediate and strong coupling results, respectively.

Recently, in order to include static and dynamical screening effects into the large polaron theory, the problem of an external charge interacting with the longitudinal optical phonon and plasmon fields has been studied.[5]

The aim of this work is to show that, even if the polaron is self-trapped when the carrier density is small, increasing the electronic density, the static and dynamical screenings of the electron-phonon interaction delocalize the polaron.

The Hamiltonian of the system contains the kinetic energy of the charge, the free phonons and plasmons energies, a coupling term between the phonons and the plasmons, the Fröhlich[1] electron-phonon interaction, and the Overhauser[6] electron-plasmon interaction in the single pole approximation for the dispersive dielectric longitudinal function.

Through a canonical transformation, the Hamiltonian becomes that of an external charge interacting with two noninteracting boson fields,[5]

$$\begin{aligned}H=&\frac{p^2}{2m}+\sum_{\vec{k}}\hbar\Omega_1(k)\left(\alpha^\dagger_{\vec{k}}\alpha_{\vec{k}}+\frac{1}{2}\right)\\&+\sum_{\vec{k}}\hbar\Omega_2(k)\left(\beta^\dagger_{\vec{k}}\beta_{\vec{k}}+\frac{1}{2}\right)\\&+\sum_{\vec{k}}(\widetilde{V}_{k,1}e^{i\vec{k}\cdot\vec{r}}\alpha_{\vec{k}}+\text{H.c.})+\sum_{\vec{k}}(\widetilde{V}_{k,2}e^{i\vec{k}\cdot\vec{r}}\beta_{\vec{k}}+\text{H.c.})\end{aligned}\tag{1}$$

with

$$\widetilde{V}_{k,1}=iV_kF_1,$$

$$\widetilde{V}_{k,2}=-iV_kF_2,$$

$$V_k=-\hbar\omega_l\sqrt{\frac{4\pi\alpha}{V}}\frac{R_p^{1/2}}{k},$$

 

$$F_1=\lambda\frac{\sqrt{\frac{1-\eta}{R_1}}+\frac{R_1^2-1}{\sqrt{(1-\eta)R_1}}}{[(1-R_1^2)^2+\lambda^2(1-\eta)]^{1/2}},$$

$$F_2=\lambda\frac{\sqrt{\frac{1-\eta}{R_2}}+\frac{R_2^2-1}{\sqrt{(1-\eta)R_2}}}{[(1-R_2^2)^2+\lambda^2(1-\eta)]^{1/2}},$$

$$\Omega_i(k)=\omega_l R_i=\omega_l\left[\frac{1}{2}[\lambda^2+1+(-1)^{i+1}\times\sqrt{(\lambda^2-1)^2+4\lambda^2(1-\eta)}]\right]^{1/2},\quad i=1,2,$$

$$\lambda=\frac{\omega_p}{\sqrt{\epsilon_\infty}\omega_l},$$

where $\omega_p$ is the plasma frequency of an electron (hole) gas of density $n$ and $V$ is the volume of the system.

The variational procedure introduced by Lee[2] for the polaron problem can be generalized to this case. The unitary transformation

$$U=e^{i[\vec{Q}-\Sigma_{\vec{k}}\vec{k}(\alpha_{\vec{k}}^\dagger\alpha_{\vec{k}}+\beta_{\vec{k}}^\dagger\beta_{\vec{k}})]\cdot\vec{r}}$$

with $\hbar\vec{Q}$ eigenvalue of the total momentum operator

$$\vec{P}=\vec{p}+\sum_{\vec{k}}\hbar\vec{k}(\alpha_{\vec{k}}^\dagger\alpha_{\vec{k}}+\beta_{\vec{k}}^\dagger\beta_{\vec{k}})$$

allows us to eliminate the electronic operators from the transformed Hamiltonian. This transformation shows that, in this approximation, the polaron moves as a free particle with wave vector $\vec{Q}$. The ground state energy and wave function are found with the variational method using the trial wave function

$$|\psi\rangle=U_1U_2|0\rangle$$

with $|0\rangle$ the vacuum for the two boson fields and

$$U_1=e^{\Sigma_{\vec{k}}(f_{\vec{k},1}\alpha_{\vec{k}}-f_{\vec{k},1}^*\alpha_{\vec{k}}^\dagger)},$$

$$U_2=e^{\Sigma_{\vec{k}}(f_{\vec{k},2}\beta_{\vec{k}}-f_{\vec{k},2}^*\beta_{\vec{k}}^\dagger)}.$$

The functions $f_{\vec{k},1}$ and $f_{\vec{k},2}$ are found by the energy minimization. At $\vec{Q}=0$ the energy minimum (the self-energy) is given by

$$E_{\rm self}^{in}=-\sum_{i=1}^{2}\sum_{\vec{k}}\frac{|\widetilde{V}_{k,i}|^2}{A_i}\tag{2}$$

and the plasma-polaron mass by

$$m^*=m\left[1+2\sum_{i=1}^{2}\sum_{\vec{k}}\frac{|\widetilde{V}_{k,i}|^2}{A_i^3}\frac{\hbar^2}{m}(\vec{k}\cdot\hat{\vec{Q}})^2\right],\tag{3}$$

where $\hat{\vec{Q}}=\vec{Q}/Q$ and

$$A_i=\hbar\Omega_i(\vec{k})+\frac{\hbar^2k^2}{2m}.$$

All the above quantities depend on the electronic density so that the plasma-polaron self-energy and mass are also density dependent. If the plasmon frequency is not spatially dispersive, $E_{\rm self}^{in}$ and $m^*$ are given by

$$E_{\rm self}^{in}=-\alpha\hbar\omega_l\left[\frac{F_1^2}{\sqrt{R_1}}+\frac{F_2^2}{\sqrt{R_2}}\right],\tag{4}$$

$$m^*=m\left[1+\frac{\alpha}{6}\left(\frac{F_1^2}{R_1^{3/2}}+\frac{F_2^2}{R_2^{3/2}}\right)\right].\tag{5}$$

We can also generalize the theory of the polaron in the strong coupling regime,[3] taking as trial variational wave function

$$|\psi\rangle=|f_1\rangle|f_2\rangle\phi(r),\tag{6}$$

where $|f_1\rangle$ depends only on the operators $\frac{1}{2}ak$ and $\alpha_{\vec{k}}^\dagger$, $|f_2\rangle$ on $\beta_{\vec{k}}$ and $\beta_{\vec{k}}^\dagger$, and

$$\phi(r)=\frac{\gamma^{3/2}}{\pi^{3/4}}e^{-(\gamma^2/2)r^2}$$

is the envelope self-trapped electronic wave function. In this case the self-energy is found minimizing the energy with respect to states $|f_1\rangle$, $|f_2\rangle$, and the parameter $\gamma$. The minimization with respect to $|f_1\rangle$ and $|f_2\rangle$ gives

$$E=\left\langle\phi\left|\frac{p^2}{2m}\right|\phi\right\rangle-\sum_{i=1}^{i=2}\sum_{\vec{k}}\frac{|\widetilde{V}_{k,i}|^2|\rho_k|^2}{\hbar\Omega_i(k)}\tag{7}$$

with

$$\rho_k=\langle\phi|e^{i\vec{k}\cdot\vec{r}}|\phi\rangle.$$

In the nondispersive case for the plasmon frequency, putting $\gamma=\alpha\gamma'$, we obtain

$$E=\hbar\omega_l\alpha^2\left[\frac{3}{2}\gamma'^2-\gamma'\sqrt{\frac{2}{\pi}}\left(\frac{F_1^2}{R_1}+\frac{F_2^2}{R_2}\right)\right].\tag{8}$$

It is possible to show that the renormalized frequencies $R_i$ and the quantities $Q_i=[(1-R_i^2)^2+\lambda^2(1-\eta)]^{1/2}$ satisfy the following relations, which are analogous to those found for the polariton problem:[7]

$$R_1^2+R_2^2=\lambda^2+1,$$

$$R_1^2R_2^2=\lambda^2\eta,$$

$$(1-R_1^2)(1-R_2^2)=-\lambda^2(1-\eta),$$

$$Q_1^2Q_2^2=\lambda^2(1-\eta)(R_1^2-R_2^2)^2,$$

$$\frac{Q_1}{Q_2}=\frac{1-R_1^2}{\lambda\sqrt{1-\eta}},$$

$$Q_1^2+Q_2^2=(R_1^2-R_2^2)^2,$$

$$Q_1^2R_2^2+Q_2^2R_1^2=(R_1^2-R_2^2)^2,$$

$$Q_1^2R_1^2+Q_2^2R_2^2=\lambda^2(R_1^2-R_2^2)^2. \tag{9}$$

As a consequence of the above relations it can be shown that for $\lambda\neq 0$

$$\frac{F_1^2}{R_1}+\frac{F_2^2}{R_2}=\frac{1}{1-\eta}. \tag{10}$$

This implies that the energy (8) does not depend on the electronic density. In fact the value $\gamma'=\frac{1}{3}(2/\pi)^{1/2}[1/(1-\eta)]$ minimizes the energy which becomes

$$E_{\rm self}^{s}=-\frac{1}{3\pi}\frac{1}{(1-\eta)^2}\hbar\omega_l\alpha^2. \tag{11}$$

It is important to remark that such a value is not coincident with that found in the strong coupling polaron theory, because it contains the extra factor $1/(1-\eta)^2$. This occurs because the vacuum state of phonon and plasmon is not coincident with that of the operators $\alpha_{\vec{k}}$ and $\beta_{\vec{k}}$. The strong coupling self-energy (11) is independent of the polarity of the crystal because it can be written in a more transparent way as

$$E_{\rm self}^{s}=-\frac{1}{3\pi}\frac{1}{2}\frac{e^2}{\epsilon_\infty a_0} \tag{12}$$

with $a_0=\hbar^2\epsilon_\infty/me^2$.

Since $E_{\rm self}^{in}=-\alpha f(\lambda,\eta)$ and $E_{\rm self}^{s}=-\alpha^2 g(\eta)$, the quantity $\alpha_m=f(\lambda,\eta)/g(\eta)$ is such that for $\alpha<\alpha_m$ it results $E_{\rm self}^{in}<E_{\rm self}^{s}$. In Fig. 1 is given $\alpha_m$ as a function of $\lambda$ for a fixed value of $\eta$. It is evident that, even for very large values of $\alpha$, for high $\lambda$, the intermediate coupling theory gives lower self-energies.

In the above discussion both fields were described in the intermediate or strong coupling limits. However, this approximation could be not appropriate. To be more confident with the above conclusions, the plasma-polaron problem has been studied with the Feynman[4] path-integral formalism.

The starting Hamiltonian is again given by (1). Using the path-integral representation and following Feynman's analysis for the polaron, we find that the two renormalized boson fields give rise to independent contributions written in terms of two dimensionless Fröhlich-like constants $\alpha_1=\alpha F_1^2$ and $\alpha_2=\alpha F_2^2$:

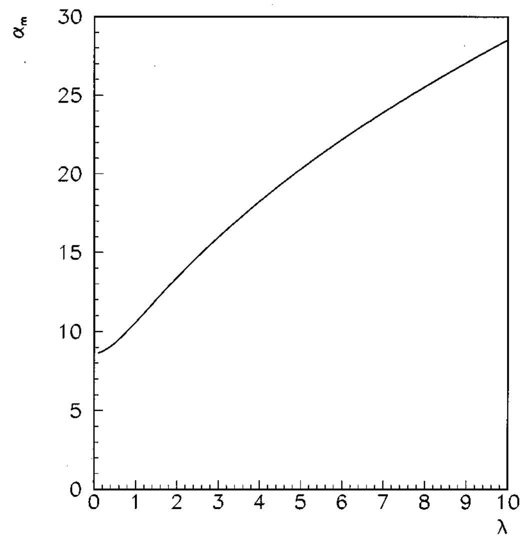


FIG. 1. $\alpha_m$ as a function of $\lambda$ for $\eta=0.05$. For values of $\alpha$ and $\lambda$ above the curve, the self-energy in the strong coupling limit is lower with respect to that in the intermediate regime; the opposite occurs in the region below the curve. The electronic density increases with $\lambda^2$ and $\lambda=1$ is an electronic density of $\sim 10^{19}$ cm$^{-3}$.

$$S_{PP}[\vec{r}(t)]=-\int_0^\beta dt\frac{m}{2}\dot{\vec{r}}^2+\sum_{i=1,2}\frac{\alpha_i}{\sqrt{2}}\int_0^\beta dt\int_0^\beta ds\frac{G_{\Omega_i}(t-s)}{|\vec{r}(t)-\vec{r}(s)|},$$

where $G_{\Omega_i}$ is the $i$th boson field Green function

$$G_\omega(u,\beta)=\frac{\cosh(\hbar\omega(\beta/2-u))}{2\sinh(\hbar\omega\beta/2)}.$$

The problem is solved variationally by considering a trial action $S_t$ in which the retarded potential $|\vec{r}(t)-\vec{r}(s)|^{-1}$ is replaced by a harmonic retarded interaction, i.e., the interaction of the electron with the boson fields is simulated through a quadratic interaction

$$S_t[\vec{r}(t)]=-\int_0^\beta dt\,\frac{m}{2}\,\dot{\vec{r}}^2-\sum_{i=1,2}\frac{c_i}{2}\int_0^\beta dt\int_0^\beta ds G_{w_i}(t-s)\times[\vec{r}(t)-\vec{r}(s)]^2$$

and $c_i$, $w_i$, for $i=1,2$, are four variational parameters.

A central quantity needed in the variational calculation is the correlation function of the electron

$$\langle e^{i\vec{k}[\vec{r}(t)-\vec{r}(s)]}\rangle=e^{-k^2D_{PP}(|t-s|)}.$$

Here $\langle\ \rangle$ denotes a path-integral average with weight $e^{S_t}$. We find the expression

$$D_{PP}(t)=\frac{1}{2}\left[\frac{v_1^2v_2^2}{\omega_1^2\omega_2^2}t\left(1-\frac{t}{\beta}\right)+2\frac{(\omega_1^2-v_1^2)(\omega_1^2-v_2^2)}{\omega_1^2(\omega_1^2-\omega_2^2)}E(\omega_1,\beta,t)+2\frac{(\omega_2^2-v_1^2)(v_2^2-\omega_2^2)}{\omega_2^2(\omega_1^2-\omega_2^2)}E(\omega_2,\beta,t)\right], \quad (13)$$

where $E(\omega,\beta,t)$ is an auxiliary function defined by

$$E(\omega,\beta,t)=\frac{\sinh(\omega t/2)\sinh(\omega(\beta-t)/2)}{\omega\sinh(\omega\beta/2)}.$$

The new modes $\omega_1$ and $\omega_2$ are functions of the variational parameters; they can be considered as independent variational parameters replacing $c_1$ and $c_2$. Since $c_1$ and $c_2$ must not be negative, then $\omega_1\geqslant\max(v_1,v_2)\geqslant\omega_2\geqslant\min(v_1,v_2)$. The function $D_{PP}$ is temperature dependent and it is the correlation function of the electron in the presence of two independent fields.

In order to calculate an upper bound for the plasma-polaron ground state energy, we take the limit of high $\beta$. The result is

$$E_{PP}=\frac{3}{2}\hbar(\omega_1+\omega_2-v_1-v_2)-\frac{3}{4}\hbar\left[\omega_1\frac{(\omega_1^2-v_1^2)(\omega_1^2-v_2^2)}{\omega_1^2(\omega_1^2-\omega_2^2)}+\omega_2\frac{(\omega_2^2-v_1^2)(v_2^2-\omega_2^2)}{\omega_2^2(\omega_1^2-\omega_2^2)}\right]-A \quad (14)$$

with

$$A=\sum_{i=1,2}\frac{\alpha_i}{\sqrt{2\pi}}\int_0^\infty dt D_\infty^{-1/2}(t)e^{-\hbar\Omega_i t}$$

and

$$D_\infty(t)=\lim_{\beta\to\infty}D_{PP}(t,\beta)=\frac{\hbar}{2m}\left[\frac{v_1^2v_2^2}{\omega_1^2\omega_2^2}t+\frac{(\omega_1^2-v_1^2)(\omega_1^2-v_2^2)}{\omega_1^2(\omega_1^2-\omega_2^2)}\frac{1-e^{-\hbar\omega_1 t}}{\omega_1}+\frac{(\omega_2^2-v_1^2)(v_2^2-\omega_2^2)}{\omega_2^2(\omega_1^2-\omega_2^2)}\frac{1-e^{-\hbar\omega_2 t}}{\omega_2}\right]. \quad (15)$$

The integrals that appear here cannot be performed in closed form so that a complete determination of $E_{PP}$ requires numerical integration.

Another quantity of interest is the effective mass, which can be found extending the Feynman polaron scheme. When $\beta$ approaches $\infty$ we find

$$m_{\text{eff}}=m\left(1+\frac{1}{3\sqrt{8\pi}}\sum_{i=1,2}\alpha_i\int_0^\infty dt t^2 D_\infty(t)^{-3/2}e^{-\hbar\Omega_i t}\right), \quad (16)$$

where the best parameters obtained for $E$ have to be inserted in $D_\infty$.

First of all we show how the Feynman polaron limit will occur in our treatment. When $\lambda\to 0$, then $\Omega_1\to\omega_l$, $\Omega_2\to 0$, $\alpha 1\to\alpha$, $\alpha 2\to 0$, and numerically we find that $\omega_2/v_2=1$. Consequently the Feynman results (self-energy and mass) for the polaron are retrieved.

In Figs. 2 and 3 we show the self-energy as a function of $\lambda$ for $\alpha=8$ and $\alpha=15$. The dotted, full, and dashed lines give the results of the calculations in the intermediate regime, in the strong limit, and in the path-integral formalism, respectively. We see that for $\alpha=8$ the strong coupling self-

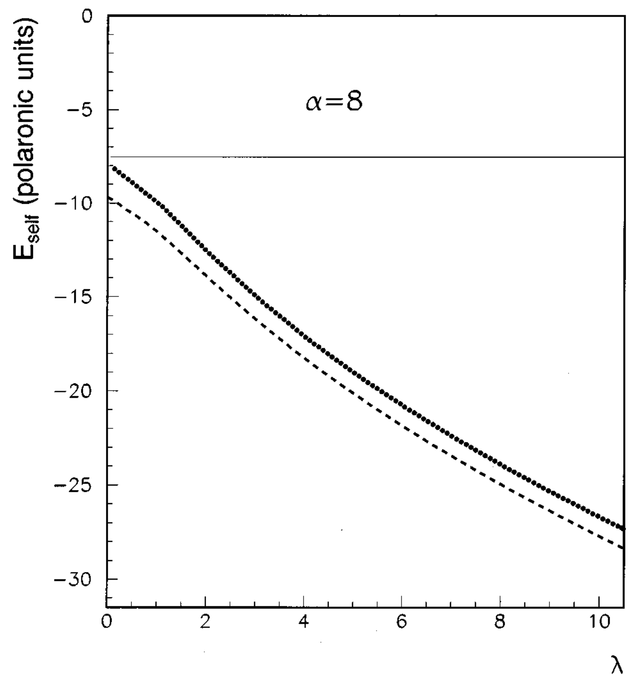


FIG. 2. The plasma-polaron self-energy as a function of the density for $\alpha=8$ and $\eta=0.05$ calculated with the intermediate regime theory (dotted line), with the strong coupling theory (full line), and with the path-integral technique (dashed line).

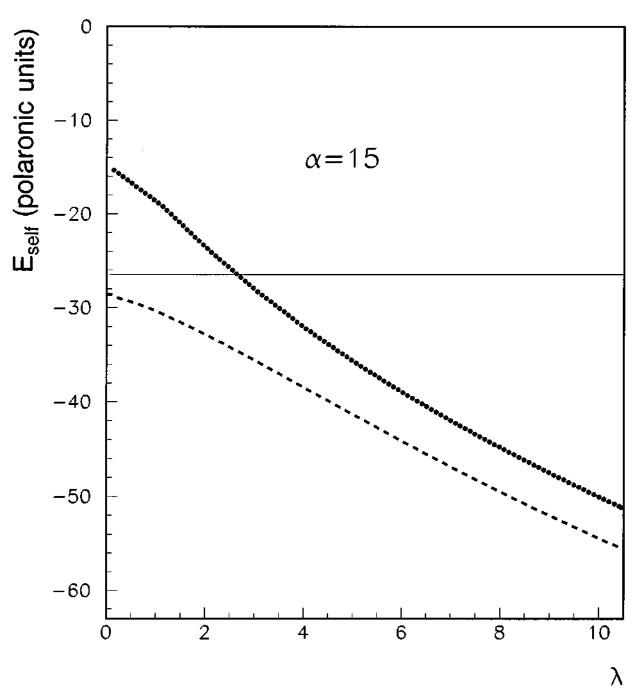


FIG. 3. The same quantities of Fig. 2 for $\alpha=15$ and $\eta=0.05$.

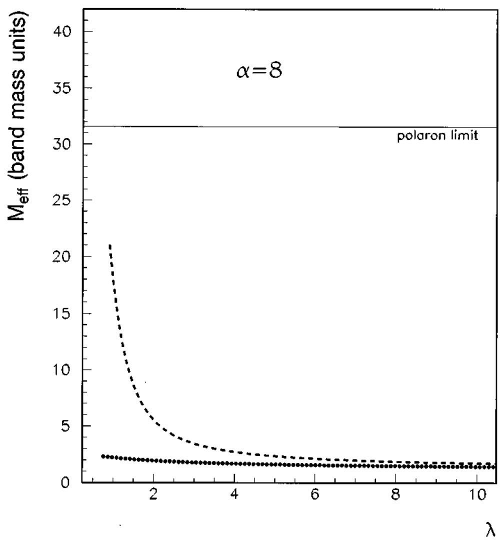


FIG. 4. The plasma-polaron mass as a function of the density for $\alpha=8$ and $\eta=0.05$ calculated with the intermediate regime theory (dotted line) and with the path-integral technique (dashed line). The divergent value of the mass for $\lambda\rightarrow 0$ in the intermediate regime is not indicated.

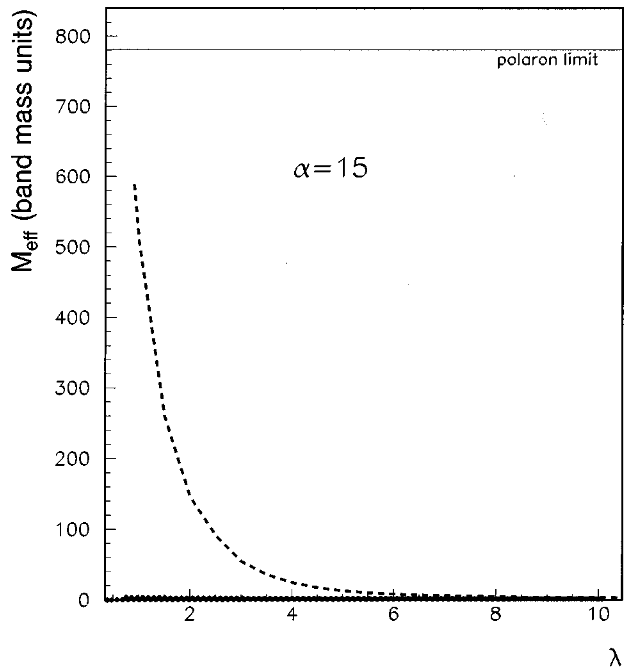


FIG. 5. The same quantities of Fig. 4 for $\alpha=15$ and $\eta=0.05$.

energy is always higher than the intermediate one, while the last is higher than the path-integral self-energy. Moreover, for any electron density, the path-integral values follow closely the intermediate ones. This indicates that the plasma polaron is not always self-trapped.

For $\alpha=15$ we find that the strong coupling self-energy is lower than the intermediate one for low electronic density and then a crossover occurs at $\lambda^*\sim 3$. For $\lambda<\lambda^*$ the plasma polaron is self-trapped and becomes untrapped for $\lambda>\lambda^*$. This different behavior comes out because the plasma oscillations screen the electron-phonon interaction. Finally the path-integral results are lower with respect to the other curves and interpolate between the intermediate and strong limit. The polaron self-trapped for low electronic density becomes not self-trapped in the opposite limit. This fact is also confirmed by studying the polaron mass changes increasing the electronic density. In Figs. 4 and 5 we show the plasma-polaron masses as a function of $\lambda$ again for $\alpha=8$ and $\alpha=15$. The full, dashed, and dotted lines give the masses in strong coupling, path-integral, and intermediate approximation, respectively. For $\alpha=8$ the intermediate and path-integral masses tend to be coincident for large $\lambda$, while they become different for small $\lambda$ ($\lambda\leqslant 4$). We note that for $\lambda\rightarrow 0$ the intermediate regime approximation is not appropriate because $m^*\rightarrow\infty$. This nonphysical limit is intrinsic to the intermediate approximation and cannot be eliminated even taking into account perturbatively the terms neglected in the Lee variational procedure.[8]

The $\alpha=15$ case (Fig. 5) shows a similar behavior, but now the increase of the mass in the path-integral calculation is greater and it appears at larger density.

We end the paper with a brief discussion on the possible relevance of our results for high $T_c$ superconductors. It is well known that increasing the doping level many high $T_c$ materials go from an isolating phase to a metallic one and then become superconducting. In the isolating phase there is experimental evidence, from far-infrared reflectivity measurements, of formation of self-trapped small polarons.[9] They seem to survive also in the metallic phase where the polaronic absorption is superimposed to a Drude term which, then, controls the reflectivity. In our opinion, this experimental evidence suggests that electron-phonon interaction in these materials is relevant and then both self-trapped small polarons and not self-trapped electrons (holes) are present. In particular we focus our attention on the Drude contribution to the reflectivity and try to explain it in terms of mobile large (not self-trapped) polarons. In this context the large polarons discussed in this paper are better candidates since they are characterized by lower effective masses. Then the result that even for very high electron-phonon coupling $\alpha$, increasing the carrier density $n$ ($\lambda$) causes the large polarons to become mobile particles is, in our opinion, relevant for high $T_c$ materials. On the other hand, we note that in a broad range of values of $\alpha$, charge carrier mass $m$ and longitudinal optical phonon energy $\omega_l$, $n^*$ ($\lambda^*$) results lower than typical values of charge carrier density in high $T_c$ superconductors ($n\sim 10^{20}$–$10^{21}$).

We also mention that a charge carrier mass decreasing with the density, as found in this paper, finds support in recent mass estimation[10] based on the Hall effect and magnetic penetration depth measurements in YBCO samples.

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