

## Phonon-plasmon cooperative effects in the dilute large-bipolaron gas: A possible mechanism for high- $T_c$ superconductivity

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A Hamiltonian for two interacting electrons coupled with longitudinal phonons and plasmons is set up. The phonons are treated in the Fröhlich scheme and the plasmons in the single-pole approximation. We are interested in systems with low electron density ( $n \leq 10^{20} - 10^{21}$ ) such as high- $T_c$  superconductors. It is shown that, in this density range, the electron dynamics do not simply screen the electrostatic interaction but, also, cooperates to the bipolaron formation. Furthermore we find that the binding energies and the effective mass depend on the electronic density. The features of the effective electron-electron potential are discussed, mainly for what concerns the self-energy terms and the long-range tail; the former depends on the pair state and the second shows that the plasmon field tends to screen all the electrostatic interactions, even the electron-phonon one. It is also shown that the Hamiltonian formulation is equivalent to a dielectric formulation where the total dielectric function of the system is the sum of the dielectric function appropriate for an ionic material and that appropriate for the electron gas. Within this model we calculate the Bose-condensation critical temperature of a system of correlated pairs and free carriers in a two-fluid model at thermodynamical equilibrium.

### I. INTRODUCTION

For many years the BCS theory<sup>1</sup> for phonon-mediated pairing has represented the theoretical framework for superconductivity. Only recently, with the discovery of high- $T_c$  superconductors have BCS theory and the mechanism responsible for the pairing been questioned.

In the last years many alternative theories have been proposed such as the resonating-valence-bond theory,<sup>2</sup> the exciton coupling,<sup>3</sup> the spin fluctuation models,<sup>4</sup> and bipolaron model.<sup>5,6</sup> It has also emphasized the specific roles of holes<sup>7</sup> and dimensionality.<sup>8</sup> Recently, there have been attempts to review Eliashberg theory<sup>9</sup> for strong electron-phonon couplings going beyond Migdal-theorem limitations.<sup>10,11</sup>

One of the most remarkable characteristics of the high- $T_c$  materials is a small coherence length of the order of a few angstroms. For this reason the Bose condensation of charged bosons,<sup>12</sup> which had been proposed even before BCS theory, has attracted much attention. In this approach many properties of the superconductivity can also be recovered.<sup>12-14</sup>

Important developments of the theory were obtained when it was shown that, considering a many-body system with an effective electron-electron attractive interaction, it is possible to go from a Bose condensation (bipolaron mechanism) to a BCS theory changing only the dilution parameter of the system ( $nR_b^3 \ll 1$  and  $nR_b^3 \gg 1$ , respec-

tively, where  $n$  is the electronic density and  $R_b$  the bipolaron radius).<sup>15</sup> Moreover, it was also shown that, in two dimensions, the existence of a bipolaron bound state makes the many-particle system unstable versus a superconductivity state; furthermore, if the binding energy is large with respect to the Fermi energy, we have a Bose condensation and in the opposite limit a BCS state.<sup>16</sup> The conclusion is that the bipolaron mechanism and BCS theory are complementary treatments of the same phenomenon.

In this scenario it is clear that a crucial problem is to understand whether or not two electrons can form a bound state through interactions with the phonon field.<sup>17-23</sup> In a previous paper<sup>24</sup> we presented results concerning the binding energy of two electrons interacting with a phonon and plasmon field. The idea was that, at low electron densities, the conduction electrons participate in the binding energy of the bipolaron not only by the screening of the electrostatic interactions, but also by a cooperative effect.<sup>25,26</sup>

In this work we extend this theory in such a way to calculate both binding energy and effective mass. We discuss the features of the effective electron-electron interaction and show the equivalence of the approach to a dielectric formulation. The electron-phonon interaction is treated in the Fröhlich approximation,<sup>27</sup> and the sea of other electrons is studied in the single-pole approximation.<sup>28</sup> We find that the problem reduces to that of two

electrons interacting with the phonon and plasmon boson fields and repelling each other with a Coulomb force; moreover, a phonon-plasmon interaction is introduced.<sup>29</sup> Since the part of the Hamiltonian referring to the boson fields is exactly diagonalized, we reduce the problem to that of two charges interacting with two independent renormalized fields. The binding energy and effective mass of the bi-plasma-polaron (BPP), i.e., the bound or metastable state formed by two electrons in interaction with the renormalized boson fields, are calculated with a self-consistent variational procedure. The main result is that these quantities depend on the electronic density (the dimensionless parameter which controls different density is  $\lambda = \omega_p / \sqrt{\epsilon_\infty} \omega_l$ , where  $\omega_p$ ,  $\omega_l$ , and  $\epsilon_\infty$  are the plasma frequency, the longitudinal optical-phonon frequency, and the high-frequency dielectric constant, respectively) and that in the regime of low density ( $\lambda \leq 1$ ) the phonon and plasmon give cooperative effects to make more bound the BPP, whereas in the opposite regime all the interactions are completely screened by the electrons and the BPP cannot form even in a metastable state. The effective electron-electron interaction can be also calculated; the self-energy terms show explicitly the dynamical and static screening effects played by the electronic density.<sup>30,31</sup> Finally, if the BPP radius becomes large, the total energy and the effective mass of the system reduce to those of two free plasma polarons (PP's), i.e., a particle in interaction with the two renormalized boson fields.

We show also that the results regarding the interaction of the external charges with the renormalized fields can be also obtained through a dielectric formulation of the problem in which the total dielectric function contains the sum of the contributions of the ionic part and the electronic one. The two formulations have different advantages, so that the effective electron-electron interaction is easy to calculate in the Hamiltonian formulation, whereas any improvement in the knowledge of the dielectric function allows us to know more realistic renormalized frequencies and consequently more reliable coupling of the external charges with the boson fields.

It can be also shown that the random-phase approximation for an electronic interaction given by the sum of the bare electron-electron and electron-phonon interaction<sup>32</sup> gives the total dielectric function considered above; consequently, our approach is not as poor as it would appear.

In all our results it appears that the corrections to the PP and BPP masses with respect to the band mass are very large. The total momentum, i.e., the sum of the center of the mass momentum of the pair and that of the phonons and plasmons, is conserved, whereas in the BCS (Ref. 1) and Eliashberg treatments<sup>9</sup> the pair momentum is zero. The consequence is that, in our case, the pairing, even in the case of zero total momentum, does not occur between particles with exactly opposite momenta.

All our calculations are done for  $\lambda \leq 2$ , so that the condition  $nR_b^3 \leq 1$  is verified, and therefore it is meaningful to consider only one pair of electrons in interaction with the others.

Finally, we calculate the critical temperature  $T_c$  for the Bose condensation of the BPP gas at thermodynamical

equilibrium with the PP one, using a two-fluid model.<sup>33</sup> Since our calculations are physically reliable for  $\lambda \leq 2$ , the highest density considered is  $n \sim 10^{20} \text{ cm}^{-3}$ . We find that  $T_c$  increases with  $\lambda$ , and it is about 50 K for  $\lambda = 2$ .

We believe that the described effects could be relevant for the high- $T_c$  superconductors, because their electronic density is small, the correlation length is comparable with the polaron radius, and their high-frequency dielectric constant is small with respect to the static one. A reasonable estimate of the electron-phonon coupling constant  $\alpha$  for the high- $T_c$  materials gives  $\alpha \sim 8$ . Such a value is near the upper (lower) limit for which the intermediate-(strong-) coupling polaronic theory applies. This means that a complete theory of the electron-phonon coupling in high- $T_c$  superconductors should take into account both features. This work is complementary to those in which the strong-coupling electron-phonon scheme is used.

In Secs. II and III the model and its equivalence with the dielectric formulation are discussed. In Sec. IV the variational self-consistent procedure for the calculation of the ground-state energy and of the effective mass is presented. In Sec. V the features of the effective electron-electron potential are shown. Finally, in Sec. VI the numerical results are discussed and in Sec. VII the critical temperature is calculated.

## II. THE MODEL

In this section we set up a Hamiltonian describing two external electrons or holes (without loss of generality, in the following we will assume that the charge carriers are electrons) interacting with each other through a Coulomb repulsion and with both longitudinal optical phonons and plasma oscillations. The physics we have in mind is the following. By singling out of an electron gas two electrons interacting through long-range plasma excitations, we reduce the many-electron problem to a two-electron problem where the action of the whole electron gas is taken into account in terms of plasmon exchange between the singled out electrons. In doing this we are dealing with a sort of mean-field approximation in which the long-range part of the electron-electron interaction is correctly represented. We have to stress that in this paper the exchange effects are not considered.

Our approach is based on a set of motion equations which describe the dynamics of the optical phonons in the Fröhlich scheme and that of the electrons in the plasmon pole approximation. As far as the phonons are concerned, we introduce the classical quantity  $\mathbf{W}(\mathbf{r}, t)$ , which represents the relative ionic displacement in the elementary cells of the lattice. Within the Fröhlich scheme the classical motion equation for the spatial Fourier transform  $\mathbf{W}_\mathbf{k}(t)$  is given by

$$\frac{d^2}{dt^2} \mathbf{W}_\mathbf{k}(t) = b_{11} \mathbf{W}_\mathbf{k}(t) + b_{12} \mathbf{E}_\mathbf{k}, \quad (1)$$

where  $b_{11}$  and  $b_{12}$  are constants to be fixed and  $\mathbf{E}_\mathbf{k}$  is the spatial Fourier transform of the total electric field. We also introduce the ionic polarization  $\mathbf{P}(\mathbf{r}, t)$ , whose spatial Fourier transform  $\mathbf{P}_\mathbf{k}(t)$  is related to the ionic displacement

ment and to the total electric field by the constitutive relation

$$\mathbf{P}_k(t) = b_{12} \mathbf{W}_k(t) + b_{22} \mathbf{E}_k, \quad (2)$$

where again the constants  $b_{12}$  and  $b_{22}$  are to be fixed.

As far as the plasmons are concerned, the effect on the dynamics of the two external electrons will be taken into account only through the electron-density fluctuation  $\rho(\mathbf{r}, t)$ , which characterizes the collective excitations of the charged gas. In the plasmon pole approximation or, equivalently, in the hydrodynamic approximation, the spatial Fourier transform of the density fluctuation satisfies the equation

$$\frac{d^2 \rho_k}{dt^2} = -\frac{\omega_p^2}{4\pi} i\mathbf{k} \cdot \mathbf{E}_k - \frac{n}{m} (\omega_k^2 - \omega_p^2) \rho_k, \quad (3)$$

where  $\omega_p^2 = (4\pi n e^2)/m$ ,  $\omega_k$  is the  $k$ -dependent plasmon frequency,  $n$  is the average electron density, and  $m$  and  $-e$  are the band mass and the electronic charge, respectively. The coupling between the ionic motion and the electron-density fluctuations is given by the total electric field  $\mathbf{E}_k$ . This field must satisfy the equations

$$i\mathbf{k} \cdot \mathbf{E}_k = 4\pi [\rho_k - i\mathbf{k} \cdot \mathbf{P}_k - e \exp(-i\mathbf{k} \cdot \mathbf{r})], \quad (4)$$

$$\mathbf{k} \times \mathbf{E}_k = 0. \quad (5)$$

Equation (4) is the Gauss law, in which the total electron density is given by the sum of the electron-density fluctuation, the charge density arising from the ionic polarization, and the external electron localized at  $\mathbf{r}$ . Equation (5) assumes that the total electric field has components only along the wave-vector direction. This assumption fixes  $b_{11} = -\omega_i^2$ , where  $\omega_i$  is the frequency of the transverse optical phonon. Moreover, if the driving field has a frequency larger than those of the phonons, the ions cannot follow the variation of the electric field, so that  $\mathbf{w}_k = 0$  and consequently  $b_{22} = (\epsilon_\infty - 1)/(4\pi)$ , where  $\epsilon_\infty$  is the background high-frequency dielectric constant.

From Eqs. (2), (4), and (5), it is possible to calculate  $\mathbf{k} \cdot \mathbf{P}_k$  and  $\mathbf{k} \cdot \mathbf{E}_k$  as a function of  $\mathbf{k} \cdot \mathbf{w}_k$  and  $\rho_k$ . By substitution in Eqs. (1) and (3), we obtain for the quantities  $u_k = \mathbf{k} \cdot \mathbf{w}_k/k$  and  $z_k = \rho_k/(ik)$  the equations

$$\frac{d^2 u_k}{dt^2} = -\omega_i^2 u_k + \frac{4\pi b_{12}}{\epsilon_\infty} z_k - \frac{4\pi b_{12} e}{ik \epsilon_\infty} \exp(-i\mathbf{k} \cdot \mathbf{r}), \quad (6)$$

$$\frac{d^2 z_k}{dt^2} = -\frac{\omega_k^2}{\epsilon_\infty} z_k + \frac{\omega_p^2 b_{12}}{\epsilon_\infty} u_k - \frac{\omega_p^2 e}{ik \epsilon_\infty} \exp(-i\mathbf{k} \cdot \mathbf{r}), \quad (7)$$

where  $\omega_l$  is the longitudinal optical frequency and  $b_{12} = [(\epsilon_\infty/4\pi)(\omega_l^2 - \omega_i^2)]^{1/2}$ . The classical Hamiltonian which gives the motion equations (6) and (7) and the equation of motion of the electron can be quantized. The result is

$$H_0 = \sum_{\mathbf{k}} \left[ \hbar \omega_l (a_{\mathbf{k}}^\dagger a_{\mathbf{k}} + \frac{1}{2}) + \frac{\hbar \omega_k}{\sqrt{\epsilon_\infty}} (b_{\mathbf{k}}^\dagger b_{\mathbf{k}} + \frac{1}{2}) + Z_k (a_{\mathbf{k}}^\dagger + a_{-\mathbf{k}}) (b_{-\mathbf{k}}^\dagger - b_{\mathbf{k}}) + (V_k e^{i\mathbf{k} \cdot \mathbf{r}} a_{\mathbf{k}} + \text{H.c.}) + (U_k e^{i\mathbf{k} \cdot \mathbf{r}} b_{\mathbf{k}} + \text{H.c.}) \right] + \frac{p^2}{2m}, \quad (8)$$

where

$$Z_k = -\frac{i\hbar}{2} \left[ \frac{\omega_p^2 \omega_l}{\omega_k \sqrt{\epsilon_\infty}} (1 - \eta) \right]^{1/2},$$

$$V_k = -\frac{1}{k} \left[ \frac{2\pi e^2 \omega_l \hbar}{V \epsilon_\infty} (1 - \eta) \right]^{1/2},$$

$$U_k = -\frac{i}{k} \left[ \frac{2\pi e^2 \omega_p^2 \hbar}{V \omega_k \epsilon_\infty^{3/2}} \right]^{1/2},$$

$$\eta = \frac{\epsilon_\infty}{\epsilon_0},$$

and where  $V$  is the volume. The operators  $a_{\mathbf{k}} (a_{\mathbf{k}}^\dagger)$  and  $b_{\mathbf{k}} (b_{\mathbf{k}}^\dagger)$  are the annihilation (creation) boson operators for phonons and plasmons, respectively.

The term containing  $Z_k$  gives the interaction between phonons and plasmons;<sup>29</sup> the term whose coefficient is  $V_k$  gives the Fröhlich<sup>27</sup> electron-phonon interaction, and the term whose coefficient is  $U_k$  gives the electron-plasmon interaction.<sup>34</sup> Summarizing, Eq. (8) gives the Hamiltonian of an electron, a phonon, and plasmon fields in interaction.

If we have two electrons interacting with the two boson fields and repelling each other through the Coulomb force, the Hamiltonian (8) becomes

$$H = \sum_{\mathbf{k}} \left[ \hbar \omega_l (a_{\mathbf{k}}^\dagger a_{\mathbf{k}} + \frac{1}{2}) + \frac{\hbar \omega_k}{\sqrt{\epsilon_\infty}} (b_{\mathbf{k}}^\dagger b_{\mathbf{k}} + \frac{1}{2}) + Z_k (a_{\mathbf{k}}^\dagger + a_{-\mathbf{k}}) (b_{-\mathbf{k}}^\dagger - b_{\mathbf{k}}) + [V_k (e^{i\mathbf{k} \cdot \mathbf{r}_1} + e^{i\mathbf{k} \cdot \mathbf{r}_2}) a_{\mathbf{k}} + \text{H.c.}] + [U_k (e^{i\mathbf{k} \cdot \mathbf{r}_1} + e^{i\mathbf{k} \cdot \mathbf{r}_2}) b_{\mathbf{k}} + \text{H.c.}] \right] + \frac{p_1^2}{2m} + \frac{p_2^2}{2m} + \frac{e^2}{\epsilon_\infty |\mathbf{r}_1 - \mathbf{r}_2|}. \quad (9)$$

The Coulomb repulsion between the two external electrons is statically screened by the background high-frequency dielectric constant  $\epsilon_\infty$ . Introducing the coordinates  $\mathbf{R}$  of the center of mass of the pair, the relative position  $\mathbf{r} = \mathbf{r}_1 - \mathbf{r}_2$ , and the conjugate variables  $\mathbf{P}$  and  $\mathbf{p}$ , the Hamiltonian becomes

$$H = \sum_{\mathbf{k}} \left[ \hbar\omega_l(a_{\mathbf{k}}^\dagger a_{\mathbf{k}+\frac{1}{2}}) + \hbar \frac{\omega_k}{\sqrt{\epsilon_\infty}} (b_{\mathbf{k}}^\dagger b_{\mathbf{k}+\frac{1}{2}}) + Z_k(a_{\mathbf{k}}^\dagger + a_{-\mathbf{k}})(b_{-\mathbf{k}}^\dagger - b_{\mathbf{k}}) \right. \\ \left. + [\rho_{\mathbf{k}}(\mathbf{r})e^{i\mathbf{k}\cdot\mathbf{R}}(V_k a_{\mathbf{k}} + U_k b_{\mathbf{k}}) + \text{H.c.}] \right] + \frac{P^2}{2M} + \frac{p^2}{2\mu} + \frac{e^2}{\epsilon_\infty r}, \quad (10)$$

with  $M = 2m$ ,  $\mu = m/2$ , and  $\rho_{\mathbf{k}}(\mathbf{r}) = 2 \cos[(\mathbf{k}\cdot\mathbf{r})/2]$ .

Hamiltonian (10) commutes with

$$\mathcal{P} = \mathbf{P} + \hbar \sum_{\mathbf{k}} \mathbf{k} (a_{\mathbf{k}}^\dagger a_{\mathbf{k}} + b_{\mathbf{k}}^\dagger b_{\mathbf{k}}), \quad (11)$$

which is the total momentum of the system. Our goal is to construct the ground state of Eq. (10), taking into account the conservation law (11). We consider first the Hamiltonian

$$H_0 = \sum_{\mathbf{k}} \left[ \hbar\omega_l(a_{\mathbf{k}}^\dagger a_{\mathbf{k}+\frac{1}{2}}) + \frac{\hbar\omega_k}{\sqrt{\epsilon_\infty}} (b_{\mathbf{k}}^\dagger b_{\mathbf{k}+\frac{1}{2}}) \right. \\ \left. + Z_k(a_{\mathbf{k}}^\dagger + a_{-\mathbf{k}})(b_{-\mathbf{k}}^\dagger - b_{\mathbf{k}}) \right] \quad (12)$$

and note that its structure is similar to that considered by Hopfield<sup>35</sup> for the polariton problem. It can be shown that the constants  $w_{1k}$ ,  $x_{1k}$ ,  $y_{1k}$ ,  $z_{1k}$ ,  $w_{2k}$ ,  $x_{2k}$ ,  $y_{2k}$ , and  $z_{2k}$  can be fixed in such a way that the canonical transformations

$$\begin{aligned} \alpha_{\mathbf{k}_1} &= w_{1k} a_{\mathbf{k}} + x_{1k} b_{\mathbf{k}} + y_{1k} a_{-\mathbf{k}}^\dagger + z_{1k} b_{-\mathbf{k}}^\dagger, \\ \alpha_{\mathbf{k}_2} &= w_{2k} a_{\mathbf{k}} + x_{2k} b_{\mathbf{k}} + y_{2k} a_{-\mathbf{k}}^\dagger + z_{2k} b_{-\mathbf{k}}^\dagger, \\ \alpha_{-\mathbf{k}_1}^\dagger &= y_{1k}^* a_{\mathbf{k}} + z_{1k}^* b_{\mathbf{k}} + w_{1k}^* a_{-\mathbf{k}}^\dagger + x_{1k}^* b_{-\mathbf{k}}^\dagger, \\ \alpha_{-\mathbf{k}_2}^\dagger &= y_{2k}^* a_{\mathbf{k}} + z_{2k}^* b_{\mathbf{k}} + w_{2k}^* a_{-\mathbf{k}}^\dagger + x_{2k}^* b_{-\mathbf{k}}^\dagger \end{aligned} \quad (13)$$

satisfy the relations

$$[\alpha_{\mathbf{k}_i}, \alpha_{\mathbf{q}_j}^\dagger] = \delta_{ij} \delta_{\mathbf{k}\mathbf{q}}, \quad [\alpha_{\mathbf{k}_i}, \alpha_{\mathbf{q}_j}] = 0, \quad (14)$$

$$[\alpha_{\mathbf{k}_i}, H_0] = \hbar\Omega_i(\mathbf{k})\alpha_{\mathbf{k}_i}, \quad [\alpha_{\mathbf{k}_i}^\dagger, H_0] = -\hbar\Omega_i(\mathbf{k})\alpha_{\mathbf{k}_i}^\dagger, \quad (15)$$

where  $\Omega_i$  are the new frequencies to calculate. The above problem has been solved;<sup>32</sup> defining  $\tilde{\omega}_k = \omega_k/\sqrt{\epsilon_\infty}$ , we obtain, where  $i=1,2$ ,

$$2\Omega_i^2(k) = \tilde{\omega}_k^2 + \omega_l^2 \\ + (-1)^{i+1} \sqrt{(\tilde{\omega}_k^2 - \omega_l^2)^2 + 16\tilde{\omega}_k\omega_l|Z_k/\hbar|^2}, \quad (16)$$

$$x_{ik} = \frac{(\tilde{\omega}_k + \Omega_i)(\omega_l^2 - \Omega_i^2)}{2T_{ki}\sqrt{\tilde{\omega}_k\Omega_i}}, \quad (17)$$

$$y_{ik} = \frac{Z_k\tilde{\omega}_k(\omega_l - \Omega_i)}{\hbar T_{ki}\sqrt{\tilde{\omega}_k\Omega_i}}, \quad (18)$$

$$w_{ik} = -\frac{Z_k\tilde{\omega}_k(\omega_l + \Omega_i)}{\hbar T_{ki}\sqrt{\tilde{\omega}_k\Omega_i}}, \quad (19)$$

$$z_{ik} = \frac{(\tilde{\omega}_k - \Omega_i)(\omega_l^2 - \Omega_i^2)}{2T_{ki}\sqrt{\tilde{\omega}_k\Omega_i}}, \quad (20)$$

$$T_{ki} = \left[ 4 \left| \frac{Z_k}{\hbar} \right|^{1/2} \tilde{\omega}_k\omega_l + (\omega_l^2 - \Omega_i^2)^2 \right]^{1/2}. \quad (21)$$

The above canonical transformation applied to the Hamiltonian (9) gives

$$H = \sum_{\mathbf{k}} \{ \hbar\Omega_1(\alpha_{\mathbf{k}_1}^\dagger \alpha_{\mathbf{k}_1} + \frac{1}{2}) + \hbar\Omega_2(\alpha_{\mathbf{k}_2}^\dagger \alpha_{\mathbf{k}_2} + \frac{1}{2}) \\ + [\rho_{\mathbf{k}}(\mathbf{r})e^{i\mathbf{k}\cdot\mathbf{R}}(\tilde{V}_k \alpha_{\mathbf{k}_1} + \tilde{U}_k \alpha_{\mathbf{k}_2}) + \text{H.c.}] \} \\ + \frac{P^2}{2M} + \frac{p^2}{2\mu} + \frac{e^2}{\epsilon_\infty r}, \quad (22)$$

where

$$\tilde{V}_k = V_k(w_{1k}^* - y_{1k}^*) + U_k(x_{1k}^* + z_{1k}^*) \quad (23)$$

and

$$\tilde{U}_k = V_k(w_{2k}^* - y_{2k}^*) + U_k(x_{2k}^* + z_{2k}^*). \quad (24)$$

In the transformed Hamiltonian (22) the problem is reduced to that of two particles interacting through a Coulomb field and with two independent renormalized boson fields. The coupling of the electrons with the new fields is through  $\tilde{V}_k$  and  $\tilde{U}_k$ , which are functions of the two old coupling constants  $V_k$  and  $U_k$ . The features of the new frequencies  $\Omega_i(k)$  and of  $\tilde{V}_k$  and  $\tilde{U}_k$  can be studied introducing the dimensionless parameters  $\lambda = \omega_p/\sqrt{\epsilon_\infty\omega_l}$ ,  $s_k = \omega_k/\omega_p$ , and  $\eta = \epsilon_\infty/\epsilon_0$ . We obtain for  $\lambda \rightarrow 0$ , i.e., for small electronic density,

$$\Omega_1(k) \rightarrow \omega_l [1 + \lambda^2(1-\eta)]^{1/2}, \quad (25)$$

$$\Omega_2(k) \rightarrow \omega_l \lambda (s_k^2 - 1 + \eta)^{1/2}, \quad (26)$$

$$\tilde{V}_k \rightarrow iV_k, \quad (27)$$

$$\tilde{U}_k \rightarrow \eta U_k \frac{s_k^{1/2}}{(s_k^2 - 1 + \eta)^{1/4}}. \quad (28)$$

It can be seen from these equations that  $\Omega_1$  goes to the phonon longitudinal frequency and the corresponding coupling reduces to the Fröhlich term, while  $\Omega_2$  and  $\tilde{U}_k$  go to limits whose meaning is clear when the plasma frequency is not dispersive ( $\Omega_2 \rightarrow \omega_p/\sqrt{\epsilon_0}$ ,  $\tilde{U}_k \rightarrow U_k \eta^{3/4}$ ).

In the opposite limit  $\lambda \gg 1$ , we have

$$\Omega_1 \rightarrow \omega_l \left[ \lambda^2 s_k^2 + \frac{1-\eta}{s_k^2} \right]^{1/2}, \quad (29)$$

$$\Omega_2 \rightarrow \omega_l \left[ 1 - \frac{1-\eta}{s_k^2} \right]^{1/2}, \quad (30)$$

$$\tilde{V}_k \mapsto U_k, \quad (31)$$

$$\tilde{U}_k \mapsto -i \frac{V_k}{[1 - (1 - \eta)/s_k^2]^{1/4}} \left[ 1 - \frac{1}{s_k^2} \right]. \quad (32)$$

From Eqs. (29) and (31) it is seen that  $\Omega_1$  goes to  $\omega_k$  and  $\tilde{V}_k$  to the electron-plasmon interaction;  $\Omega_2$  and  $\tilde{U}_k$  go to complicated limits whose physical meaning can be extracted setting  $s_k = 1$ . In this case,  $\Omega_2 \rightarrow \omega_l$  and  $\tilde{U}_k \rightarrow 0$ , indicating a complete screening.

### III. DIELECTRIC FORMULATION

An alternative and fruitful approach to the Hamiltonian discussed above makes use of the knowledge of a dielectric function appropriate to the specific system under investigation. The line of reasoning is the following. First, let us consider the model Hamiltonian

$$H = \sum_{\mathbf{k}, n} \omega_{n, \mathbf{k}} b_{\mathbf{k}, n}^\dagger b_{\mathbf{k}, n} + \sum_{\mathbf{k}, n} C_{n, \mathbf{k}} (b_{\mathbf{k}, n} e^{i\mathbf{k} \cdot \mathbf{r}(t)} + \text{H.c.}), \quad (33)$$

where  $C_{n, \mathbf{k}}$  and  $\omega_{n, \mathbf{k}}$  are parameters to be determined, and assume that the external charge is moving along an arbitrary path  $\mathbf{r}(t)$ . The Fermi golden rule leads immediately to the energy transfer  $\Delta E$  from the external charge to the boson field,<sup>36</sup>

$$\Delta E = \frac{2\pi}{\hbar} \int_0^\infty d\omega \frac{V}{(2\pi)^3} \int d^3k \sum_n \omega |C_{n, \mathbf{k}}|^2 |\rho_{\text{ex}}(\mathbf{k}, \omega)|^2 \times \delta(\omega_{n, \mathbf{k}} - \omega), \quad (34)$$

where

$$\rho_{\text{ex}}(\mathbf{k}, \omega) = \int_{-\infty}^\infty dt e^{i[\mathbf{k} \cdot \mathbf{r}(t) - \omega t]}.$$

Let us now calculate the same quantity  $\Delta E$  starting from the Maxwell equations and considering a medium characterized by a dielectric function  $\epsilon(k, \omega)$ . We have<sup>36</sup>

$$\Delta E = 8\pi e^2 \int \frac{d^3k}{(2\pi)^3} \int_0^\infty \frac{d\omega}{2\pi} \frac{\omega}{k^2} |\rho_{\text{ex}}(\mathbf{k}, \omega)|^2 \times \text{Im} \left[ -\frac{1}{\epsilon(k, \omega)} \right]. \quad (35)$$

Comparing Eqs. (34) and (35), we obtain

$$\sum_n |C_{n, \mathbf{k}}|^2 \delta(\omega_{n, \mathbf{k}} - \omega) = \frac{4e^2 \hbar}{Vk^2} \text{Im} \left[ -\frac{1}{\epsilon(k, \omega)} \right]. \quad (36)$$

Note that  $\epsilon(k, \omega)$  does not include the effects of the external particle.

As an example of the utility of (36), let us consider the case of an electron in a polar material. If the material is described by the approximate dielectric function<sup>32</sup>

$$\epsilon(k, \omega) = \epsilon_\infty \left[ 1 - \frac{\omega_l^2 - \omega_t^2}{\omega^2 - \omega_t^2} \right], \quad (37)$$

it is easy to show that

$$\text{Im} \left[ -\frac{1}{\epsilon(k, \omega)} \right] = \frac{\pi}{2} \frac{\omega_l^2 - \omega_t^2}{\epsilon_\infty \omega} \delta(\omega - \omega_l).$$

By using Eq. (36) and noting that our model dielectric constant has only one zero at  $\omega = \omega_l$ , we can recover the usual electron-phonon Fröhlich coupling constant.<sup>27</sup> The same scheme can be used to derive the electron-plasmon coupling constant within the plasmon-pole approximation for the electron-gas dielectric function.

The case we are interested in is the interaction of an external particle with a bath of phonons and plasmons. A widely used approximation for the dielectric function<sup>32</sup> is

$$\epsilon(k, \omega) = \epsilon_\infty \left[ 1 - \frac{\omega_l^2 - \omega_t^2}{\omega^2 - \omega_t^2} - \frac{\tilde{\omega}_p^2}{\omega^2 + \tilde{\omega}_p^2 - \omega^2(k)} \right], \quad (38)$$

where  $\omega(k)$  is the plasmon dispersion and  $\tilde{\omega}_p = \omega_p / \sqrt{\epsilon_\infty}$ . The zeros of  $\epsilon(k, \omega)$  give the eigenfrequencies  $\Omega_i$  of the coupled phonon-plasmon system, which are the same as those found in Eq. (16). It is convenient to rewrite (38) in the form

$$\frac{1}{\epsilon(k, \omega)} = \frac{1}{\epsilon_\infty} \left[ 1 + \frac{R_1(k)}{\omega^2 - \Omega_1^2(k)} + \frac{R_2(k)}{\omega^2 - \Omega_2^2(k)} \right], \quad (39)$$

where

$$R_i(k) = (-1)^i \frac{\Omega_i^2(\tilde{\omega}_p^2 + \omega_l^2 - \omega_t^2) - \Omega_1^2 \Omega_2^2 - \omega_t^2(\tilde{\omega}_p^2 - \omega_k^2)}{\Omega_2^2 - \Omega_1^2}, \quad i = 1, 2, \quad (40)$$

so that it is immediate to have

$$\text{Im} \left[ -\frac{1}{\epsilon(k, \omega)} \right] = \frac{\pi}{2\omega\epsilon_\infty} [R_1(k)\delta(\omega - \Omega_1(k)) + R_2(k)\delta(\omega - \Omega_2(k))]. \quad (41)$$

From (36) the coupling coefficients are

$$|C_{i, \mathbf{k}}|^2 = \frac{2\pi e^2 \hbar}{Vk^2 \epsilon_\infty} \frac{R_i(k)}{\Omega_i(k)}. \quad (42)$$

It is only a matter of lengthy algebra to show that these coupling coefficients are identical with those derived in the previous section. The reason for this identity lies on the internal consistency between the model dielectric function of Eq. (38) and the equations of motion (1) and (3).

The equivalence shown gives the possibility, using improved total dielectric functions, to know more realistic coupling coefficients between the external charge and the renormalized fields. In particular, the knowledge of the dielectric function for the free electrons  $\epsilon_{el}(k, \omega)$  permits one to calculate the plasmon dispersion relation. In fact, from Eq. (38), neglecting the ionic contribution, we obtain

$$\omega^2(k) = \tilde{\omega}_p^2 \frac{\epsilon_{el}(k, 0)/\epsilon_\infty}{\epsilon_{el}(k, 0)/\epsilon_\infty - 1}. \quad (43)$$

It is worth pointing out that the two equivalent formula-

tions of the problem have different advantages. Whereas the dielectric formulation, summarized in Eq. (36), provides us with the ability to obtain more reliable renormalized frequencies  $\Omega_i$  and coupling coefficients of the fields with the external charge any time a better model dielectric function is available, the Hamiltonian formulation permits us to construct reliable PP and BPP states and effective electron-electron potentials, as we will see in the following sections.

The equivalence indicates also that our Hamiltonian formulation has good physical grounds, because a dielectric function written as sum of the ionic and electronic

parts is the random-phase approximation for an electronic interaction sum of the bare electron-electron and electron-phonon interactions.<sup>32</sup>

#### IV. VARIATIONAL METHOD

The conservation law of the total momentum [Eq. (11)] is taken into account through the unitary transformation

$$U = \exp \left[ i \left[ \mathbf{Q} - \sum_{\mathbf{k}} \mathbf{k} \alpha_{\mathbf{k}_1}^\dagger \alpha_{\mathbf{k}_1} - \sum_{\mathbf{k}} \mathbf{k} \alpha_{\mathbf{k}_2}^\dagger \alpha_{\mathbf{k}_2} \right] \cdot \mathbf{R} \right]. \quad (44)$$

The transformed Hamiltonian reads

$$\begin{aligned} \tilde{H} = U^{-1} H U = & \frac{\hbar^2}{2M} \left[ \mathbf{Q} - \sum_{\mathbf{k}} \mathbf{k} \alpha_{\mathbf{k}_1}^\dagger \alpha_{\mathbf{k}_1} - \sum_{\mathbf{k}} \mathbf{k} \alpha_{\mathbf{k}_2}^\dagger \alpha_{\mathbf{k}_2} \right]^2 + \frac{p^2}{2\mu} + \frac{e^2}{\epsilon_{\infty} r} \\ & + \sum_{\mathbf{k}} \left\{ \hbar \Omega_1 (\alpha_{\mathbf{k}_1}^\dagger \alpha_{\mathbf{k}_1} + \frac{1}{2}) + \hbar \Omega_2 (\alpha_{\mathbf{k}_2}^\dagger \alpha_{\mathbf{k}_2} + \frac{1}{2}) + [\rho_{\mathbf{k}}(\mathbf{r}) (\tilde{V}_{\mathbf{k}} \alpha_{\mathbf{k}_1} + \tilde{U}_{\mathbf{k}} \alpha_{\mathbf{k}_2}) + \text{H.c.}] \right\}, \end{aligned} \quad (45)$$

where  $\hbar \mathbf{Q}$  is the eigenvalue of  $\mathcal{P}$ . The trial ground state is

$$|\psi\rangle = U_1(\mathbf{r}) U_2(\mathbf{r}) |0\rangle \phi(\mathbf{r}), \quad (46)$$

where  $|0\rangle$  is the vacuum of  $\alpha_{\mathbf{k}_1}$  and  $\alpha_{\mathbf{k}_2}$ . The envelope function  $\phi$  is chosen to be as a 1s hydrogeniclike wave function and the operators  $U_1$  and  $U_2$  are given by

$$U_1(\mathbf{r}) = \exp \left[ \sum_{\mathbf{k}} [f_{\mathbf{k}}(\mathbf{r}) \alpha_{\mathbf{k}_1} - f_{\mathbf{k}}^*(\mathbf{r}) \alpha_{\mathbf{k}_1}^\dagger] \right], \quad (47)$$

$$U_2(\mathbf{r}) = \exp \left[ \sum_{\mathbf{k}} [g_{\mathbf{k}}(\mathbf{r}) \alpha_{\mathbf{k}_2} - g_{\mathbf{k}}^*(\mathbf{r}) \alpha_{\mathbf{k}_2}^\dagger] \right]. \quad (48)$$

The set of functions  $\phi$ ,  $f_{\mathbf{k}}(\mathbf{r})$ , and  $g_{\mathbf{k}}(\mathbf{r})$  are determined variationally minimizing the total energy

$$\begin{aligned} E_T(\mathbf{Q}) = \langle \psi | \tilde{H} | \psi \rangle \\ = \left\langle \phi \left| \left[ \frac{\hbar^2}{2M} (\mathbf{Q} - \mathbf{K}_1 - \mathbf{K}_2)^2 + \frac{1}{2\mu} (\mathbf{j}_1 + \mathbf{j}_2)^2 + \frac{p^2}{2\mu} + \frac{e^2}{\epsilon_{\infty} r} + \frac{\hbar^2}{2\mu} \sum_{\mathbf{k}} [|\nabla f_{\mathbf{k}}(\mathbf{r})|^2 + |\nabla g_{\mathbf{k}}(\mathbf{r})|^2] \right. \right. \\ \left. \left. + \frac{\hbar^2}{2M} \sum_{\mathbf{k}} [k^2 |f_{\mathbf{k}}(\mathbf{r})|^2 + k^2 |g_{\mathbf{k}}(\mathbf{r})|^2] + \sum_{\mathbf{k}} [\hbar \Omega_1 |f_{\mathbf{k}}(\mathbf{r})|^2 + \hbar \Omega_2 |g_{\mathbf{k}}(\mathbf{r})|^2] \right. \right. \\ \left. \left. - \sum_{\mathbf{k}} \{ \rho_{\mathbf{k}} [\tilde{V}_{\mathbf{k}} f_{\mathbf{k}}^*(\mathbf{r}) + \tilde{U}_{\mathbf{k}} g_{\mathbf{k}}^*(\mathbf{r})] + \text{c.c.} \} \right] \right| \phi \rangle, \end{aligned} \quad (49)$$

where we have defined

$$\mathbf{K}_1 = \sum_{\mathbf{k}} \mathbf{k} |f_{\mathbf{k}}(\mathbf{r})|^2, \quad (50)$$

$$\mathbf{K}_2 = \sum_{\mathbf{k}} \mathbf{k} |g_{\mathbf{k}}(\mathbf{r})|^2, \quad (51)$$

$$\mathbf{j}_1 = \frac{\hbar}{2i} \sum_{\mathbf{k}} [f_{\mathbf{k}}(\mathbf{r}) \nabla f_{\mathbf{k}}^*(\mathbf{r}) - f_{\mathbf{k}}^*(\mathbf{r}) \nabla f_{\mathbf{k}}(\mathbf{r})], \quad (52)$$

$$\mathbf{j}_2 = \frac{\hbar}{2i} \sum_{\mathbf{k}} [g_{\mathbf{k}}(\mathbf{r}) \nabla g_{\mathbf{k}}^*(\mathbf{r}) - g_{\mathbf{k}}^*(\mathbf{r}) \nabla g_{\mathbf{k}}(\mathbf{r})]. \quad (53)$$

The functional variation of  $E_T(\mathbf{Q})$  with respect to  $f_{\mathbf{k}}^*(\mathbf{r})$  and  $g_{\mathbf{k}}^*(\mathbf{r})$  gives differential equations

$$\begin{aligned} -\frac{\hbar^2}{2\mu} \left[ \nabla^2 f_{\mathbf{k}}(\mathbf{r}) + \frac{1}{\phi^2} \nabla \phi^2 \cdot \nabla f_{\mathbf{k}}(\mathbf{r}) \right] + \left[ \hbar \Omega_1 + \frac{\hbar^2 k^2}{2M} - \frac{\hbar^2}{M} \mathbf{k} \cdot (\mathbf{Q} - \mathbf{K}_1 - \mathbf{K}_2) \right] f_{\mathbf{k}}(\mathbf{r}) \\ - \frac{\hbar}{2i\mu} \frac{1}{\phi^2} \{ \nabla \cdot [(\mathbf{j}_1 + \mathbf{j}_2) \phi^2 f_{\mathbf{k}}(\mathbf{r})] + \phi^2 (\mathbf{j}_1 + \mathbf{j}_2) \cdot \nabla f_{\mathbf{k}}(\mathbf{r}) \} = \tilde{V}_{\mathbf{k}} \rho_{\mathbf{k}} \end{aligned} \quad (54)$$

and

$$-\frac{\hbar^2}{2\mu} \left[ \nabla^2 g_{\mathbf{k}}(\mathbf{r}) + \frac{1}{\phi^2} \nabla \phi^2 \cdot \nabla g_{\mathbf{k}}(\mathbf{r}) \right] + \left[ \hbar \Omega_2 + \frac{\hbar^2 k^2}{2M} - \frac{\hbar^2}{M} \mathbf{k} \cdot (\mathbf{Q} - \mathbf{K}_1 - \mathbf{K}_2) \right] g_{\mathbf{k}}(\mathbf{r}) - \frac{\hbar}{2i\mu} \frac{1}{\phi^2} \{ \nabla \cdot [(\mathbf{j}_1 + \mathbf{j}_2) \phi^2 g_{\mathbf{k}}(\mathbf{r})] + \phi^2 (\mathbf{j}_1 + \mathbf{j}_2) \cdot \nabla g_{\mathbf{k}}(\mathbf{r}) \} = \tilde{U}_k \rho_k . \quad (55)$$

These differential equations are nonlinear and coupled through  $j_{1,2}$  and  $\mathbf{K}_{1,2}$ . Setting  $Q=0$  and assuming  $\mathbf{j}_1 = \mathbf{j}_2 = \mathbf{K}_1 = \mathbf{K}_2 = \mathbf{0}$ , it is seen that the solutions  $f_{\mathbf{k}}(\mathbf{r})$  and  $g_{\mathbf{k}}(\mathbf{r})$  are consistent with this assumption. However, this is not true when  $Q \neq 0$ . In this case we use an approximation borrowed from polaron theory, which allows us to linearize the equations. This approximation consists in substituting  $\mathbf{K}_1$  and  $\mathbf{K}_2$  with their average values on  $\phi$ ,

$$\langle \phi | \mathbf{K}_{1,2} | \phi \rangle = \sigma_{1,2} \mathbf{Q} , \quad (56)$$

where the assumed proportionality to  $\mathbf{Q}$  allows the self-consistent evaluation of the constants  $\sigma_{1,2}$ . The contributions from  $\mathbf{j}_{1,2}$  are negligible.

Solutions to Eqs. (54) and (55) can be searched for in the form of series of spherical harmonics,

$$f_{\mathbf{k}}(\mathbf{r}) = \left[ \frac{(2\pi)^3}{V} \right]^{1/2} \frac{1}{k} \sum_{l,m} \tilde{f}_{\mathbf{k},l}(r) Y_{l,m}^*(\omega_{\mathbf{k}}) Y_{l,m}(\omega_{\mathbf{r}}) , \quad (57)$$

$$g_{\mathbf{k}}(\mathbf{r}) = \left[ \frac{(2\pi)^3}{V} \right]^{1/2} \frac{1}{k} \sum_{l,m} \tilde{g}_{\mathbf{k},l}(r) Y_{l,m}^*(\omega_{\mathbf{k}}) Y_{l,m}(\omega_{\mathbf{r}}) . \quad (58)$$

The envelope function  $\phi$  is chosen to be of the form

$$\phi(r) = \frac{(2\gamma)^{\beta+3/2}}{\sqrt{\Gamma(2\beta+3)}} r^{\beta} e^{-\gamma r} , \quad (59)$$

with  $\gamma$  and  $\beta$  variational parameters. With these approximations it is possible to find an exact solution of both Eqs. (54) and (55),

$$\begin{aligned} \tilde{f}_{\mathbf{k},l} = & \frac{2\mu}{\hbar^2} \left[ \frac{V}{(2\pi)^3} \right]^{1/2} \tilde{V}_k 4\pi i^l \xi_1^l r^{2l} e^{-\xi_1 r/2} \frac{\Gamma(a_1)}{\Gamma(b)} \\ & \times \left[ \Phi(a_1, b, \xi_1 r) \int_{\xi_1 r}^{\infty} dt t^{b-\xi_1} e^{-t(1-\epsilon_1/2\xi_1)} j_l(kt/2\xi_1) [1+(-1)^l] \Psi(a_1, b, \xi_1 t) \right. \\ & \left. \times \Psi(a_1, b, \xi_1 r) \int_0^{\xi_1 r} dt t^{b-\xi_1} e^{-t(1-\epsilon_1/2\xi_1)} j_l(kt/2\xi_1) [1+(-1)^l] \Phi(a_1, b, \xi_1 t) \right] , \quad (60) \end{aligned}$$

where

$$\xi = \frac{[(2\beta+1)^2 + 4l(l+1)]^{1/2} - (2\beta+1)}{2} , \quad (61)$$

$$\xi_1 = \left[ 4\gamma^2 + \frac{8\mu}{\hbar^2} \left[ \hbar \Omega_1 + \frac{\hbar^2 k^2}{2M} - \frac{\hbar^2}{M} \mathbf{k} \cdot \mathbf{Q} (1-\sigma) \right] \right]^{1/2} , \quad (62)$$

$$\epsilon_1 = \xi_1 - 2\gamma , \quad (63)$$

$$b = 2(\beta+1+\xi) , \quad (64)$$

$$a_1 = \xi + \frac{\epsilon_1}{\xi_1} (\beta+1) , \quad (65)$$

$$\sigma = \sigma_1 + \sigma_2 . \quad (66)$$

The solution for  $g_{\mathbf{k},l}$  is identical in form to Eq. (60) and is obtained by changing the subscript 1 with 2.

The actual calculation of the BPP binding energies and

effective mass is organized in several steps. The first step is the self-consistent determination of  $\sigma_1$  and  $\sigma_2$  for chosen values of the variational parameters  $\gamma$  and  $\beta$ . The second step consists in the calculation of the total energy [Eq. (49)] corresponding to a given  $\mathbf{Q}$ . This procedure is repeated for several  $\gamma$ 's and  $\beta$ 's until a minimum in the total energy is found.

It can be shown analytically that for  $\gamma=0$  and  $\beta=0$  the total energy  $E_T(Q)$  reduces to that of two free PP's,  $E_{as}(Q)$ , so that it is correct to define the BPP binding energy  $E_b$  as the difference between the total energy and that of two separated PP's both calculated at  $Q=0$ .

When  $E_b \leq 0$  the BPP can form and it is in a bound state. Physically, this is due to the competition between the polarization energy and the Coulomb repulsion. The choice of the envelope function [Eq. (59)] reflects this competition.

In the limit  $Q \rightarrow 0$ ,  $E_T(Q)$  and  $E_{as}(Q)$  can be written in the form

$$E_T(Q) = E_{T,0} + \frac{\hbar^2}{2M^*} Q^2, \quad (67)$$

$$E_{as}(Q) = E_{as,0} + \frac{\hbar^2}{4m^*} Q^2, \quad (68)$$

where  $M^*$  and  $m^*$  are the BPP and PP effective masses, respectively. Their estimation is obtained by fitting Eqs. (67) and (68) to the numerical data. It is found that  $M^*$  and  $m^*$  are independent on  $Q$  for a large range of this quantity in the Brillouin zone.

## V. EFFECTIVE POTENTIAL

The minimization of Eq. (49) with respect to the state  $\phi$  shows that it is an eigenstate of the Schrödinger-like equation of a particle of mass  $\mu$  with the effective self-consistent electron-electron potential

$$\begin{aligned} V_{\text{eff}}(r) = & \frac{e^2}{\epsilon_\infty r} + \frac{\hbar^2}{2\mu} \sum_{\mathbf{k}} [|\nabla f_{\mathbf{k}}(\mathbf{r})|^2 + |\nabla g_{\mathbf{k}}(\mathbf{r})|^2] \\ & + \frac{\hbar^2}{2M} \sum_{\mathbf{k}} [k^2 |f_{\mathbf{k}}(\mathbf{r})|^2 + k^2 |g_{\mathbf{k}}(\mathbf{r})|^2] \\ & + \sum_{\mathbf{k}} [\hbar\Omega_1 |f_{\mathbf{k}}(\mathbf{r})|^2 + \hbar\Omega_2 |g_{\mathbf{k}}(\mathbf{r})|^2] \\ & - \sum_{\mathbf{k}} \{ \rho_k [\tilde{V}_k f_{\mathbf{k}}(\mathbf{r}) + \tilde{U}_k g_{\mathbf{k}}(\mathbf{r})] + \text{c.c.} \}. \end{aligned} \quad (69)$$

This quantity has very interesting properties. It will be studied systematically in a future work, showing here only that it contains many important physical features. We consider only the case  $Q=0$ .

In the limit  $r \rightarrow \infty$ ,  $V_{\text{eff}}$  can be calculated from (69) taking for  $f_{\mathbf{k}}(\mathbf{r})$  and  $g_{\mathbf{k}}(\mathbf{r})$  the corresponding asymptotic expression

$$f_{\mathbf{k}}(\mathbf{r}) = \frac{2\mu}{\hbar^2} \tilde{V}_k \frac{e^{i(\mathbf{k}\cdot\mathbf{r})/2} + e^{-i(\mathbf{k}\cdot\mathbf{r})/2}}{[\gamma^2 + (2\mu/\hbar)\Omega_1 + \frac{1}{2}k^2]^{1/2}}, \quad (70)$$

$$g_{\mathbf{k}}(\mathbf{r}) = \frac{2\mu}{\hbar^2} \tilde{U}_k \frac{e^{i(\mathbf{k}\cdot\mathbf{r})/2} + e^{-i(\mathbf{k}\cdot\mathbf{r})/2}}{[\gamma^2 + (2\mu/\hbar)\Omega_2 + \frac{1}{2}k^2]^{1/2}}. \quad (71)$$

The  $V_{\text{eff}}$  obtained in this way contains a constant term  $E_{as,0}$ , which represents the self-energy of the pair. This conclusion is similar to that obtained by other authors for the excitons.<sup>30,31</sup> To understand the features of  $V_{\text{eff}}$ , it is useful to calculate it using for  $f_{\mathbf{k}}(\mathbf{r})$  and  $g_{\mathbf{k}}(\mathbf{r})$  Eqs. (70) and (71) with  $\gamma=0$  and assuming that the plasma frequency is not dispersive.

The terms not depending on  $r$  give  $E_{as,0}$ , i.e.,

$$E_{as,0} = -2\alpha\hbar\omega_l \left[ \frac{F_1^2}{\sqrt{R_1}} + \frac{F_2^2}{\sqrt{R_2}} \right], \quad (72)$$

where

$$\begin{aligned} F_1 = & \lambda \frac{\sqrt{(1-\eta)/R_1} + (R_1^2 - 1)/\sqrt{(1-\eta)R_1}}{[(1-R_1^2)^2 + \lambda^2(1-\eta)]^{1/2}}, \\ F_2 = & -\lambda \frac{\sqrt{(1-\eta)/R_2} + (R_2^2 - 1)/\sqrt{(1-\eta)R_2}}{[(1-R_2^2)^2 + \lambda^2(1-\eta)]^{1/2}}, \end{aligned} \quad (73)$$

and  $R_1 = \Omega_1/\hbar\omega_l$ ,  $R_2 = \Omega_2/\hbar\omega_l$ . From this result some particular cases can be studied.

(a) If  $\lambda \rightarrow 0$ ,  $-2\alpha\hbar\omega_l$  is found, which is the self-energy of two free electrons interacting only with the phonon field.

(b) If  $\eta \rightarrow 1$ , which means that the ionicity of the crystal is not considered,  $E_{as,0} \rightarrow E_{pl} = -e^2/\epsilon_\infty R_{pl}$ , with  $R_{pl} = (\hbar\sqrt{\epsilon_\infty}/2m\omega_p)^{1/2}$ . This is the self-energy of two electrons interacting with the plasmon field in an approximation similar to that of Fröhlich for the phonon field. This value, as that for se polaron, is due to the dynamics of the external electrons and plasmons.

(c) If  $\lambda \rightarrow \infty$ , we obtain the same limit as point (b). This means that the increase of the electron density screens the ionic effects, but the self-energy due to the plasmon field tends to  $-\infty$ . In Fig. 1 we show the quantity  $\Delta = E_{as,0} - E_{pl}$  as function of  $\lambda$ .  $\Delta$  can be interpreted as the phonon contribution to the self-energy of the pair, including the effects due to the dynamics of other electrons. We see that for  $\lambda \leq 2$  (i.e.,  $n \leq 10^{20} \text{ cm}^{-3}$ ) the dynamics of the electrons tends to screen partially the electron-phonon interaction. This fact is relevant for bipolaron formation. On the other hand, for metallic densities ( $n \sim 10^{22} \text{ cm}^{-3}$ ), the electron screening is complete, as expected.

In  $V_{\text{eff}}$  there is also a long-range tail given by

$$V_{\text{LR}}(r) = \frac{e^2}{\epsilon_\infty r} - \frac{e^2}{\epsilon_\infty r} \left( \frac{F_1^2}{R_1} + \frac{F_2^2}{R_2} \right), \quad (74)$$

whose properties are the following.

(a) For  $\lambda \rightarrow 0$ , it becomes  $e^2/\epsilon_0 r$ . This means that,

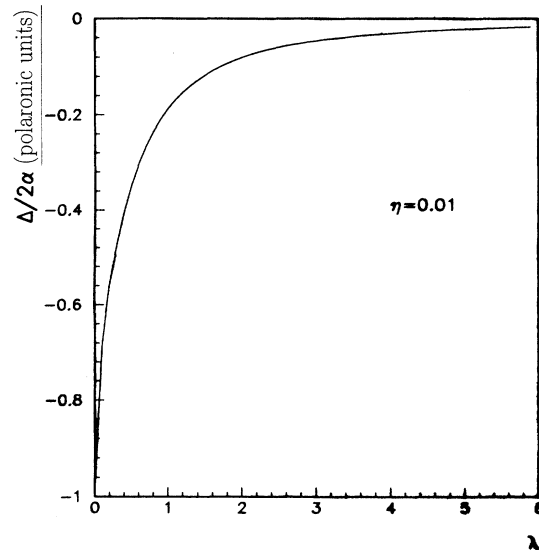


FIG. 1. Difference between the pair self-energy in the phonon and plasmon field and that of the plasmon pair self-energy,  $\Delta/2\alpha$ , as a function of  $\lambda$  ( $\Delta$  measures only the ionic contribution). We see that the phonon effects are meaningful for electronic densities less than  $\lambda=4$  (i.e.,  $n \sim 10^{20} \text{ cm}^{-3}$ ). We stress that in the high- $T_c$  superconductors the densities are only slightly higher.



when there is only the phonon field, the electron-electron potential, at large distance, is screened by the static dielectric constant. This result has been also obtained in the case of the excitons.<sup>37</sup>

(b) For  $\eta \rightarrow 1$ , the long-range tail  $V_{LR}$  is zero. This means that, if we have only the plasmon field, the electron-electron potential is screened by the other electrons.

(c) For  $\lambda \rightarrow \infty$ , we obtain the same result as point (b), which means that, in the large density limit, the free electrons screen completely the electron-phonon and electron-electron interactions.

The remaining terms are exponentially decaying with screening constant given by  $R_p^{-1} = (2m\omega_l/\hbar)^{1/2}$  and  $R_{pl}^{-1}$ . The relative weight of these depends on  $\lambda$ : For  $\lambda \rightarrow 0$ , the screening factor is  $R_p^{-1}$  and in the opposite limit is  $R_{pl}^{-1}$ .

In the above discussion it appears that, also when the electronic density is large, the self-energy of the polarons is always due to dynamical effects, whereas in the exciton case<sup>30,31</sup> it has been shown that it is  $-e^2k_0/\epsilon_\infty$ , where  $k_0$  is the Thomas-Fermi screening constant. We do not discuss such problems in detail in this work, but we show only that, taking for example for  $\epsilon_{el}(k,0)$  the Thomas-Fermi expression to calculate through Eq. (43) the dispersion of the plasmon branch, such a limit is contained in our model. In fact, we obtain

$$\omega(k) = \omega_p \left[ 1 + \frac{k^2}{k_0^2} \right]^{1/2}, \quad (75)$$

which is approximated by the expression

$$\omega(k) = \omega_p \left[ 1 + \frac{k^2}{2k_0^2} \right] \quad (76)$$

for small  $k$  to permit analytical calculations. Neglecting ionicity effects, the self-energy is given by

$$E_{as,0} = -\frac{e^2}{\epsilon_\infty R_{pl}} \left[ \left( 1 + \frac{1}{2R_{pl}^2 k_0^2} \right)^{1/2} - \frac{1}{\sqrt{2}R_{pl}k_0} \right]. \quad (77)$$

From this expression we see that in the limit of high density, i.e.,  $R_{pl}^2 k_0^2 \ll 1$ ,  $E_{as,0} \rightarrow -e^2k_0/\sqrt{2}\epsilon_\infty$ , and in the opposite limit  $E_{as,0} \rightarrow -e^2/\epsilon_\infty R_{pl}$ , which clearly shows that in our model the dynamical effects are more important than the static one in the low-density limit and vice versa. We note that in the high-density limits  $E_{as,0}$  is a fraction of that usually found in the literature. This is due to the development of  $\omega(k)$  done in Eq. (76). Finally, we find that, also in this case, the long-range tail of the electron-electron potential is always zero and that the short-range part decreases exponentially with the Thomas-Fermi screening factor  $k_0$  in the high-density limit and with  $R_{pl}^{-1}$  in the opposite limit.

## VI. RESULTS AND DISCUSSION

Assuming the plasma frequency not dispersive, we have calculated the total energy  $E_T(Q)$  of the BPP, the

self-energy  $E_{as}(Q)$  of two single PP's, the effective mass  $M^*$ , the radius  $R_b$  of the BPP, and the mass  $m^*$  of the PP. Using as units for energies and lengths  $\hbar\omega_l$  and  $R_p$ , respectively, and for the effective masses  $M^*$  and  $m^*$  the band mass  $m$ , the above quantities are functions of  $\lambda$ ,  $\alpha$ , and  $\eta$ . The first parameter is connected to the electronic density through the plasma frequency; its value depends on the electronic density, but also on the band mass of the electron, on the energy  $\hbar\omega_l$  of the longitudinal optical phonon, and on  $\epsilon_\infty$ . Although the results of this section are independent on the values of these quantities, we need to fix them for the calculations of the next one. We take then  $m = 5m_e$ ,  $m_e$  being the free-electron mass,  $\hbar\omega_l = 70$  meV, and  $\epsilon_\infty = 2.5$ , so that  $\lambda = 1$  means an electronic density of  $n = 4.45 \times 10^{19} \text{ cm}^{-3}$ .

We minimize the total energy (49) with respect to  $\beta$  and  $\gamma$ , using, for  $f_k(r)$  and  $g_k(r)$ , Eqs. (57) and (58). In Figs. 2 and 3 we report the binding energies  $E_b = E_{T,0} - E_{as,0}$  and the self-energy  $E_{as,0}/2$  of one free PP for fixed values of  $\alpha$  and  $\eta$  as a function of  $\lambda$ . The value of  $\alpha$  chosen is 8 and those of  $\eta$  are 0.01 and 0.05. These choices are reliable for high- $T_c$  superconductor materials. We know<sup>22</sup> that when the bipolaron exists ( $\alpha \geq \alpha_c$ ) there is a maximum value of  $\eta$ ,  $\eta_c$  such that for  $\eta \geq \eta_c$ , the bipolaron cannot form. The values of  $\eta$  considered are such that, for  $\alpha = 8$ ,  $\eta = 0.01$  and 0.05 are smaller and larger than  $\eta_c$ , respectively.

We see that, for  $\eta = 0.01$ , the binding energy  $E_b$  is negative for small  $\lambda$  (the BPP can form), but becomes positive with increasing  $\lambda$ . In these last cases there is a relative minimum for  $E_{T,0}$ , so that the BPP exists in a metastable state. It appears clearly from the numerical results for  $\lambda \leq 1$  that cooperative effects between plasmons and phonons increase the stability of the BPP; for larger  $\lambda$ , the screening effects become more important and the

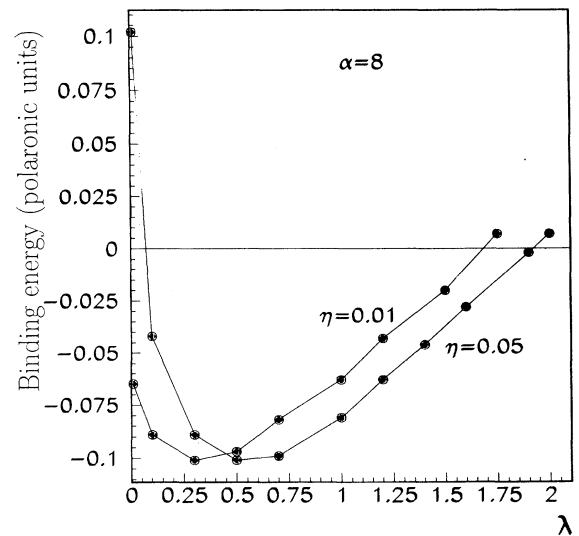


FIG. 2. Binding energies of the bi-plasma-polaron are shown as function of  $\lambda$  for  $\alpha = 8$  and  $\eta = 0.01$  and 0.05.  $\lambda = 1$  indicates an electronic density of  $4.45 \times 10^{19} \text{ cm}^{-3}$ . The energies are in units of  $\hbar\omega_l$ .

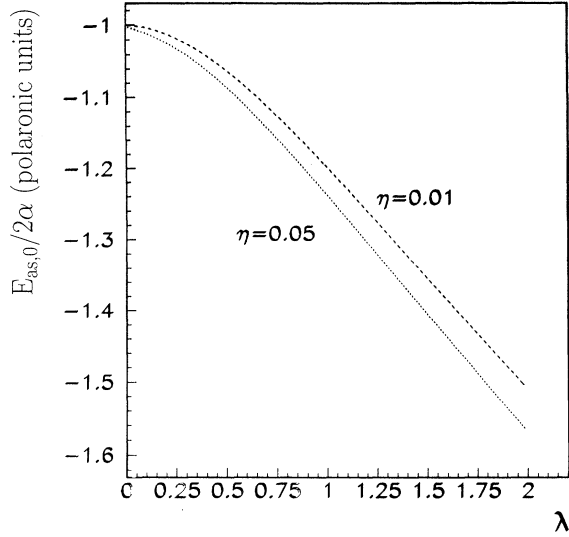


FIG. 3. Since the self-energy of a free plasma polaron is proportional to  $\alpha$ , the quantity  $E_{as,0}/2\alpha$  is drawn as function of  $\lambda$  for  $\eta=0.01$  and  $0.05$ . The energies are in units of  $\hbar\omega_l$ .

BPP state becomes metastable. From Fig. 3 it can be seen that the PP self-energy increases going from  $\eta=0.01$  to  $0.05$ . Since a larger  $\eta$  implies a smaller ionicity of the lattice, the increase of the self-energy is ascribed to plasmon exchange. A similar behavior can also be seen in Fig. 2 where, for  $\eta=0.05$ ,  $E_b$ , for small  $\lambda$ , is larger with respect to  $\eta=0.01$ ; the BPP state starts out as metastable and then becomes bound. Increasing  $\lambda$ ,  $E_b$  becomes comparable in the two cases while the states are found. Finally, increasing further  $\lambda$ ,  $E_b$  increases and becomes zero or slightly positive. We have done our calculations for  $\lambda \leq 2$  because the dilution parameter  $nR_b^3$  does not become larger than 1, as we will see later.

The BPP radius is calculated through

$$R_b = \int r^3 \phi(r)^2 dr, \quad (78)$$

so that also the dilution parameter  $nR_b^3$  can be known. We find that the BPP radii are nearly constant for given  $\alpha$  and  $\eta$  as function of  $\lambda$ ; their values, for  $\alpha=8$ , are 3.7 and 3.9 for  $\eta=0.01$  and  $\eta=0.05$ , respectively. The dilution parameter ranges from  $10^{-5}$  for  $\lambda=0.01$  to 1 for  $\lambda=2$ , and there are no significant variations with  $\eta$ . We see that the BPP radius and the dilution parameter are so small as to justify the hypothesis to consider only one electron pair and to neglect the correlation and exchange terms. We can also see that the Pippard coherence length  $\xi_0 = (\sqrt{3}/2\pi)R_b$  is small (tens of Å) and that  $k_F \xi_0$ , where  $k_F$  is the Fermi wave number, is always  $\leq 1$ , in agreement with the results for the high- $T_c$  superconductors.

Finally, in Figs. 4 and 5 we show  $M^*$  and  $m^*$ , as functions of  $\lambda$ , for fixed  $\alpha$  and  $\eta$ . Since the plasmon branch is assumed not dispersive,  $m^*$ , the PP effective mass, can be calculated analytically. We find

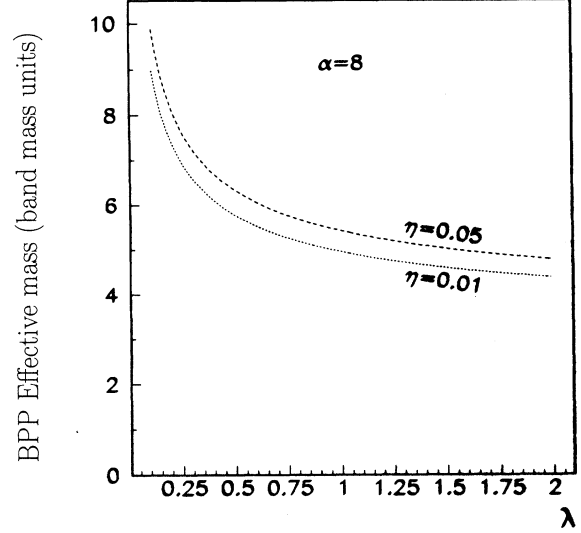


FIG. 4. Bi-plasma-polaron effective mass  $M^*$  are shown as function of  $\lambda$  for  $\alpha=8$  and  $\eta=0.01$  and  $0.05$ .

$$\frac{m^*}{m} = 1 + \frac{\alpha}{6} \left[ \frac{F_1^2}{R_1^{3/2}} + \frac{F_2^2}{R_2^{3/2}} \right]. \quad (79)$$

From Fig. 4 it can be seen that  $m^*$  is divergent for  $\lambda \rightarrow 0$ . This divergence deserves a comment. First of all, we observe that the branch  $\Omega_2$  vanishes for  $\lambda \rightarrow 0$ . It is well known from the polaron theory<sup>36</sup> that when the boson frequency vanishes an unphysical divergency of the mass is introduced which can be removed by taking into account two or more boson processes. As far as our calculation is concerned, we do not attempt to account for those high-order processes. As a consequence, the results

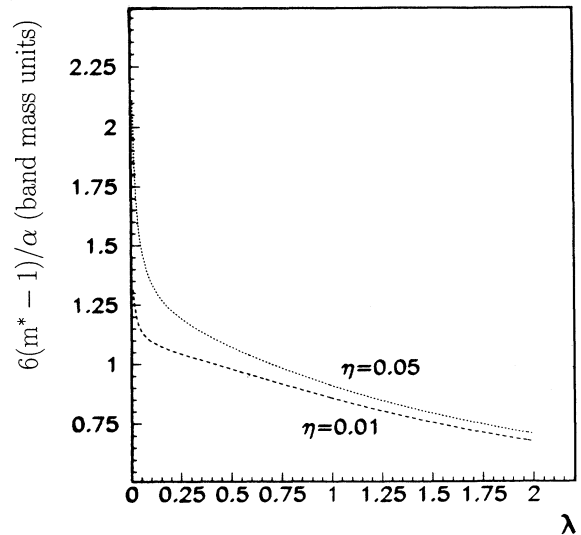


FIG. 5. Quantity  $6(m^* - 1)/\alpha$  is shown as function of  $\lambda$  for  $\eta=0.01$  and  $0.05$ .

concerning the PP and BPP cannot be trusted for  $\lambda < 0.1$ .

From Fig. 4 it can be seen that  $M^*$  behaves in a way similar to  $m^*$ . In particular, for large  $\lambda$  it tends to  $2m^*$ , as we expect. On the other hand,  $m^*$  is a decreasing function of  $\lambda$  and for  $\lambda \rightarrow \infty$  goes to  $m$ .

### VII. BPP BOSE CONDENSATION

Since we have found that the BPP radii are so small that, for any density considered,  $nR_b^3 \leq 1$ , we consider these composite particles as well-defined bosons responsible for the supercurrent in the high- $T_c$  materials. The properties of the system can be studied following the approach introduced by Blatt,<sup>33</sup> who considered a two-fluid model where PP's, and BPP's are in thermodynamical equilibrium, respectively. We calculate both the Bose-condensation critical temperature and the PP and BPP relative populations as follows. Because of the particle conservation, the total number of particles,  $N$ , in the system can be written as the sum of the PP's and BPP's present,

$$N = \sum_k \{ \exp[\beta_b(\epsilon_k - \mu)] + 1 \}^{-1} + 2 \sum_k \{ \exp[\beta_b(\eta_k - 2\mu)] - 1 \}^{-1}, \quad (80)$$

where  $\beta_b = 1/K_b T$  ( $K_b$  and  $T$  are the Boltzmann constant and the temperature, respectively) and  $\epsilon_k$  and  $\eta_k$  are the excitation spectra of the PP and BPP, respectively. On the basis of the results of the previous sections, the spectra for both kinds of particles are

$$\epsilon_k = \frac{\hbar^2 k^2}{2m^*}, \quad \eta_k = E_b + \frac{\hbar^2 k^2}{2M^*}. \quad (81)$$

In Eq. (80) we have used the condition for the chemical equilibrium<sup>32</sup> which fixes the relation between the two chemical potentials  $\mu = \mu_{PP} = \mu_{BPP}/2$ . From Eq. (80) we obtain  $\mu = \mu(N, \beta_b)$ . In particular, the critical temperature  $T_c$  is found from the same Eq. (80), setting  $\mu = E_b/2$ .

In Fig. 6 we show the critical temperature versus the normalized total particle density  $\lambda$ , for  $\alpha = 8$  and  $\eta = 0.01$ . We note that  $T_c$  is near to the Bose-condensation temperature  $\bar{T}_c$ ,

$$\bar{T}_c = \frac{h^2}{5.224\pi k_B} \frac{n^{2/3}}{M^*} \quad (82)$$

of a gas of density  $n = N/V$  for small  $\lambda$ ; for higher  $\lambda$ , we have small deviations due to the increase of the fermion's population.

Our rough estimate of the critical temperature suffers from an overestimation of the screening effects. This follows from the fact that the binding energy  $E_b$  is always calculated as if all the particles participate in the screening as unbound PP's. In reality, as soon as BPP's are formed, the number of PP's participating in the screening reduces, leading to a reduction of the screening itself. This effect is known in the case of the excitons when the screening associated with them<sup>31</sup> is negligible compared to that of the electrons.

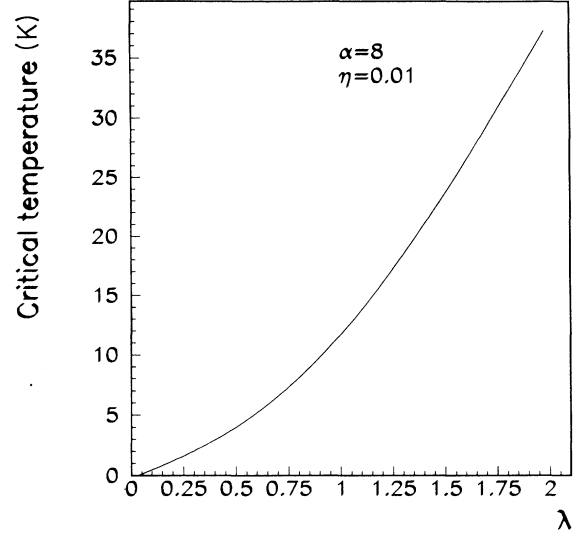


FIG. 6.  $T_c$  is drawn as a function of  $\lambda$  for  $\alpha = 8$  and  $\eta = 0.01$ . We assume  $m = 5m_e$ ,  $\hbar\omega_l = 70$  meV, and  $\epsilon_\infty = 2.5$ .

### VIII. CONCLUSIONS

In this work we have constructed a Hamiltonian in which two repelling external charges are interacting with a longitudinal optical-phonon field and with a sea of other electrons. The interaction of the phonons with the external charges is treated in the Fröhlich scheme,<sup>27</sup> the sea of the free electrons is treated in the single-pole approximation,<sup>28</sup> and its interaction with the external charges is described through an electron-plasmon term.<sup>29</sup> The dispersion of the plasmon branch is taken also into account.

It is shown that the model is equivalent to a description of the system through a dielectric function in which the ionic and electronic contributions are simply summed. Such a dielectric function is the random-phase approximation of the sum of the bare Coulomb and the electron-phonon interaction.<sup>32</sup>

The boson fields terms in the Hamiltonian and their interaction can be diagonalized and new interactions between the external charges and the renormalized fields can be defined. The couplings depend now on the density of the electrons through the plasma frequency.

The Hamiltonian commutes with the total momentum of the system, i.e., the sum of the center-of-mass momentum of the pair and the phonons and plasmons momenta. The ground state takes into account such a conservation law, and it is found using a self-consistent variational method. The envelope function of this state depends on the two parameters  $\beta$  and  $\gamma$  such that, if they tend to zero, the energy of the state and the wave function become that of two free PP's. It is then physically reliable to define the binding energy of the pair as the difference between the total energy and that of two free PP's at zero total momentum. We find that there are absolute or local minima of the total energy, so that the pair can be in a bound or a metastable state. Since the total momentum

is conserved, it is possible to define the effective BPP mass and compare it with that of two PP's.

We find that for small density ( $\lambda \leq 1$ ) the phonons and plasmons give cooperative effects to make more stable the BPP and to increase its effective mass with respect to those of two free PP's. For larger  $\lambda$  screening effects become more important, so that the binding energy becomes positive and the effective BPP mass tends to that of two free PP's. On the other hand, the PP effective mass decreases monotonically increasing the density (it goes from the polaron mass to band mass), because the screening effects are more important for increasing  $\lambda$ .

It is possible to define an effective electron-phonon potential which takes into account the polarization effects due to the phonons and plasmons. Its asymptotic limit is the self-energy of the pair; furthermore, the phonons contribute to a Coulomb long-range tail for the potential and the electrons tend to screen the interactions.

In the case considered of a nondispersive plasma branch, the self-energy terms are due only to the dynamical screening. We have also found that, using a Thomas-Fermi screening for the plasmons, there is a competition between the static and dynamical screening effects such that for small electronic density the dynamical effects are more important and for high density static effects are.

Using these results, it is possible, in a two-fluid model,<sup>33</sup> to calculate the Bose-condensation temperature of the BPP gas in thermodynamical equilibrium with the PP gas. We find that  $T_c$  increases with  $\lambda$  and becomes of the

order of 50 K for  $n \sim 10^{20} \text{ cm}^{-3}$ . For all the electronic densities considered, the dilution parameter  $nR_b^3$  is not longer than 1, so that it is physically meaningful to consider a pair as a defined single excitation.

If we would apply the obtained results to the high- $T_c$  superconductors, we stress that such materials are strongly ionic<sup>5,38</sup> ( $\eta \ll 1$ ) and the effective carrier mass and longitudinal optical-phonon frequency are large, as we assumed. Many of them are strongly anisotropic, but there are also isotropic materials such as the fullerene and  $\text{Ba}_{1-x}\text{K}_x\text{BiO}_{3-y}$ . Our theory refers to the last. We predict an increase of the critical temperature with the density, whereas the experimental data show that for larger density the critical temperature decreases. We are not able to consider, in our single-pair approximation, larger densities and consequently calculate the critical temperature because we need to construct a many-body wave function for the system.

It appears that, at the electronic densities studied in this work, the electron dynamics is relevant both for bi-plasma-polaron formation and the effective mass. A reasonable estimate of the electron-phonon coupling constant gives a value near to the upper (lower) limit for which the intermediate (strong) coupling is valid. The conclusion is that in high- $T_c$  superconductors a complete treatment of the electron-phonon interaction should include both features. This work is therefore complementary to those in which the strong electron-phonon coupling scheme is used.

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