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Phase Diagram Approach to the Polaron Problem

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Abstract

The problem of an electron interacting with acoustic as well as with optical modes of lattice vibrations is investigated within the modified variational scheme of the Lee-Low-Pines theory. It is shown that the polaron changes its state according to the values of the electron-optical phonon and the electron-acoustic phonon coupling constants by examining the ground-state energy, the mean numbers of the optical and acoustic phonons in the cloud around the electron and the spatial localization of the electron as functions of the coupling constants. Using the experimental information on the drift mobility of electrons in ionic crystals we estimate with the help of the phase diagram the upper limits of the deformation potential for several materials.

1. Introduction

The problem of an electron interacting with acoustic as well as with optical modes of lattice vibrations has been of considerable interest.¹⁾ The system for this polaron can be in the simplified version characterized by three parameters : the electron-optical phonon coupling constant α , the electron-acoustic phonon coupling constant β and the Debye cut-off wave number k_0 . In the absence of the acoustic coupling the system has long been studied as the standard polaron problem.^{2,3)} The polaron state is classified into two types according to the value of the coupling constant α : the nearly-free state and the self-trapped state. When the interaction is relatively weak the electron behaves more or less like a free particle dressed with a few phonons. The nearly free polaron is well understood in terms of the perturbation⁴⁾ and the intermediate coupling⁵⁾ theories. On the other hand when the interaction is strong enough to cause a strong correlation between the phonons, the electron is captured in a self-induced potential which is built by the field of correlated virtual phonons. The self-trapped polaron is treated with the strong-coupling theory.⁶⁾ There are several theories which interpolate between the weak- and the strong-coupling theories.⁷⁾ The modified variational scheme of the Lee-Low-Pines theory proposed by Huybrechts⁸⁾ is one of such theories. According to Feynman's path integral method the polaron changes gradually its state from the nearly free state to the self-trapped one as the coupling constant increases.⁹⁾ On the other hand in terms of the modified variational scheme of the Lee-Low-Pines theory the polaron changes suddenly its state at a certain value of the coupling constant.^{10,11)} In the optical polaron problem the Debye cut-off k_0 can be taken as infinity because of its long

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range nature of the electron-optical phonon interaction. In the absence of the optical coupling in place of the acoustic the system describes the acoustic polaron via the deformation potential which has been studied by many workers.^{12,13)} As to the acoustic polaron via the deformation potential it is shown that there are also two types of state : the nearly free state and the self-trapped state, and that the acoustic polaron changes suddenly its state from the nearly free state to the self-trapped one at a certain value of the coupling constant β_c within the path integral scheme due to Feynman¹⁴⁾ and the modified variational scheme of the Lee-Low-Pines theory.¹¹⁾ We must take into account the Debye cut-off dependence because of the short range nature of the electron-acoustic phonon interaction. The transition point β_c becomes small with the increasing k_0 . In the presence of both couplings the system has been studied with particular attention to self-trapping within Feynman's path integral method¹⁴⁾ and the adiabatic approximation.¹⁵⁾

In this paper we investigate a system in which an electron interacts simultaneously with acoustic and optical modes of lattice vibrations within the modified variational scheme of the Lee-Low-Pines theory mentioned above. This scheme is rather simple and has been successfully applied not only to the bulk^{10,16-18)} but also to the low-dimensional polaron problems.¹⁹⁻²²⁾ We calculate variationally the ground-state energy, the mean numbers of the optical and acoustic phonons in the cloud around the electron and the spatial localization of the electron as functions of the electron-optical phonon coupling constant α and the electron-acoustic phonon coupling constant β for the definite Debye cut-off k_0 . Phase-transition-like behaviors are seen in the changes of these quantities. The polaron state is classified into three types : the nearly free state, the self-trapped state with the mean number of the optical phonons in the cloud around the electron which is larger than that of the acoustic phonons and the self-trapped state where the mean number of the acoustic phonons in the cloud around the electron is larger than that of the optical phonons, which will be hereafter denoted by F, S and L, respectively. These states can be stable or metastable according to the values of the parameters α and β . Such features are summarized in the phase diagram.

The phase diagram for the polaron is useful in understanding some physical properties of the system. As an application we first consider polar semiconductors such as GaAs and InSb for which the electron-acoustic phonon coupling constants β as well as the electron-optical phonon coupling constants α are experimentally determined.²³⁾ Based on the phase diagram prepared for each material it is shown that the polarons in the polar semiconductors are in the F state. The drift mobility of electrons in these materials should be large consistent with the experiment. Next we consider the ionic crystals such as AgCl and KBr for which the electron-acoustic phonon coupling constants β have not yet been determined. In this case using the experimental information on the drift mobility of electrons in the ionic crystals²⁴⁾ we estimate with the help of the phase diagram the upper limits of the deformation potential for these materials.

In § 2 the Hamiltonian for the system is given. We obtain in the modified variational scheme the expressions for the ground-state energy, the mean numbers of the optical and acoustic phonons in the cloud around the electron and the spatial localization of the electron.

In § 3 these quantities are numerically calculated as functions of the electron-optical phonon coupling constant α and the electron-acoustic phonon coupling constant β for the definite Debye cut-off k_o , which reveals the phase-transition-like behaviors. Such features are well summarized in the phase diagram. In § 4 we apply our phase diagram to the polaronic systems of ionic crystals and polar semiconductors. The final section is devoted to the conclusion.

2. Calculations of the ground-state energy, the mean numbers of phonons and the spatial localization of the electron

We consider a system in which an electron interacts with the longitudinal optical and acoustic phonons in an ionic crystal or a polar semiconductor. The Hamiltonian for the system can be written as

$$\begin{aligned}
 H = & \frac{1}{2} p^2 + \sum_k a_k^\dagger a_k + \sum_k \frac{k}{k_o} b_k^\dagger b_k \\
 & + \left(\frac{2\sqrt{2} \pi \alpha}{V} \right)^{1/2} \sum_k \frac{1}{k} (a_k + a_{-k}^\dagger) \exp(ik \cdot r) \\
 & + \left(\frac{2\sqrt{2} \pi \beta}{V} \right)^{1/2} \sum_k \sqrt{k} (b_k + b_{-k}^\dagger) \exp(ik \cdot r).
 \end{aligned} \tag{1}$$

The first term represents the kinetic energy of the electron. The second and third terms stand respectively for the energies of the optical and acoustic phonon fields. The fourth and last terms represent the interactions of the electron with the optical phonon and with the acoustic phonon fields. Here p and k are the momentum and position of the electron. a_k^\dagger (b_k^\dagger) and a_k (b_k) are creation and annihilation operators for the optical (acoustic) phonon of wave vector k , k_o is the Debye cut-off for the phonon wave number and V is the normalization volume. The dimensionless electron-optical phonon coupling constant α is

$$\alpha = e^2 \left(\frac{1}{\epsilon_\infty} - \frac{1}{\epsilon_o} \right) \left(\frac{m}{2\hbar^3 \omega_o} \right)^{1/2} \tag{2}$$

where e is the electronic charge, m the electron band mass and ω_o the optical phonon frequency. ϵ_o and ϵ_∞ are respectively the static and high-frequency dielectric constants. The dimensionless electron-acoustic phonon coupling constant β is

$$\beta = \frac{\sqrt{2} D^2 m^2}{8\pi\rho \hbar^3 s} \tag{3}$$

where D is the deformation potential constant, ρ the density of the crystal and s the speed of sound. In writing down the Hamiltonian we have made a simplified assumption

$$\hbar \omega_o = \hbar s k_o \tag{4}$$

and used units $\hbar = m = \omega_o = s k_o = 1$.

We introduce a modified Lee-Low-Pines transformation for the present system

$$U_1 = U_1^a U_1^b \tag{5}$$

where

$$U_1^a = \exp\left(-i a \sum_k k \cdot r a_k^\dagger a_k\right) \quad (6)$$

$$U_1^b = \exp\left(-i b \sum_k k \cdot r b_k^\dagger b_k\right) \quad (7)$$

Here a and b are parameters. When a (b)=1 equation (6) ((7)) gives the Lee-Low-Pines transformation. In the limit a (b) \rightarrow 0 it is the identical transformation which leads to the strong coupling approximation. Under the transformation (5) the Hamiltonian can be transformed into

$$\begin{aligned} H' &= U_1^{-1} H U_1 \\ &= \frac{1}{2} (p - a \sum_k k a_k^\dagger a_k - b \sum_k k b_k^\dagger b_k)^2 \\ &\quad + \sum_k a_k^\dagger a_k + \sum_k \frac{k}{k_0} b_k^\dagger b_k \\ &\quad + \left(\frac{2\sqrt{2}\pi\alpha}{V}\right)^{1/2} \sum_k \frac{1}{k} (a_k + a_{-k}^\dagger) \exp(i(1-a)k \cdot r) \\ &\quad + \left(\frac{2\sqrt{2}\pi\beta}{V}\right)^{1/2} \sum_k \sqrt{k} (b_k + b_{-k}^\dagger) \exp(i(1-b)k \cdot r). \end{aligned} \quad (8)$$

To proceed, we make an ansatz about the form of the ground-state wavefunction for the system

$$|e\rangle = |\Phi\rangle \quad (9)$$

where $|e\rangle$ is the electronic part and $|\Phi\rangle$ is the lattice wavefunction. We take a gaussian-type wavefunction for $|e\rangle$

$$|e\rangle = \left(\frac{\lambda}{\pi}\right)^{3/4} \exp\left(-\frac{1}{2} \lambda r^2\right) \quad (10)$$

where λ is a variational parameter. On the other hand in terms of the second Lee-Low-Pines transformation

$$U_2 = \exp\left(\sum_k (f_k a_k^\dagger - f_k^* a_k) + \sum_k (g_k b_k^\dagger - g_k^* b_k)\right) \quad (11)$$

the lattice wavefunction can be expressed as

$$|\Phi\rangle = U_2 |0\rangle \quad (12)$$

where f_k and g_k are variational functions and the phonon ground-state $|0\rangle$ is defined as

$$a_k |0\rangle = 0, \quad b_k |0\rangle = 0,$$

$$\langle 0|0\rangle = 1. \quad (13)$$

Then the ground-state energy for the system can be easily calculated as

$$E = \langle \Phi | \langle e | H' | e \rangle | \Phi \rangle$$

$$\begin{aligned}
&= \frac{3}{4} \lambda + \frac{1}{2} a^2 \sum_k k^2 |f_k|^2 + \frac{1}{2} b^2 \sum_k k^2 |g_k|^2 \\
&\quad + \sum_k |f_k|^2 + \sum_k \frac{k}{k_o} |g_k|^2 \\
&\quad + \left(\frac{2\sqrt{2} \pi \alpha}{V} \right)^{1/2} \sum_k \frac{1}{k} (f_k + f_{-k}^*) \exp\left(-\frac{1}{4\lambda} (1-a)^2 k^2\right) \\
&\quad + \left(\frac{2\sqrt{2} \pi \beta}{V} \right)^{1/2} \sum_k \sqrt{k} (g_k + g_{-k}^*) \exp\left(-\frac{1}{4\lambda} (1-b)^2 k^2\right).
\end{aligned} \tag{14}$$

Here we have used the relations

$$\sum_k k |f_k|^2 = 0 \quad \text{and} \quad \sum_k k |g_k|^2 = 0 \tag{15}$$

which come from the momentum conservation

$$\langle \Phi | \langle e | U_1^{-1} (p + \sum_k k a_k^\dagger a_k + \sum_k k b_k^\dagger b_k) U_1 | e \rangle | \Phi \rangle = 0. \tag{16}$$

Minimization of the ground-state energy with respect to the variational functions f_k and g_k yields

$$\begin{aligned}
&E(\lambda, a, b, \alpha, \beta, k_o) \\
&= \frac{3}{4} \lambda - \frac{\sqrt{2} \alpha}{\pi} \int_0^{k_o} dk \frac{\exp\left(-\frac{1}{2\lambda} (1-a)^2 k^2\right)}{\left(1 + \frac{1}{2} a^2 k^2\right)} \\
&\quad - \frac{\sqrt{2} \beta}{\pi} \int_0^{k_o} dk \frac{k^2 \exp\left(-\frac{1}{2\lambda} (1-b)^2 k^2\right)}{\left(\frac{1}{k_o} + \frac{1}{2} b^2 k\right)},
\end{aligned} \tag{17}$$

$$f_k = - \frac{\left(\frac{2\sqrt{2} \pi \alpha}{V}\right)^{1/2} \frac{1}{k} \exp\left(-\frac{1}{4\lambda} (1-a)^2 k^2\right)}{\left(1 + \frac{1}{2} a^2 k^2\right)} \tag{18}$$

$$g_k = - \frac{\left(\frac{2\sqrt{2} \pi \beta}{V}\right)^{1/2} \sqrt{k} \exp\left(-\frac{1}{4\lambda} (1-b)^2 k^2\right)}{\left(\frac{k}{k_o} + \frac{1}{2} b^2 k^2\right)} \tag{19}$$

The mean number of the optical phonons in the cloud around the electron is then calculated as

$$\begin{aligned}
&N^o(\lambda, a, b, \alpha, \beta, k_o) \\
&= \langle e | \langle 0 | (U_1 U_2)^{-1} \sum_k a_k^\dagger a_k (U_1 U_2) | 0 \rangle | e \rangle \\
&= \sum_k |f_k|^2 \\
&= \frac{\sqrt{2} \alpha}{\pi} \int_0^{k_o} dk \frac{\exp\left(-\frac{1}{2\lambda} (1-a)^2 k^2\right)}{\left(1 + \frac{1}{2} a^2 k^2\right)^2}
\end{aligned} \tag{20}$$

The mean number of the acoustic phonons in the cloud around the electron is given by

$$\begin{aligned}
& N^A(\lambda, a, b, \alpha, \beta, k_o) \\
&= \langle e | \langle 0 | (U_1 U_2)^{-1} \sum_k b_k^+ b_k (U_1 U_2) | 0 \rangle | e \rangle \\
&= \sum_k |g_k|^2 \\
&= \frac{\sqrt{2}\beta}{\pi} \int_0^{k_o} dk \frac{k \exp(-\frac{1}{2\lambda}(1-b)^2 k^2)}{(\frac{1}{k_o} + \frac{1}{2}b^2 k)^2}
\end{aligned} \tag{21}$$

The spatial localization of the electron is calculated as

$$\begin{aligned}
& r(\lambda, a, b, \alpha, \beta, k_o) \\
&= (\langle e | \langle 0 | (U_1 U_2)^{-1} r^2 (U_1 U_2) | 0 \rangle | e \rangle)^{1/2} \\
&= \left(\frac{3}{2\lambda}\right)^{1/2}
\end{aligned} \tag{22}$$

Not only the ground-state energy but also other physical quantities should be obtained by minimizing equation (17) with respect to the parameters λ , a and b . As their values vary with the electron-phonon coupling constants α and β and with the Debye cut-off k_o they should be calculated numerically.

3. Numerical results and the phase diagram

In this section we calculate numerically the ground-state energy, the mean numbers of the optical and acoustic phonons in the cloud around the electron and the spatial localization of the electron for the wide ranges of the coupling constants α and β . We take $k_o = 20$. The characteristic features of our results will be summarized in the phase diagram. We show the ground-state energies in figure 1, the mean numbers of the optical and acoustic phonons in figure 2 and the spatial localizations of the electron in figure 3 as a function of the coupling constant β for four different values of the coupling constant α : (a) 0.0; (b) 5.3; (c) 7.0; (d) 9.0. First we explain the notations employed. F denotes the nearly free state, S the self-trapped state with the mean number of the optical phonons in the cloud around the electron which is larger than that of the acoustic phonons and L the self-trapped state with the mean number of acoustic phonons in the cloud around the electron that is larger than that of the optical phonons. F_F , E_S and E_L show respectively the ground-state energies for the F, S and L states. N_F^A , N_S^A and N_L^A denote the mean numbers of the acoustic phonons in the cloud around the electron for the F, S and L states. N_F^O , N_S^O and N_L^O show the mean numbers of the optical phonons in the cloud around the electron for the F, S and L states. r_F , r_S and r_L are the spatial localizations of the electron for the F, S and L states, where r_F is infinity. When there is no electron-optical phonon interaction ($\alpha = 0$) the ground-state energy depends on β linearly until the point $\beta_c = 0.0090$ and then it varies with a curve as shown in figure 1 (a). There are two branches. The first branch for small β which corresponds to the F state is

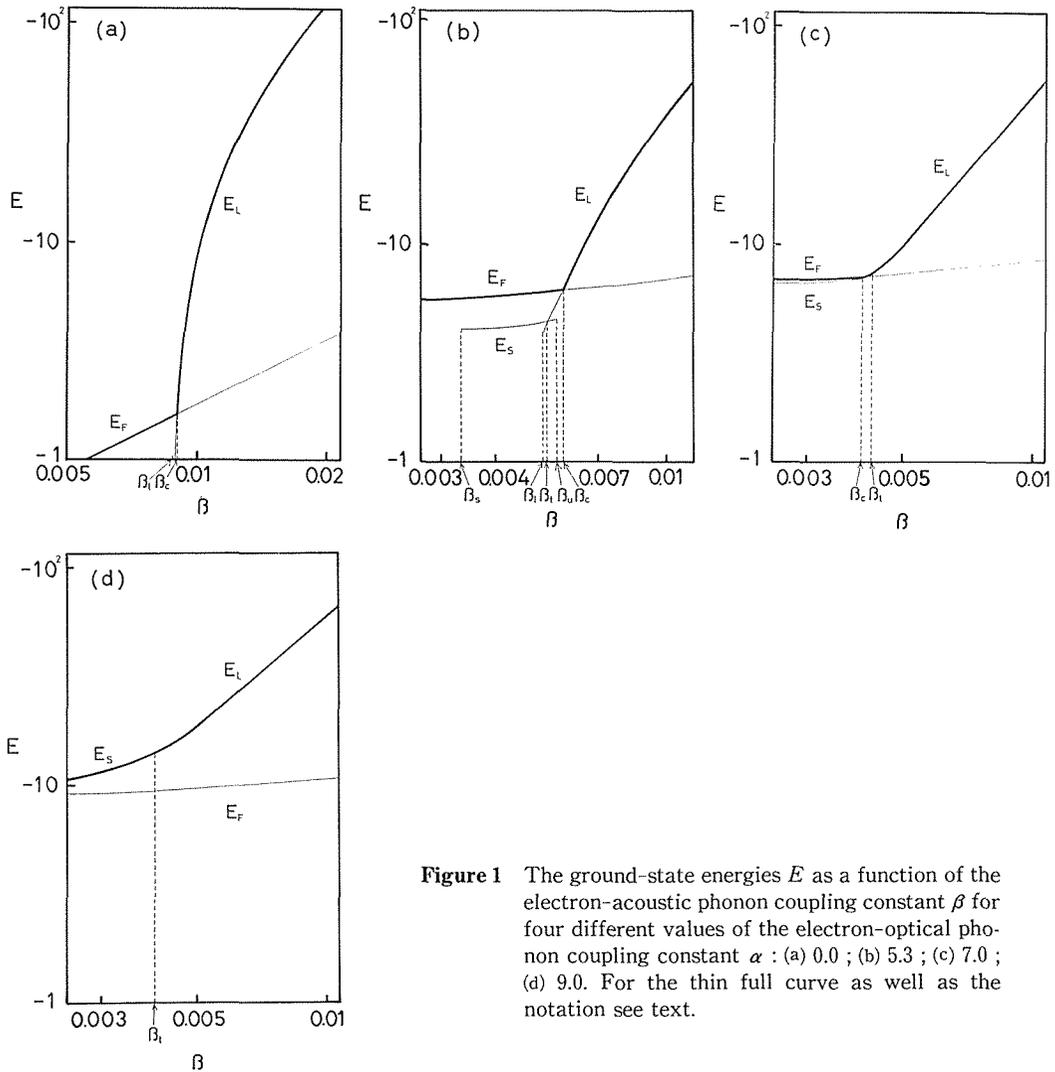


Figure 1 The ground-state energies E as a function of the electron-acoustic phonon coupling constant β for four different values of the electron-optical phonon coupling constant α : (a) 0.0 ; (b) 5.3 ; (c) 7.0 ; (d) 9.0. For the thin full curve as well as the notation see text.

the same as the Lee-Low-Pines approximation and it terminates at the point $\beta_f = 0.41$. The second branch which begins from the point $\beta_l = 0.0088$ corresponds to the strong-coupled L state. The polaron changes its state at the point β_c from a nearly free type to a self-trapping one. For $\beta_l < \beta < \beta_c$ the self-trapped state and for $\beta_c < \beta < \beta_f$ the nearly free state are metastable. The mean number of the acoustic phonons around the electron increases discontinuously at the point β_c as shown in figure 2 (a). On the other hand the spatial localization of the electron which spreads over the crystal for small β ($< \beta_c$) shrinks at the same point (figure 3 (a)). When the electron-optical phonon interaction enters the transition point β_c becomes small with increasing α . $\beta_c = 0.0058$ for $\alpha = 5.3$ and $\beta_c = 0.0040$ for $\alpha = 7.0$. β_c vanishes when α exceeds 7.9. Another branch which corresponds to the strong-coupled S state appears when α exceeds 4.4. For $\alpha = 5.3$ the branch starts at $\beta_s = 0.0034$ and ends at $\beta_u = 0.0056$ (figure 1 (b)). It crosses at $\beta_l = 0.0053$ with the branch corresponding to the L state which starts from $\beta_l = 0.0052$. The mean number of the optical phonons in the cloud around

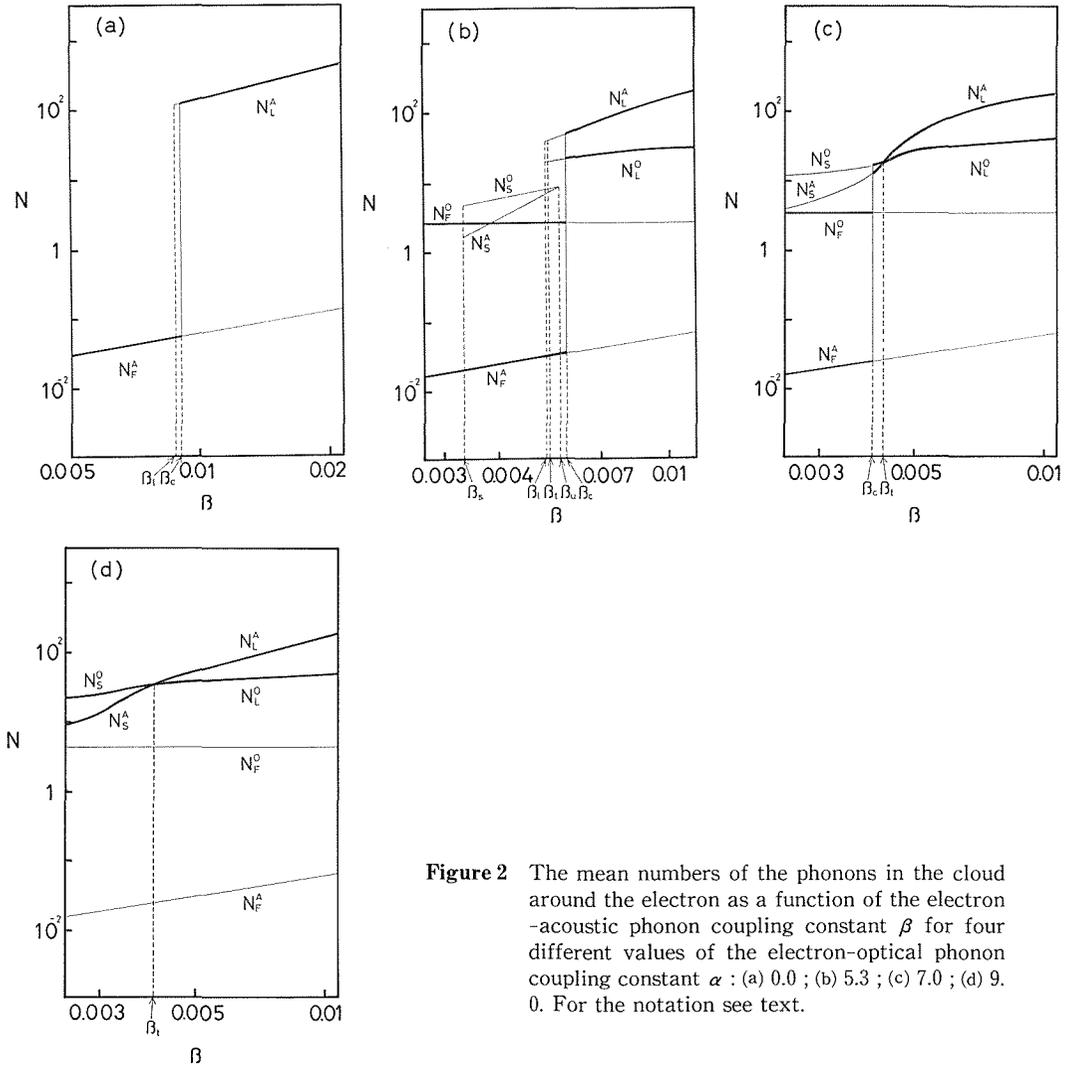


Figure 2 The mean numbers of the phonons in the cloud around the electron as a function of the electron-acoustic phonon coupling constant β for four different values of the electron-optical phonon coupling constant α : (a) 0.0; (b) 5.3; (c) 7.0; (d) 9.0. For the notation see text.

the electron is larger than that of the acoustic phonons for the S state and vice versa for the L state as it should be (figure 2 (b)). For $\beta_s < \beta < \beta_t$ the S state, for $\beta_u < \beta < \beta_c$ the L state and for $\beta_c < \beta < \beta_f$ the F state are metastable. For $\beta_t < \beta < \beta_l$ the S state is the first metastable state and the L state is the second metastable state and vice versa for $\beta_l < \beta < \beta_c$. As α increases the starting point β_s for the branch corresponding to the S state becomes small. β_s vanishes when α exceeds 5.8. For $\alpha = 7.0$ the branch for the F state crosses at $\beta_c = 0.0040$ with that for the S state (figure 1 (c)). The branch for the S state changes gradually at $\beta_t = 0.0043$ into the branch for the L state. The spatial localization of the electron changes smoothly with β around β_t (figure 2 (c)). On the other hand the number of the optical phonons in the cloud around the electron is larger (smaller) than that of the acoustic phonons for $\beta < \beta_t$ ($\beta > \beta_t$) (figure 3 (c)). For $\alpha = 9.0$ the branch for the S state changes smoothly at $\beta_t = 0.040$ to that for the L state (figure 1 (d)). The characteristic features of our results are well summarized in the phase diagram in figure 4 where the polaron state is represented in

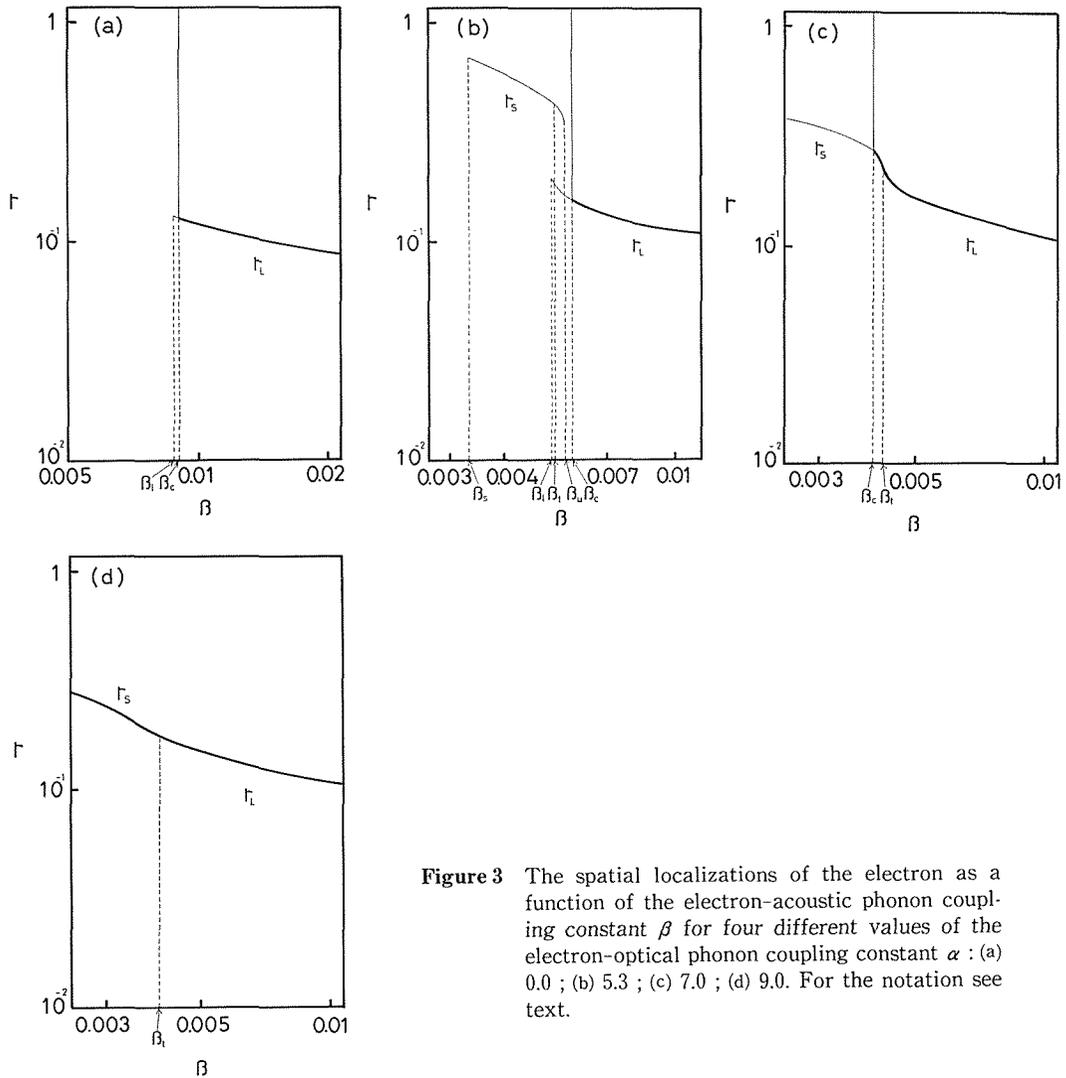


Figure 3 The spatial localizations of the electron as a function of the electron-acoustic phonon coupling constant β for four different values of the electron-optical phonon coupling constant α : (a) 0.0; (b) 5.3; (c) 7.0; (d) 9.0. For the notation see text.

the (α, β) plane. We have several regions: the region F where the F state is the only stable state, the region S where the S state is the only stable state, the region L where the L state is the only stable state, the region F (S) where the F state is stable and the S state is metastable, and vice versa in the region S (F). The region F (L) where the F state is stable and the L state is metastable, and vice versa in the region L (F). The region F (S, L) where the F state is a stable state, the S state is the first metastable state and the L state is the second metastable state. The region F (L, S) where the F state is the stable state, the L state is the first metastable state and the S state is the second metastable state. The phase diagram for the polaron will be useful to understand some physical properties of the polaronic system as we have shown in the case of the bound polaron.¹⁸⁾ In the next section we consider the application of the phase diagram to the real polaronic systems.

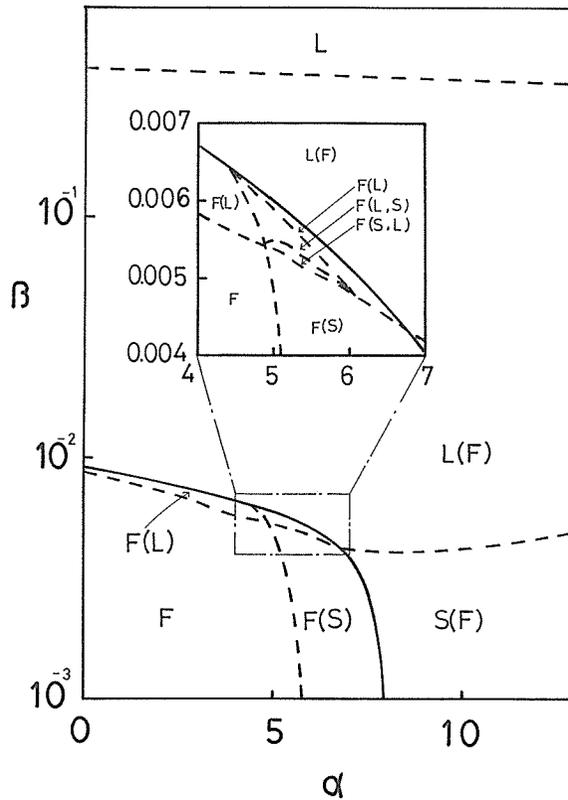


Figure 4 The phase diagram for the polaron. For the notation see text.

4. Phase diagram approach to the real polaronic systems

First we consider polar semiconductors such as GaAs and InSb for which the electron-optical phonon coupling constants α , the electron-acoustic phonon coupling constants β and the Debye cut-off wave numbers k_0 are experimentally determined. We make a list of the values of α , β and k_0 for several polar semiconductors in table 1. Here we have taken the values of α from the list by Kartheuser²⁵⁾ and calculated β through equation (3) using the experimental results on the deformation potential²³⁾ which are also listed in table 1. As the Debye cut-off wave number k_0 as well as the units used are different for each material we must prepare a phase diagram for each case. By comparing the phase diagrams obtained with the experimentally known values of α and β we conclude that all the polarons in these materials are in the F state. This is consistent with the experimental fact that the drift mobility of electrons in the polar semiconductors are rather large. Next we consider ionic crystals such as AgCl and KBr for which so far as we know the electron-acoustic phonon coupling constants β have not yet been determined. As the experimental results on the drift mobility of electrons in such ionic crystals are well explained in terms of the Fröhlich polaron theory²⁴⁾ it is known that the F state for the polarons in these materials are stable.¹⁾ Using this fact we estimate with the help of the phase diagram the upper limits of the electron

Table 1 Parameters k_0 , α , β , the deformation potentials D (eV) and the polaron states determined from the phase diagrams for various polar semiconductors. α and D are taken from the lists by Kartheuser²⁵⁾ and by Neuberger,²³⁾ respectively.

Polar semiconductors	k_0	α	β	D (eV)	state
AlSb	65.3	0.023	8.8×10^{-8}	6.2	F
GaAs	31.2	0.068	2.33×10^{-5}	16.8	F
GaSb	38.0	0.025	2.33×10^{-5}	22.6	F
GaP	12.3	0.201	3.46×10^{-4}	12.7	F
InAs	54.3	0.053	3.0×10^{-9}	— 5.2	F
InSb	72.8	0.022	1.8×10^{-9}	6.4	F
InP	25.8	0.113	4.4×10^{-5}	21	F

Table 2 Parameters k_0 , α , calculated upper limits of β and the deformation potential D (eV) for various ionic crystals. α are taken from the list by Kartheuser.²⁵⁾

Ionic crystals	k_0	α	β	D (eV)
AgCl	18.9	1.94	0.0082	56
AgBr	24.8	1.56	0.0051	54
KBr	14.9	3.05	0.011	38
NaCl	8.41	4.86	0.013	16
ZnO	20.2	0.849	0.0081	96

-acoustic phonon coupling constant β or equivalently of the deformation potential for these materials. The results are represented in table 2 where the values of α are taken from the list by Kartheuser.²⁵⁾

5. Conclusion

We have considered the problem of an electron interacting with acoustic as well as with optical modes of lattice vibrations within the modified variational scheme of the Lee-Low-Pines theory. We calculate the ground-state energy, the mean numbers of the optical and acoustic phonons in the cloud around the electron and the spatial localization of the electron as functions of the electron-optical phonon coupling constant α and the electron-acoustic phonon coupling constant β for the definite Debye cut-off $k_0=20$. All the quantities numerically calculated show phase-transition-like behaviors. Consequently we can make the phase diagram for the polaron. With the help of the phase diagram prepared for each material we show that the polarons in polar semiconductors such as GaAs and InSb are all in the F state and we estimate the upper limits of the deformation potential for ionic crystals such as AgCl and KBr. Although we are in the present work concerned with the relatively simple polaronic systems our phase diagram approach will be useful to understand the various physical behaviors of electrons and holes in ionic crystals and polar semiconductors.

We are now investigating the problem of hole polarons in AgBr and AgCl stimulated with the experimental work by Laredo et al.²⁶⁾

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