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Self-trapping of electrons at the field-effect junction of a molecular crystal

N. Kirova[1,2] and M.-N. Bussac[3]
[1]*POMA, Université d'Angers, France*
[2]*LPTMS, Université Paris-Sud, Orsay, France*
[3]*Ecole Polytechnique, Palaiseau, France*

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We consider the interface of a molecular crystal with a polar dielectrics. Coulomb interaction of free electrons in the molecular crystal with surface polar phonons of the dielectrics can lead to self-trapping of carriers. For typical parameters of molecular field-effect transistors the binding energy is found to be high enough to allow for the formation of a strongly coupled polaron. The effect is further enhanced in the presence of a bias electric field.

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I. INTRODUCTION

During the past few years there has been an increasing number of experimental studies of the transport properties of molecular crystals. The intriguing and still controversial issue is their high field-effect mobilities. The most attention has been devoted to possible application of organic thin films in field-effect transistors (FET's). It is important to notice that the gate materials (SiO_2, Al_2O_3, etc.) are usually the polar dielectrics, which can affect electronic surface states in the molecular crystal. The interaction of an electron in the molecular crystal with surface phonons of the polar dielectrics leads to its self-trapping and formation of the long-range surface polaron. Polaronic effects have been discussed for electrons and excitons in the bulk.[1] But in typical nonpolar molecular crystal only intramolecular on-site polarons can be formed which requires a very strong coupling and, if it takes place, would drastically reduce the carriers mobility.[2] But polarons at the FET junction will have a large radius, keeping a high in-plane mobility. The influence of surface phonons on the electron polaronic state inside the ionic crystal was studied a long time ago[3] and is still being studied.[4] Here we address the opposite situation when the electron resides in the nonpolar molecular crystal. We consider electronic surface states and possible surface polaron formation in the molecular crystal due to the interaction with the surface phonons of the polar dielectrics. According to the value of the coupling constant α, various approaches to polaron formation have been used. In the weak- or intermediate-coupling limit, the method of canonical transformation was introduced by Lee, Low, and Pines.[5] Being exact at $\alpha \to 0$, this method works well only up to intermediate values of α. For the strong-coupling limit, there are two approaches, the adiabatic one developed originally by Pekar[6] and the Feynman path integral method.[7] The Feynman method, though giving reasonably good estimates for the energy of the polaron in the entire range of α, (Ref. 8), is not exact at any α. On the other hand, in the Pekar approach the total energy functional is asymptotically exact for $\alpha \gg 1$. Unfortunately, the solution of the corresponding nonlinear Schrödinger-like equation is not feasible, however. Therefore one needs to use the variational method just for solving for the wave function ψ.

Below we demonstrate the possibility of surface polaron formation based on the Pekar approach. As we shall see, Sec. V, we are in the strong-coupling limit also at low electric field. Moreover, this is also justified as we are in the adiabatic limit, where the kinetic energy of the phonons is much smaller than the bandwidth of the typical molecular crystal.[9] For such a surface polaron we obtain the ground-state wave function ψ, the binding energy W_b, and the effective mass M_p. We study also the squeezing effect of the bias electric field. To demonstrate its strong influence on the properties of the surface polaron we perform numerical studies for parameters of the interface between pentacene and Al_2O_3.

II. GENERAL EQUATIONS

Let the interface be perpendicular to the z axis, the semi-infinite space $z>0$ being occupied by the molecular crystal whereas $z<0$ by the polar dielectrics. The electron is confined within the molecular crystal, its wave function $\psi(\vec{r},z)$ being distributed near the interface, $\vec{r}=(x,y)$. The only dipole-active excitations of the polar dielectrics, to which the electron is coupled, are the surface phonon modes. Following the general scheme of Refs. 6 and 10, the electron-phonon interaction can be divided into two parts. The first one comes from the interaction with high-frequency phonons which is reduced to the classical limit of the image charge potential:

$$H_{i\text{-}ch} = -\frac{e^2}{4\epsilon_\infty^*}\left\langle \psi^*(\vec{r},z)\right|\frac{1}{z}\left|\psi(\vec{r},z)\right\rangle, \quad \frac{1}{\epsilon_\infty^*} = \frac{\epsilon_\infty - \epsilon_1}{\epsilon_1(\epsilon_\infty + \epsilon_1)}. \tag{1}$$

Here ϵ_1 is the dielectric susceptibility of the molecular crystal and ϵ_∞ is the high-frequency dielectric susceptibility of the polar dielectrics. Another part comes from the interaction with low-frequency phonons and it results in polaron formation. For the surface phonons, it can be written as[11]

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$$H_{e\text{-}ph}=-\sum_s\sum_q\frac{e}{\sqrt{q}}\sqrt{\frac{\pi\hbar\omega_s}{S\tilde{\epsilon}^*}}\times\langle\psi^*|e^{-q|z|}(e^{-i\vec{q}\cdot\vec{r}}b_q+e^{i\vec{q}\cdot\vec{r}}b_q^*)|\psi\rangle$$
$$=-\sum_s\sum_q\frac{e}{\sqrt{q}}\sqrt{\frac{\pi\hbar\omega_s}{S\tilde{\epsilon}^*}}\int_0^\infty dz e^{-q|z|}\times\int(e^{-i\vec{q}\cdot\vec{r}}b_q+e^{i\vec{q}\cdot\vec{r}}b_q^*)\rho(\vec{r},z)d\vec{r}. \tag{2}$$

Here $\rho(\vec{r},z)=|\psi(\vec{r},z)|^2$, b_q and b_q^* are the annihilation and creation operators of the surface phonon field, $\vec{q}=(q_x,q_y)$, ω_s are the surface phonon frequencies of the polar dielectrics, S is the surface area, and

$$\frac{1}{\tilde{\epsilon}^*}=\frac{\epsilon_\infty-\epsilon_1}{\epsilon_1(\epsilon_\infty+\epsilon_1)}-\frac{\epsilon_0-\epsilon_1}{\epsilon_1(\epsilon_0+\epsilon_1)}, \tag{3}$$

where $\epsilon_0=\epsilon(\omega\to 0)$ is the low-frequency limit for the dielectric susceptibility of the polar dielectrics.

The total Hamiltonian, in the presence of a bias electric field E perpendicular to the interface, can be written as

$$H=\langle\psi^*|-\frac{\hbar^2}{2m}\Delta|\psi\rangle+H_{i\text{-}ch}+H_{e\text{-}ph}+eE\langle\psi^*|z|\psi\rangle+\sum_{q,s}\hbar\omega_s b_q b_q^*, \tag{4}$$

where m is the band mass of a free electron in the molecular crystal. The last term in Eq. (4) is the surface phonon field energy.

III. SURFACE POLARON

In the adiabatic limit we can treat the phonon field as a classical one. Using the standard variational procedure for the case of the single surface phonon mode, we obtain

$$b_q=-\int\int\frac{e}{\sqrt{q}}\sqrt{\frac{\pi}{S\tilde{\epsilon}^*\hbar\omega_s}}e^{-qz}e^{-i\vec{q}\cdot\vec{r}}\rho(\vec{r},z)d\vec{r}\,dz. \tag{5}$$

Then the total energy can be written as

$$W=\min\{H\}=W_{el}+W_1. \tag{6}$$

Here

$$W_{el}=\langle\psi^*|-\frac{\hbar^2}{2m}\Delta|\psi\rangle-\frac{e^2}{4\epsilon_\infty^*}\left\langle\psi^*\left|\frac{1}{z}\right|\psi\right\rangle+eE\langle\psi^*|z|\psi\rangle \tag{7}$$

and

$$W_1=-\frac{\pi e^2}{S\tilde{\epsilon}^*}\sum_q\frac{1}{q}\int\int e^{-q(z+z')}e^{-i\vec{q}(\vec{r}-\vec{r}')}\times\rho(\vec{r},z)\rho(\vec{r}',z')d\vec{r}\,dz\,d\vec{r}'\,dz'. \tag{8}$$

In the continuum limit W_1 can be written as

$$W_1=-\frac{e^2}{4\pi\tilde{\epsilon}^*}\int\frac{d\vec{q}}{q}\left|\int_0^\infty\rho_q(z)e^{-qz}\right|^2, \tag{9}$$

where

$$\rho_q(z)=\int e^{-i\vec{q}\cdot\vec{r}}\rho(\vec{r},z)d\vec{r}. \tag{10}$$

The energy W_1 can be also written as

$$W_1=-\frac{e^2}{4\pi\epsilon^*}\int\frac{\rho(\vec{r},z)\rho(\vec{r}',z')d\vec{r}d\vec{r}'dz\,dz'}{\sqrt{(z+z')^2+(\vec{r}-\vec{r}')^2}}. \tag{11}$$

We can write the total energy as follows:

$$W_p=W_0\int d^2\vec{r}dz\times\left[-a_B^2\psi^*\Delta\psi-\frac{a_B}{2\epsilon^*}\psi^*\frac{1}{z}\psi-\frac{eE}{W_0}\psi^*z\psi\right]-W_0\frac{a_B}{\tilde{\epsilon}^*}\int dq\left[\int_0^\infty\rho_q(z)e^{-qz}dz\right]^2-W_e, \tag{12}$$

where $W_0=\hbar^2/2ma_B^2$, $a_B=\hbar^2/me^2$ is the effective Bohr radius for the band electron, and W_e is the ground-state energy of the free electron in the given E as it would be without self-trapping[12]:

$$W_e=1.86W_0\left(\frac{eEa_B}{W_0}\right)^{2/3}. \tag{13}$$

A. Ground-state energy

The ground state of the polaron can be studied by variational method. For an isolated electron at the external electric field E perpendicular to the contact interface the wave function should have z-axial symmetry. The trying functions are defined in the half-space $z>0$ with the boundary condition $\psi(\vec{r},0)=0$. Consider first the case $E=0$. The set of tested wave functions with corresponding energies is listed in the Appendix. Among them,

$$\psi=\frac{4}{3b\sqrt{\pi a}}\frac{z}{a}\left(1+\frac{r}{b}\right)\exp\left(-\frac{z}{a}\right)\exp\left(-\frac{r}{b}\right) \tag{14}$$

gives the lowest ground-state energy $W=W_0w$ with

$$w=\frac{1}{a^2}+\frac{1}{3b^2}-\frac{1}{2a\epsilon_\infty^*}-\frac{2}{b\tilde{\epsilon}^*}I\left(\frac{a}{b}\right), \tag{15}$$

$$I(\eta)=\int_0^\infty \frac{dt(1+t^2/6)^2}{(1+t^2)^7(1+\eta t)^6}. \tag{16}$$

Here and below we use $a_B=1$. Let introduce $n=a/b$. Then Eq. (15) can be written as

$$w=\frac{1}{a^2}\left(1+\frac{n^2}{3}\right)-\frac{1}{a}\left(\frac{1}{2\epsilon_\infty^*}+\frac{2n}{\tilde{\epsilon}^*}I(n)\right). \tag{17}$$

Minimizing Eq. (17) over parameters a, n, we arrive at the system of equations for a and a:

$$\frac{1}{a}+\frac{n^2}{3a}-\frac{1}{4\epsilon_\infty^*}-\frac{n}{\tilde{\epsilon}^*}I(n)=0, \tag{18}$$

$$\frac{n}{3a^2}-\frac{2}{a\tilde{\epsilon}^*}I(n)-\frac{2n}{a\tilde{\epsilon}^*}\frac{dI(n)}{dn}=0. \tag{19}$$

The above equations have an obvious solution

$$nI(n)=-\frac{\tilde{\epsilon}^*}{4\epsilon_\infty^*}\,\frac{1}{a}=0,$$

which corresponds to a maximum of w [notice that $nI(n)\geqslant 0$] and describes delocalized electrons with $w=0$. For the localized polaron solution ($a\neq 0$, $b\neq 0$) after some transformation we arrive at a closed equation for n alone,

$$\frac{3}{\tilde{\epsilon}^*}\frac{d}{dn}(nI)+\frac{n^2}{\tilde{\epsilon}^*}\frac{dI}{dn}-\frac{n}{4\epsilon_\infty^*}=0 \tag{20}$$

and the following relation between a and n:

$$a-\frac{3}{\tilde{\epsilon}^* n}\frac{d}{dn}[nI(n)]=0. \tag{21}$$

The numerical solution for the general case $E\neq 0$ is given in Sec. IV.

TABLE I. Dielectric susceptibilities and surface phonon frequency from Ref. 14 used in numerical estimates.

ϵ_1	ϵ_0	ϵ_∞	$\hbar\omega_s$
2.2	3.0	11	0.046 eV

B. Polaron effective mass

The polaron effective mass M_p can be found using standard procedure[13] by taking into account the low-frequency dispersion of $\widetilde{\epsilon^*}$:

$$\frac{1}{\widetilde{\epsilon_0^*}}\Rightarrow\frac{1}{\widetilde{\epsilon_0^*}}\left(1-\frac{1}{\omega_1^2}\frac{\partial^2}{\partial t^2}\right).$$

Then for the polaron moving with a velocity $\mathbf{v}$ the kinetic energy is

$$\delta W=\frac{e^2}{4\pi\widetilde{\epsilon_0^*}}\frac{1}{\omega_1^2}\int\frac{d\mathbf{q}}{q}(\mathbf{qv})^2\left[\int_0^\infty\rho_q(z)e^{-qz}dz\right]^2\equiv\frac{1}{2}M_p\mathbf{v}^2$$

and finally

$$\frac{M_p}{m}=\frac{W_0}{\hbar\omega_s}\frac{2a_B^3}{\widetilde{\epsilon_0^*}}\int_0^\infty q^2dq\left[\int_0^\infty\rho_q(z)e^{-qz}dz\right]^2. \tag{22}$$

IV. NUMERICAL RESULTS: EFFECT OF BIAS ELECTRIC FIELD

For all numerical estimates we have considered the Al_2O_3/pentacene interface. Characteristic values for dielectric susceptibilities [ϵ_1 and $\epsilon(\omega)$ for molecular crystal and polar dielectrics correspondingly] and the surface phonon energy $\hbar\omega_s$ have been taken from Ref. 14 and are listed in Table I.

For $E=0$ we find $n=0.91$; then $a=22.6$ Å and $b=24.5$ Å, and hence the wave function is almost symmetric. The corresponding polaron binding energy is very small, $W_b=0.05$ eV, and the effective mass is naturally only

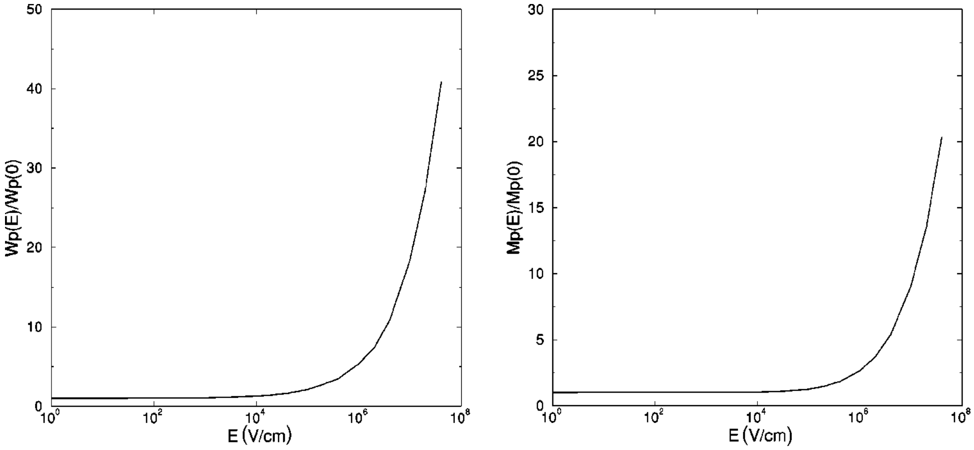

FIG. 1. Binding energy (left) and effective mass (right) for the surface polaron as functions of the applied electric field calculated for the pentacene-Al_2O_3 interface.

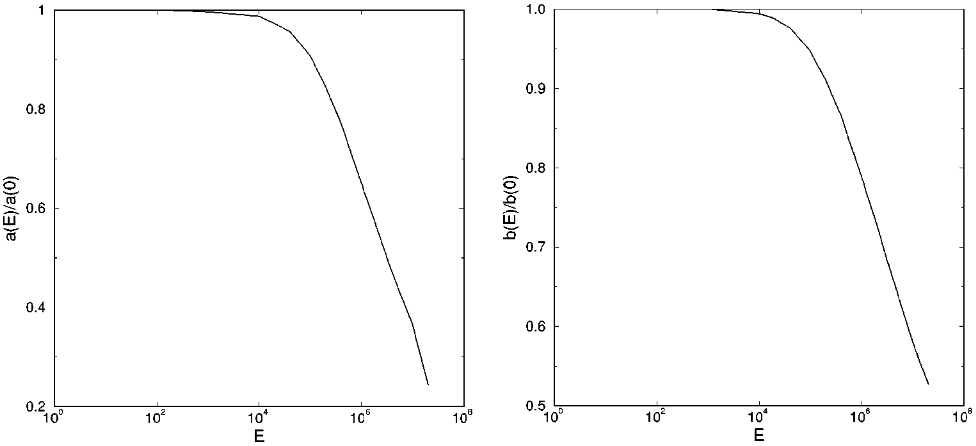

FIG. 2. The characteristic size of the polaron wave function perpendicular (left) and parallel (right) to the interface.

weakly enhanced, $M_p \approx 1.1$ m. In the presence of bias field the polaron binding energy W_b and polaron mass M_p increase rather slowly up to $E = 10^5$ V/cm $[W_p(E = 10^5\ \mathrm{V/cm}) \approx 0.11$ eV, $M_p(E = 10^5\ \mathrm{V/cm}) \approx 1.25 M_p(E = 0)]$ and then there is the strong increase of both. Corresponding numerical results are presented in Fig. 1.

At the same time, being almost unchanged in the plane, parallel to the junction surface, the wave function continues to be squeezed in the perpendicular direction. So the external electric field makes the polaron wave function distribution more two-dimensional. The calculated parameters of the polaron wave function at various bias electric field are presented in Fig. 2. The corresponding form of polaron wave functions without and in the presence of an external electric field are presented in Fig. 3.

V. DISCUSSION, CONCLUSIONS, AND PERSPECTIVES

Above we have exploited the strong-coupling limit of the polaron theory.[6] To verify it, we introduce the dimensionless coupling constant α in analogy with bulk polarons as

$$\alpha = \frac{e^2}{\tilde{\epsilon}^*}\sqrt{\frac{M_p}{2\hbar^3\omega_s}} = \frac{M_p}{m}\frac{1}{\tilde{\epsilon}^*}\sqrt{\frac{W_0}{\hbar\omega_s}}. \tag{23}$$

For $\hbar\omega_s \approx 0.046$ eV we obtain $\alpha = 4.3 M_p/m$. It is commonly accepted that the strong-coupling adiabatic limit for the bulk is valid if $\alpha > 6$. Nevertheless, for the surface polaron the condition is weaker: $\alpha > 1.5$ (Ref. 15). The next important point is that in our case the effective coupling constant strongly depends on the bias electric field via the polaron effective mass (see Fig. 4) and we are in the strong-coupling limit even at zero electric field.

We conclude that the interaction of the electron near the surface of the molecular semiconductor with polarization of the gate dielectric results in the formation of the surface polaron. Their existence will show up in the enhanced effective mass, midgap states, and pseudogap regime in the case of tunneling experiments. The bias electric field, even for its typical values $\sim 10^6$ eV/cm, profoundly stabilizes the polaronic state. At even higher fields a finite concentration of polarons can be achieved which can readily, because of the enhanced mass, can lead to their Wigner crystallization already known in conventional metal-oxide-semiconductor FET (MOSFETs) (Ref. 16 and 17). A more exotic opportunity is that the strong enough polaronic effect can overcome the e-e Coulomb repulsion and lead to the formation of bipolarons. (Remember that without this repulsion taken into account, the energy gain per particle is 4 times higher for the shallow bipolaron in comparison to two polarons). More cal-

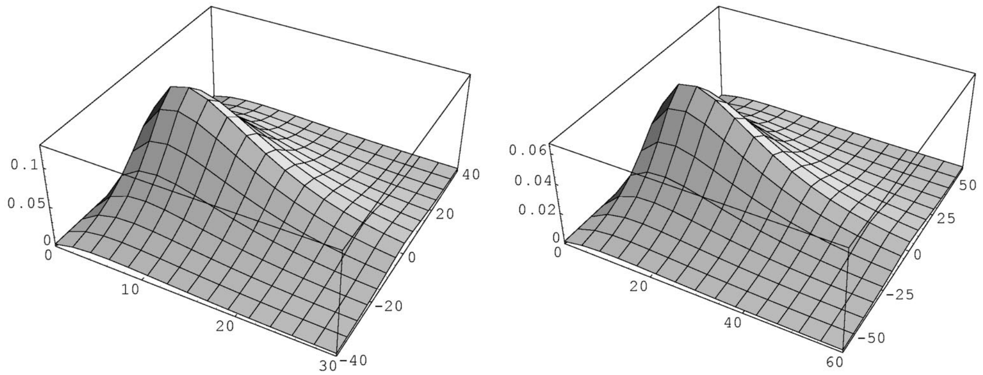

FIG. 3. Electron wave function for $E = 0$ (left) and for $E = 5 \times 10^6$ V/cm (right) calculated from the pentacene-$\mathrm{Al_2O_3}$ interface.

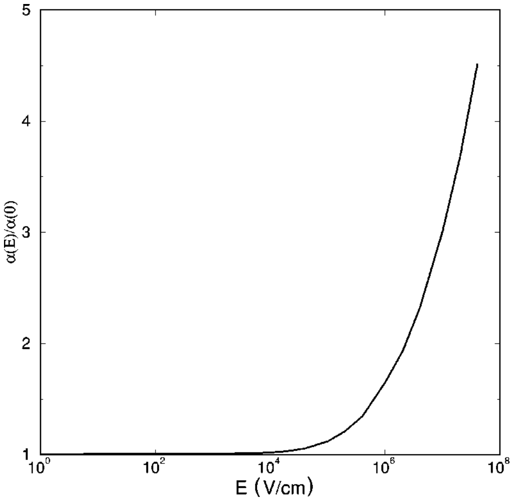

FIG. 4. The effective coupling constant for the surface polaron as a function of the bias electric field.

culations are necessary to check for the stability of bipolarons as has been already done for the one-dimensional case.[18] The appearance of bipolarons in the bulk has been well confirmed for conducting polymers experimentally (see Refs. 19 and 20 and references therein) and motivated theoretically.[21,22] It was intensively considered for oxides.[23–25]. The following scenario would branch between the Wigner crystallization of bipolarons and their Bose condensation.[26] The latter possibilities, leading potentially to FET superconductivity, have been discussed already for polymers[27] and for cuprates[28] and found to be marginal with respect to the necessary strengths of available bias fields. Unfortunately, this goal has been undermined recently by exaggerated reports on corresponding achievements which have not been confirmed. But the real progress by many other groups was very fast and today the really existing fields are only a few times below target values. Moreover, there are less attended experiments[29] where the concentration of carries within the layer of a high-T_c oxide was widely changed by making a junction to a ferroelectric instead of a usual gate material. Turning on and off the superconductivity by applying a bias voltage (greatly amplified by the FE polarization) was clearly confirmed. Interestingly, our calculations can be easily adjusted to this very original case.

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APPENDIX: TRIAL FUNCTIONS AND CORRESPONDING ENERGIES

We have checked the exponential and Gaussian type of wave functions. The wave functions and corresponding ground-state energies are listed in Table II.

For each type of wave functions, values for α, β can be found by minimizing the corresponding energies. We have

TABLE II. Trial wave functions and corresponding energies.

k	Wave functions (ψ_k)	Energies (w_k)	$I_k\left(n=\frac{a}{b}\right)$
1	$\frac{2\sqrt{2}}{ab\sqrt{a\pi}}z^{-z/a}e^{-r/b}$	$\frac{1}{a^2}+\frac{1}{b^2}-\frac{1}{2a\epsilon_\infty^*}-\frac{2}{b\tilde{\epsilon}^*}I_1$	$\int_0^\infty \frac{dt}{(1+t^2)^3(1+nt)^6}$
2	$\frac{4}{3ab\sqrt{a\pi}}z\left(1+\frac{r}{b}\right)e^{-z/a}e^{-r/b}$	$\frac{1}{a^2}+\frac{1}{3b^2}-\frac{1}{2a\epsilon_\infty^*}-\frac{2}{b\tilde{\epsilon}^*}I_2$	$\int_0^\infty \frac{dt(1+t^2/6)^2}{(1+t^2)^7(1+nt)^6}$
3	$\frac{4}{3a^2b\sqrt{3a\pi}}z^2\left(1+\frac{r}{b}\right)e^{-z/a}e^{-r/b}$	$\frac{1}{3a^2}+\frac{1}{3b^2}-\frac{1}{4a\epsilon_\infty^*}-\frac{2}{b\tilde{\epsilon}^*}I_3$	$\int_0^\infty \frac{dt(1+t^2/6)^2}{(1+t^2)^7(1+nt)^{10}}$
4	$\frac{2\sqrt{2}}{ab\sqrt{a\pi}}ze^{-z/a}e^{-r^2/b^2}$	$\frac{1}{a^2}+2\frac{1}{b^2}-\frac{1}{2a\epsilon_\infty^*}-\frac{2}{b\tilde{\epsilon}^*}I_4$	$\int_0^\infty \frac{\exp(-t^2)dt}{(1+nt)^6}$
5	$\frac{2\sqrt{2}}{a^2b\sqrt{3a\pi}}z^2e^{-z/a}e^{-r^2/b^2}$	$\frac{1}{3a^2}+\frac{1}{b^2}-\frac{1}{4a\epsilon_\infty^*}-\frac{2}{b\tilde{\epsilon}^*}I_5$	$\int_0^\infty \frac{\exp(-t^2)dt}{(1+nt)^{10}}$
6	$\frac{4}{ab}\left(\frac{2}{a^2\pi^3}\right)^{1/4}ze^{-z^2/a^2}e^{-r^2/b^2}$	w_6 is always positive	No polaronic state

found that the exponential wave function $k=2$ with a finite first derivative at the interface $\psi'(\vec{r},0)\neq 0$ gives the lowest energy with respect all other types. To check for the stability of our results, we have also considered the wave function

$$\psi=Az(1+A_1z+A_2z^2)(1+B_1r+B_2r^2)e^{-z/a}e^{-r/b}.$$

We obtained indeed that the minimal energy corresponds to $A_1=A_2=B_2=0$, $B_1=1$.

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