Effects of quantum-mechanical corrections to image potentials on the binding energies of surface polarons outside polar crystals

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Binding energies of surface polarons outside polar-crystal surfaces are calculated by considering interactions of electrons outside with both electronic and ionic polarizations of crystals quantum mechanically. Our results show that quantum-mechanical modifications of image potentials due to electronic polarizations reduce polaron binding energies by 40–55 % compared to those calculated with image potentials due to electronic polarization approximated by their electrostatic limits; substitution of infinitely high frequencies for the finite vibrational frequencies of electronic polarizations, however, changes the calculated polaron binding energies by less than 10% for the wide-band materials considered.

I. INTRODUCTION

Surface-polaron states outside polar-crystal surfaces have been extensively studied[1-6] ever since the work of Sak[1] and of Evans and Mills.[2] In most of these works,[1,3,5,6] however, image potentials acting upon electrons due to electronic polarizations—which localize electrons to surfaces and determine dominantly surface-polaron binding energies—are approximated by their classical electrostatic limits. The seemingly reasonable argument for this is that the vibrational frequencies of electronic polarizations are much higher than those of ionic polarizations caused by the relative separations of positive and negative ions when crystals vibrate in optical modes. Recently, a number of papers[7-10] have reported on quantum-mechanical calculations of image potentials, with the conclusions that quantum-mechanical corrections to electrostatic image potentials are not negligible, especially when the electrons are in the vicinities of crystal surfaces. Evans and Mills[2] pointed out that quantum-mechanical nonlocal image potentials will reduce surface-polaron binding energies by a factor of $\frac{16}{25}$ compared with those calculated with electrostatic image potentials, though in their calculation they have neglected electronic polarizations by assuming that the high-frequency dielectric constant $\epsilon_\infty = 1$. In this paper, we report a calculation of binding energies of surface polarons outside polar-crystal surfaces with image potentials due to both electronic and ionic polarizations treated quantum mechanically to study how much the quantum-mechanical corrections to the image potentials due to electronic polarizations will change the polaron binding energies.

The electronic polarization of the crystal considered is approximated by point dipoles vibrating harmonically at a single frequency; that is, we approximate the electronic structures of the atoms in every Winger-Seitz (WS) cell by a two-level system. It is a rather straightforward exercise

to refine the model by considering a many-level system for the electronic structures of the atoms in every WS cell; that is, we consider point dipoles vibrating at several different frequencies if necessary. An electron outside the polar-crystal surface interacts with both the electronic and ionic polarization fields. The calculational technique is mathematically equivalent to that for a polaron near a polyatomic crystal surface with many optical vibrational branches.[11-13]

II. THEORY

The surface-polaron system we considered consists of an electron outside a polar semiconductor which occupies space $z < 0$. The Hamiltonian of surface polarons where electrons couple with ionic polarizations, which have a single branch of vibrational modes, has been derived by many authors.[1,2,14-16] In what follows, we outline briefly the derivation of the Hamiltonian of our system with emphasis on the treatment unique for the multibranch vibrational mode model.

In the Drude-Lorentz model, the point dipoles associated with the atoms in one WS cell due to the electronic transition from ground to excited states, when an electric field is applied to the cell in the two-level system approximation, satisfies the following equation:[17]

$$\ddot{\mathbf{p}}(\mathbf{r},t) = -\omega_1^2 \mathbf{p}(\mathbf{r},t) + e^2 \mathbf{E}_{\text{loc}}(\mathbf{r},t)/m_e , \qquad (1)$$

where ω_1 is the electronic transition frequency between ground and excited states and $\mathbf{E}_{\text{loc}}(\mathbf{r},t)$ is the local electric field acting upon the dipole due to electric dipoles from other cells. By introducing an effective relative displacement of the electron $\mathbf{u}_1(\mathbf{r},t) = \mathbf{p}(\mathbf{r},t)/-e$, Eq. (1) can be rewritten as

$$m_e \ddot{\mathbf{u}}_1(\mathbf{r},t) = -m_e \omega_1^2 \mathbf{u}_1(\mathbf{r},t) - e\mathbf{E}_{\text{loc}}(\mathbf{r},t) . \qquad (2)$$

The relative displacement $\mathbf{u}_2(\mathbf{r},t)$ of the positive and neg-

ative ions in one WS cell satisfies the following equation:[15]

$$\mu \ddot{\mathbf{u}}_2(\mathbf{r},t) = -\mu\omega_2^2 \mathbf{u}_2(\mathbf{r},t) + e^* \mathbf{E}_{\text{loc}}(\mathbf{r},t) , \qquad (3)$$

where we have assumed that the polar semiconductor consists of one pair of positive and negative ions per WS cell. In Eq. (3), μ is the reduced mass of the ion pair, ω_2 is the frequency associated with the short-range part of the ionic interaction, and e^* is the effective charge of the ions. We rewrite Eqs. (2) and (3) as

$$\mu_i \ddot{\mathbf{u}}_i(\mathbf{r},t) = -\mu_1 \omega_i^2 \mathbf{u}_i(\mathbf{r},t) + e^* \mathbf{E}_{\text{loc}}(\mathbf{r},t) , \qquad (4)$$

with the subindex $i = 1,2$ referring to quantities of electronic and ionic polarizations, respectively,

$$\mu_i = \begin{cases} m_e & (i=1) \\ \mu & (i=2) \end{cases} \quad \text{and} \quad e_i^* = \begin{cases} -e & (i=1) \\ e^* & (i=2) \end{cases} . \qquad (5)$$

The total polarization field $\mathbf{P}(\mathbf{r},t)$ is given as the sum of both electronic and ionic polarizations

$$\mathbf{P}(\mathbf{r},t) = \sum_i n e_i^* \mathbf{u}_i(\mathbf{r},t) , \qquad (6)$$

with n the density of WS cells.

Because of the translational invariance along the interface, we can introduce two-dimensional Fourier transformations along the x-y plane and work in $(\mathbf{q},z)$ space, where $\mathbf{q}$ is the two-dimensional in-plane wave vector. The polarization field $\mathbf{P}(\mathbf{r},t)$ can be decoupled in to an s-polarized part and a p-polarized part. Since electrons do not couple to the s-polarized part, we will consider only p polarization in the following analysis. In the unretarded limit, the eigenmodes of the polarization field can be obtained from the classical electrostatics, with the p-polarized eigenmodes determined by the following equations:[15]

$$\chi^{-1}(\omega) P_q(\mathbf{q},z)$$
$$= \tfrac{1}{2} \int_\infty^0 dz' e^{-|z-z'|q}$$
$$\times [-P_q(\mathbf{q},z') - i\,\text{sgn}(z-z') P_z(\mathbf{q},z')] , $$
$$\qquad (7)$$

$$\chi^{-1}(\omega)\epsilon(\omega) P_z(\mathbf{q},z)$$
$$= \tfrac{1}{2} \int_\infty^0 dz' e^{-|z-z'|q} [P_z(\mathbf{q},z') - i\,\text{sgn}(z-z') P_q(\mathbf{q},z')] , $$

where P_q and P_z are the components of the p-polarization field parallel and normal to $\mathbf{q}$, respectively. $\text{sgn}(z)$ is the signum function. $\chi(\omega) = \epsilon(\omega) - 1$ and the dielectric function $\epsilon(\omega)$ is given by

$$\epsilon(\omega) = \frac{(\omega_{L1}^2 - \omega^2)}{(\omega_{T1}^2 - \omega^2)} \frac{(\omega_{L2}^2 - \omega^2)}{(\omega_{T2}^2 - \omega^2)} \qquad (8)$$

with ω_{Li} and ω_{Ti} ($i = 1,2$) determined by

$$(\omega_1^2 - \omega_{Li}^2)(\omega_2^2 - \omega_{Li}^2) + \tfrac{2}{3} \sum_i \frac{(ne_i^*)^2}{n\mu_i}(\omega_i^2 - \omega_{Li}^2) = 0 \qquad (9a)$$

and

$$(\omega_1^2 - \omega_{Ti}^2)(\omega_2^2 - \omega_{Ti}^2) - \tfrac{1}{3} \sum_i \frac{(ne_i^*)^2}{n\mu_i}(\omega_i^2 - \omega_{Ti}^2) = 0 . \qquad (9b)$$

The surface optical (SO) modes are obtained from the equation

$$\epsilon(\omega_{sj}) = -1 \quad (j = 1,2) , \qquad (10)$$

with ω_{s1} and ω_{s2} ($\omega_{s1} \gg \omega_{s2}$) originating from the electronic and ionic polarizations, respectively. The longitudinal optical (LO) modes are given by the equation

$$\epsilon(\omega_{Lj}) = 0 \quad (j = 1,2) . \qquad (11)$$

Substituting Eqs. (10) and (11) into (7), one finds that the space distributions of the eigenmodes of the polarization field are independent of the branch index j. To make the eigenmodes orthogonal, we introduce the eigenfunction S_j with

$$S_1 = \begin{bmatrix} 1 \\ 0 \end{bmatrix} \quad \text{and} \quad S_2 = \begin{bmatrix} 0 \\ 1 \end{bmatrix} . \qquad (12)$$

Similar to the treatment of electronic spins when spin-orbital interaction is neglected, the orthogonal eigenmodes of the p-polarization field are given by

$$\mathbf{P}_{\gamma j}(\mathbf{q},\mathbf{r}) = \mathbf{P}_\gamma(\mathbf{q},z) e^{i\mathbf{q}\cdot\boldsymbol{\rho}} S_j , \qquad (13)$$

where the subindex γ indicates SO and LO modes with the corresponding $\mathbf{P}_\gamma(\mathbf{q},z)$ obtained from Eq. (7) and $\boldsymbol{\rho}$ is the in-plane component of the positional vector of the electron. The polarization field can be expanded with $\mathbf{P}_{\gamma j}(\mathbf{q},\mathbf{r})$,

$$\mathbf{P}(\mathbf{r},t) = \sum_{\gamma,j,\mathbf{q}} P_{\gamma j\mathbf{q}}(t) \mathbf{P}_\gamma(\mathbf{q},z) e^{i\mathbf{q}\cdot\boldsymbol{\rho}} S_j$$
$$= \sum_j \mathbf{P}_j(\mathbf{r},t) S_j , \qquad (14)$$

where $\mathbf{P}_j(\mathbf{r},t)$ is the polarization field arriving from the eigenmodes of branch j and $p_{\gamma j\mathbf{q}}(t) = p_{\gamma j\mathbf{q}}^{(0)} \exp(-i\omega_{\gamma j}t)$, with $p_{\gamma j\mathbf{q}}^{(0)}$ the amplitude of the polarization in mode $(\gamma, j, \mathbf{q})$. The interaction energy between the electron and polarization field is given by the sum of the interaction energies between the electron and polarization fields of the different branches and reads

$$H_I = \int d\mathbf{r}' \nabla_{\mathbf{r}} \frac{\hat{\mathbf{e}} \cdot \mathbf{P}(\mathbf{r}',t)}{|\mathbf{r} - \mathbf{r}'|} , \qquad (15)$$

where $\hat{\mathbf{e}} = eS_0$, $S_0 = \binom{1}{1}$ and the dot in Eq. (15) means the inner product between S_0 and S_j. Quantizing the polarization field, we obtain the Hamiltonian of the surface polaron in the systems we considered,

$$H_{\text{pol}} = -\frac{\hbar^2}{2m_e}\nabla_{\mathbf{r}}^2 + \sum_{j\mathbf{q}} \hbar\omega_{sj} a_{\mathbf{q}j}^\dagger a_{\mathbf{q}j}$$
$$+ \sum_{j\mathbf{q}} [V_{\mathbf{q}j}(z) e^{i\mathbf{q}\cdot\boldsymbol{\rho}} a_{\mathbf{q}j} + \text{H.c.}] , \qquad (16)$$

where m_e is the electron mass outside the surface ($z > 0$), $a_{\mathbf{q}j}^\dagger$ creates a surface vibrational mode in branch j with in-plane wave vector $\mathbf{q}$ and $V_{\mathbf{q}j}(z)$ is the interaction constant between the electron and surface vibrational modes,

$$V_{qj}(z) = -i \left[\frac{4\pi e^2 \hbar}{qA_0 \left. \dfrac{d\epsilon}{d\omega} \right|_{\omega=\omega_{sj}}} \right]^{1/2} \exp(-qz) , \quad (17)$$

with A_0 ($A_0 \rightarrow \infty$) the surface area of the crystal. The electron–LO mode interaction vanishes when the electron is outside the semiconductor surface. The derivation of H_{pol} can be easily extended to cases where electrons interact with polarization fields which have n branches of vibrational modes by extending S_j ($j=0,1,2$) to S_j ($j=0,1,2,\ldots,n$), where

$$S_0^{(n)} = \begin{bmatrix} 1 \\ 1 \\ \vdots \\ 1 \end{bmatrix} , \quad (18a)$$

with 1 in each of the n rows and

$$S_j^{(n)} = \begin{bmatrix} 0 \\ \vdots \\ 1 \\ \vdots \\ 0 \end{bmatrix} \quad (j=1,2,\ldots,n) , \quad (18b)$$

for which the only nonvanishing entry is a 1 in the jth row.

In order to retain the familiar surface-polaron theory, where one considers a single branch of the vibrational modes of the ionic polarization and approximates the electronic polarization by an electrostatic image potential when one lets ω_{L1} and ω_{T1} go to infinite [and so does the frequency of the electronic transition ω_1 in $\epsilon(\omega)$, Eq. (8)], we define the high-frequency dielectric constant of the crystal by

$$\epsilon_\infty = \frac{\omega_{L1}^2}{\omega_{T1}^2} . \quad (19)$$

If we set $\omega = 0$ in Eq. (8), we have

$$\frac{\epsilon_s}{\epsilon_\infty} = \frac{\omega_{L1}^2}{\omega_{T1}^2} , \quad (20)$$

where $\epsilon_s = \epsilon(0)$ is the static dielectric constant of the crystal.

For simplicity, we assume that the bottom of the conduction band of the crystal lies high above the vacuum level, so the crystal surface represents an infinitely high potential barrier. The ansatz of the polaron ground state is taken as

$$\psi = \varphi(z) U_1 U_2 |0\rangle , \quad (21)$$

where $|0\rangle$ is the vacuum state of the surface-polarization fields, $\varphi(z)$ is the trail wave function of the electron distribution perpendicular to the surface, and U_1 and U_2 are the Lee-Low-Pines unitary transformations often used in surface-polaron theory with

$$U_1 = \exp \left[-i \sum_{q,j} \mathbf{q} \cdot \rho a_{qj}^\dagger a_{qj} \right] \quad (22)$$

and

$$U_2 = \exp \left[\sum_{q,j} (a_{qj}^\dagger f_{qj} - a_{qj} f_{qj}^*) \right] . \quad (23)$$

The polaron ground-state energy with the polaron in-plane wave vector $\mathbf{k}_\parallel = 0$ is given by minimizing the expectation value of H_{pol} over ψ (21) with respect to f_{qj},

$$E_g = \left\langle \varphi \left| \left[-\frac{\hbar^2}{2m_e} \nabla_r^2 \right] \right| \varphi \right\rangle - \sum_{q,j} \frac{|\langle \varphi | V_{qj}(z) | \varphi \rangle|^2}{\hbar\omega_{sj} + \hbar^2 q^2 / 2m_e} . \quad (24)$$

If we suppose the electron is localized near $z = z_0$, and take $\varphi(z) = \delta(z - z_0)$ as a consequence, then the potential-energy term in Eq. (24) becomes

$$V_{eff}(z_0) = -\sum_j \alpha_{sj} \hbar\omega_{sj} \int_0^\infty dq \frac{\exp(-2u_{sj} z_0 q)}{1+q^2}$$
$$\rightarrow -\frac{\epsilon_s - 1}{\epsilon_s + 1} \frac{e^2}{4z_0} , \quad (25)$$

with

$$\alpha_{sj} = 4m_e e^2 \left[\hbar^2 u_{sj} \omega_{sj} \left. \frac{d\epsilon}{d\omega} \right|_{\omega=\omega_{sj}} \right]^{-1}$$

the polar coupling constant of the surface mode ω_{sj}. $V_{eff}(z_0)$ approaches the electrostatic image potential [the last limit in Eq. (25)] when the electron is far away from the surface ($z_0 \gg u_{sj}^{-1} = \sqrt{\hbar/2m_e\omega_{sj}}$). When the electron is in the near vicinity of the surface ($z_0 \rightarrow 0$), $V_{eff}(z_0)$ is finite, while the electrostatic image potential goes to infinite. The effective potential associated with the vibrational mode of branch $j = 1$ in Eq. (25) is the image potential due to the electronic polarization. To show this, we set ω_{L1} and $\omega_{T1} \rightarrow \infty$ in Eq. (25), but keep $\omega_{L1}^2 / \omega_{T1}^2 = \epsilon_\infty$, and we have

$$V_{eff,1}(z_0) = -\alpha_{sj} \hbar\omega_{sj} \int_0^\infty dq \frac{\exp(-2u_{sj} z_0 q)}{1+q^2} \Bigg|_{j=1}$$
$$\rightarrow -\frac{\epsilon_\infty - 1}{\epsilon_\infty + 1} \frac{e^2}{4z_0} . \quad (26)$$

When the electron has a spatial distribution perpendicular to the surface, the potential-energy term in Eq. (24) cannot in principle be approximated by a local one-electron image potential. The polaron energy and effective potential of Eq. (24) must be calculated self-consistently. We calculate the polaron ground-state energy E_g [Eq. (24)] variationally utilizing the function

$$\varphi(z) = 2\beta^{3/2} z e^{-\beta z} , \quad (27)$$

where β is the variational parameter. To make comparisons, we also calculate the polaron ground-state energy by approximating the potential energy associated with the vibrational mode of branch $j = 1$ in Eq. (24) by the lo-
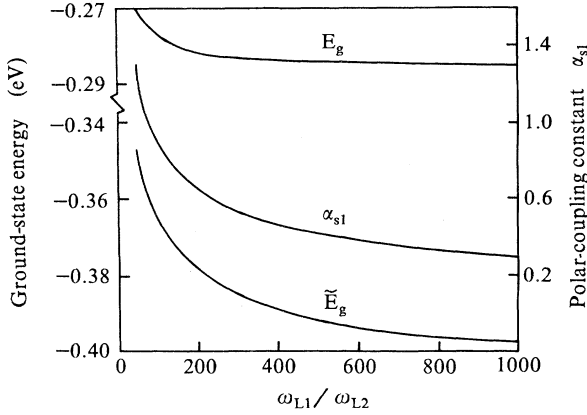
FIG. 1. Ground-state energies of surface polarons outside a polar-crystal surface calculated with the nonlocal image potentials and with the image potential due to the electronic polarization approximated by its electrostatic limit. The calculated ground-state energies are denoted, respectively, as E_g and $\tilde{E}_g$ (left scale). Also given is the polar coupling constant α_{s1} between the electron and surface-polarization field of branch $j=1$ (right scale). The results are given as functions of ω_{L1}/ω_{L2} with other parameters taken as those of a ZnO crystal (see Table I).

cal potential $V_{\text{eff},1}(z)$ [Eq. (26)]. That is, we calculate

$$\tilde{E}_g = \left\langle \varphi \middle| \left[-\frac{\hbar^2}{2m_e}\nabla_r^2 + V_{\text{eff},1}(z) \right] \middle| \varphi \right\rangle$$
$$- \sum_q \frac{|\langle \varphi | V_{qj}(z) | \varphi \rangle|^2}{\hbar\omega_{sj} + \hbar^2 q^2/2m_e} \Bigg|_{j=2} . \quad (28)$$

If we set ω_{L1} and $\omega_{T1} \to \infty$ but keep $\omega_{L1}^2/\omega_{T1}^2 = \epsilon_\infty$, $\tilde{E}_g$ goes to the polaron ground-state energy calculated in the literature[1,3,5,6] with the assumption that the image potential due to the electronic polarization is approximated by its electrostatic limit. The numerical results are given in the next section.

III. RESULTS AND DISCUSSIONS

The ground-state energy E_g of the surface polaron with the electron outside the semiconductor surface calculated with the wave functions given by Eqs. (21) and (27) is

plotted as a function of ω_{L1}/ω_{L2} in Fig. 1 (left scale) with other parameters taken as those of a ZnO surface (see Table I). Also plotted in Fig. 1 (left scale) is the ground-state energy $\tilde{E}_g$ calculated with the image potential due to the electronic polarization approximated by a local potential.

Two points are worth noticing from the numerical results in Fig. 1. (i) The polaron binding energy $(E_b = -E_g)$ calculated with the nonlocal image potentials is much less than that $(\tilde{E}_b = -\tilde{E}_g)$ calculated with an approximated local image potential due to the electronic polarization, even in the limit $\omega_{L1} \to \infty$. (ii) If we assume that $\hbar\omega_{T1}$ is about the same order of magnitude of the crystal band gap, for the ZnO surface we have $\omega_{L1}/\omega_{L2} \cong 100$. From Fig. 1 we see that substitution of an infinitely high frequency for the finite vibrational frequency of the electronic polarization changes the polaron binding energy by less than 5% (see the line of E_g in Fig. 1), while the polaron binding energy is overestimated by 40% for the calculation which approximates the nonlocal image potential due to electronic polarization by its electrostatic limit (compare the lines of E_g and $\tilde{E}_g$ in Fig. 1). In Table I we list polaron binding energies calculated for a number of polar-crystal surfaces (i) with nonlocal image potentials and $\hbar\omega_{T1}$ taken as the crystal band gap, (ii) with nonlocal image potentials and $\hbar\omega_{T1} \to \infty$, and (iii) with image potentials due to electronic polarizations approximated by their electrostatic limits and $\hbar\omega_{T1} \to \infty$. The calculated binding energies are denoted, respectively, for the three cases by E_g, $E_g^{(0)}$, and $\tilde{E}_g^{(0)}$. Comparing the results in the last three columns of Table I, except for InSb which will be discussed later, we find that the substitution of an infinitely high frequency for the finite vibrational frequencies of the electronic polarizations changes the binding energies of the surface polarons by less than 10%, while the approximation of the nonlocal quantum-mechanical electronic potentials due to the electronic polarizations by their electrostatic image potentials overestimates the binding energies of the surface polarons by 40–55%. The effects of quantum-mechanical corrections of image potentials due to electronic polarizations on binding energies of surface polarons outside polar crystals are not negligible.

The calculations also show that if one approximates finite vibrational frequencies of electronic polarizations

TABLE I. Binding energies of surface polarons calculated for a number of polar-crystal surfaces (i) with nonlocal image potentials and $\hbar\omega_{T1}$ taken as the crystal band gap, (ii) with nonlocal image potentials and $\hbar\omega_{T1} \to \infty$, and (iii) with image potentials due to electronic polarizations approximated by their electrostatic limits and $\hbar\omega_{T1} \to \infty$. The calculated binding energies are denoted, respectively, for the three cases by E_g, $E_g^{(0)}$, and $\tilde{E}_g^{(0)}$. The polar coupling constants α_{s1}, between the electrons and surface-polarization fields of branch $j=1$, are calculated with $\hbar\omega_{T1}$ taken as the band gaps of the semiconductors which are taken from Ref. 18. The other experimental parameters used in the calculations are taken from Ref. 19. The energy unit is in eV.

Polar crystal	ϵ_s	ϵ_∞	$\hbar\omega_{T1}$	$\hbar\omega_{L2}$	α_{s1}	E_b	$E_b^{(0)}$	$\tilde{E}_b^{(0)}$
ZnO	8.59	4.0	3.70	0.0720	0.915	0.2769	0.2860	0.4080
CdS	8.58	5.86	2.40	0.0368	1.240	0.2936	0.3096	0.4675
CdTe	10.31	6.90	1.44	0.0212	1.628	0.3081	0.3335	0.5072
GaAs	12.35	10.48	1.35	0.0368	1.694	0.3512	0.3827	0.5927
InSb	17.90	15.70	0.18	0.0239	4.503	0.3234	0.4278	0.6655

by infinitely high frequencies, the effects of nonlocal image potentials on polaron binding energies can be described by introducing a multiplying factor α into electrostatic image potentials due to electronic polarizations; that is, one takes the image potentials due to electronic polarizations in the form

$$V_{\text{eff},1}(z) = -\alpha \frac{\epsilon_\infty - 1}{\epsilon_\infty + 1} \frac{e^2}{4z} \ .$$

For the trial wave function $\varphi(z)$ we used [Eqs. (21) and (27)] α equals $\frac{4}{5}$ and is independent of material parameters.

The Lee-Low-Pines unitary transformation method we used in the present paper applies to cases where the polar coupling constants of electrons α_{sj} are assumed to be less than 2.5.[20] The binding energies of the surface polarons are determined dominantly by the polarizations of branch $j = 1$ for the semiconductors we considered. In Table I we listed polar coupling constants α_{s1} of the semiconductor surfaces calculated with $\hbar\omega_{T1}$ taken as the band gaps

of the semiconductors. Except for InSb, the polar coupling constant α_{s1} of the wide-gap semiconductor is about the order of magnitude of one and decreases as ω_{L1} increases (see Fig. 1), satisfying the condition for the application of the Lee-Low-Pines unitary transformation method. For the narrow-gap semiconductor, such as InSb considered in our paper, the polar coupling constant α_{s1} is much larger than 2.5 (see Table I). The binding energy calculated with Lee-Low-Pines unitary transformation method becomes unreliable. We included the results of InsB in Table I only to indicate that more complicated calculations, which apply to the whole range of coupling strengths, are needed to investigate the problem proposed in our paper. The calculations are now being carried out in our group.

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