Phonon-induced shift in shallow donor levels of semiconductor quantum structures

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Using perturbational, variational, and Feynman-path-integral techniques, we have calculated shifts in impurity binding energies of shallow hydrogenic donor levels in two-dimensional semiconductor systems due to the electron—LO-phonon interaction. We find that polaron shifts in donor energy levels are of the order of 1—10 % in weakly polar $Al_xGa_{1-x}As$-GaAs quantum wells, and 10—20 % in a more polar CdTe-HgTe system. We find Lamb-shift corrections due to the electron-phonon interaction to be negligibly small. Different theoretical techniques and their quantitative implications for experimental measurements are critically discussed.

I. INTRODUCTION

A number of theoretical[1-3] and experimental[4,5] papers in recent literature have discussed the issue of the "hydrogenic" binding of an electron to a donor impurity in a semiconductor quantum well or heterostructure. This problem is the two-dimensional (2D) analog of the well-studied[6] *shallow* impurity-bound state problem in bulk three-dimensional systems. Existing theoretical work has so far considered[7] effects of impurity position in the well, well size, finite potential barrier at the interface, and finite barrier size on the shallow-donor-level energies. Only $Ga_{1-x}Al_xAs$-GaAs-$Ga_{1-x}Al_xAs$ quantum-well structures have been experimentally (and theoretically) studied so far. There is, in general, fairly good agreement[7] between theory and experiment for a GaAs quantum well—the disagreement of about 5% (calculated energies being slightly deeper than the experimentally measured binding energies) can, perhaps, be explained by uncertainties in the precise knowledge of various parameters (e.g., size of the well, position of the dopant atom, band-gap discontinuity) entering the calculation.

In this paper, we report on our calculation of the effect of the electron-phonon interaction on the binding energy of shallow hydrogenic donor levels in two-dimensional semiconductor structures. Since the III-V (or the II-VI) materials used in producing typical quantum-well structures are polar in nature, an electron, weakly bound to a hydrogenic donor impurity in this system, interacts with the LO phonons of the host semiconductor via the well-known long-range polar Fröhlich interaction, and this interaction tends to increase the donor binding energies. Thus the impurity-bound electron is really a 2D Fröhlich polaron, which carries with itself the lattice polarization due to the dipolar interaction between the lattice ions and the electron. It turns out that since GaAs is a weakly polar material (with Fröhlich coupling constant $\alpha \sim 0.07$), the correction to the impurity binding energy, due to the electron—LO-phonon interaction, is small ($< 10\%$) for the well-studied GaAs-$Al_xGa_{1-x}As$ quantum-well structure.

However, the effect we are discussing (namely, the renormalization of the hydrogenic binding energy in a semiconductor by the electron—LO-phonon interaction) is general and, being a lattice effect, is always present in a system. In more polar materials like HgTe-CdTe quantum wells or heterostructures, the correction, due to the Fröhlich interaction, could be of substantive quantitative significance. The corresponding effect has been studied in bulk three-dimensional systems by a number of authors.[8-11] We note that the phonon-interaction effect considered in this paper *increases* the donor binding energy and, nominally, worsens the slight disagreement between theory and experiment in GaAs quantum wells. However, the disagreement is still within the experimental uncertainty in our knowledge of material parameters in GaAs quantum wells. Thus the direct experimental consequence of our theoretical considerations remains unclear at the present time.

II. THEORY AND RESULTS

The basic Hamiltonian for the single conduction-band electron coupled to a Coulombic impurity and interacting with the LO-phonon field can be written within the effective-mass approximation as

$$H = H_0 + H_1 + H_2 , \qquad (1)$$

where

$$H_0 = p^2 - \beta^2/r , \qquad (2)$$

$$H_1 = \sum_k b_k^\dagger b_k , \qquad (3)$$

and

$$H_2 = \sum_k \frac{(2\pi\alpha)^{1/2}}{\sqrt{k}} (b_k^\dagger e^{-i\mathbf{k}\cdot\mathbf{r}} + b_k e^{i\mathbf{k}\cdot\mathbf{r}}) . \qquad (4)$$

H_0 describes the one-electron Hamiltonian for the bound electron with its kinetic energy p^2 and the attractive Coulombic potential term $-\beta^2/r$, where the impurity is

supposed to be at the origin and $\beta^2 = e^2/\epsilon_0$, ϵ_0 being the static background dielectric constant. We neglect any image effect arising from the small difference in background dielectric constants of the two materials (e.g., GaAs and $Al_xGa_{1-x}As$), and take ϵ_0 to correspond to the well material (e.g., GaAs). We choose units such that $2m = \hbar = \omega_{LO} = 1$, where m is the electron band mass (without polaronic renormalization), and ω_{LO} is the LO-phonon frequency. H_1 is the kinetic energy of the LO phonons, which are assumed to be dispersionless Einstein oscillators. H_2 is the long-range Fröhlich interaction between the electron and the LO-phonon field in two dimensions with k as a two-dimensional wave vector. The Hamiltonian, as written in Eqs. (1)–(4), is valid only in the strictly 2D limit for zero quantum-well thickness. The modifications necessary for the actual quasi-2D system will be discussed later in the paper. In our units β^2 is the 2D Rydberg (i.e., the magnitude of the $1s$ ground-state energy of the 2D hydrogen atom). In Eq. (4), the Fröhlich coupling constant α is the effective dimensionless electron-phonon interaction parameter.

No exact solution for the total Hamiltonian defined by Eq. (1) is known. For small α (and the III-V and II-VI systems we are interested in are weak-coupling systems with $\alpha < 1$), one can treat the electron-phonon interaction term H_2 [given by Eq. (4)] as a perturbation to the rest of the Hamiltonian $(H_0 + H_1)$. The ground-state energy shift in the leading-order perturbation theory[12] is given by

$$\Delta E = \sum_n \frac{|\langle n | H_2 | 0 \rangle|^2}{E_0 - E_n - 1} , \qquad (5)$$

where the sum over the intermediate states n must cover all the hydrogenic states (including the continuum) and E_0 is the unperturbed ground-state energy. Substituting Eq. (4) in Eq. (5) we get

$$\Delta E = (2\pi\alpha) \sum_n \sum_k \frac{\langle n | e^{ik\cdot r} | 0 \rangle \langle 0 | e^{-ik\cdot r} | n \rangle}{k(E_0 - E_n - 1)} . \qquad (6)$$

There are a number of ways of evaluating the expression on the right-hand side of Eq. (6). We follow Bajaj[10] and write Eq. (6) approximately as

$$\Delta E \simeq (2\pi\alpha) \sum_{q,k} \frac{|\langle 0 | q \rangle|^2}{k(-k^2 + 2k\cdot q - 1)} , \qquad (7)$$

where $|q\rangle$ is the plane-wave state with wave vector q (i.e., $\langle r | q \rangle = e^{iq\cdot r}$) and $|0\rangle$ is the ground state of the 2D hydrogen atom. The approximation given by Eq. (7) for the energy shift is equivalent to summing over all the plane-wave states q, weighting each term by the probability $|\langle 0 | q \rangle|^2$ of finding the particular plane wave in the unperturbed ground-state wave function $|0\rangle$.

In Fig. 1, we show the calculated ΔE as a function of the unperturbed ground-state energy $E_0 = \beta^2$ (the unit of energy is ω_{LO}) for a strictly 2D GaAs quantum well (for GaAs, $\alpha = 0.07$). Results for other systems are obtained trivially by scaling with the proper Fröhlich coupling constants ($\alpha = 0.03$, 0.07, and 0.4 for InSb, GaAs, and CdTe, respectively).

One can also obtain[8–11] the polaronic shift to the bound-state energy by using a simple effective-mass argu-

ment which gives

$$\Delta E = \beta^2 \left[\frac{m_p}{m} - 1 \right] + \varepsilon_p , \qquad (8)$$

where m_p is the renormalized polaronic mass of the *free* polaron (with m as the bare band mass). In Eq. (8), ε_p is the bound-state energy of a *free* polaron which for a 2D system is given[13] by $\varepsilon_p = \pi\alpha/2$. For 2D systems, it has been shown[13] that $m_p = (1 + \pi\alpha/8)m$, and therefore, within the effective-mass (strictly) 2D approximation, we get

$$\Delta E = \beta^2 \frac{\pi\alpha}{8} + \frac{\pi\alpha}{2} . \qquad (9)$$

The result of Eq. (9) [which is linear in both β^2 and α] is shown in Fig. 1 as the dashed line. The rationale behind the simple effective-mass approximation defined by Eq. (8) is that in the "leading-order" theory, the "bound"-polaron problem is equivalent to a "free" polaron (with mass m_p) bound to a Coulombic impurity center. Thus the effect of the perturbation H_2 on the Hamiltonian H_0 is approximated by changing H_0 to H_0', where H_0' has the bare electron mass m replaced by the polaron mass m_p:

$$H_0' = p^2 \frac{m}{m_p} - \frac{\beta^2}{r} - \varepsilon_p . \qquad (10)$$

This effective-mass approximation [Eq. (8) or (10)] is intuitively expected to work well for small α and for small β^2 (i.e., $\beta^2 \ll \hbar\omega_{LO}$), since in this weak-binding limit one can think of the electron dressed with the phonon cloud to be adiabatically bound to the Coulombic impurity center.[14] We note that the two curves in Fig. 1 are in fact very close to each other for small β^2.

For larger β^2 (with small α), Eq. (7) is not a valid approximation since, for tightly bound electrons, the plane-wave summation becomes increasingly meaningless. Thus both the results depicted in Fig. 1 fail as β^2 becomes larger. However, an exact calculation[11] of Eq. (6) in three dimensions shows that the effective-mass approximation [i.e., Eq. (8)] has a much larger regime of validity than the
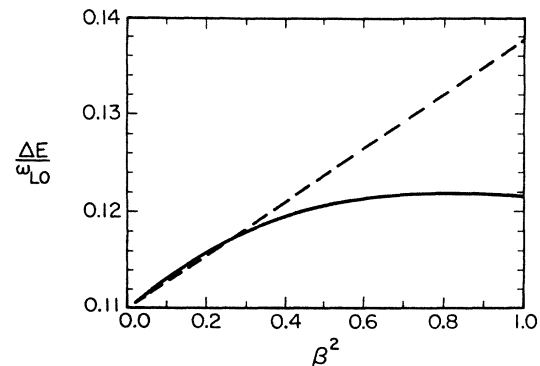

FIG. 1. Ground-state binding-energy shift ΔE shown as a function of the 2D Rydberg β^2 for the perturbation theory (solid line) and the effective-mass approximation (dashed line). We take the Fröhlich coupling constant $\alpha = 0.07$ (GaAs) and the results are for the strict 2D limit (the unit of energy is the LO-phonon energy $\hbar\omega_{LO}$).

approximate perturbative result of Eq. (7). We contend that the same is true[14] in two-dimensional systems (in fact, arguments leading[11] to this conclusion can be shown to be equally valid in two dimensions). Therefore, we take the effective-mass result (the dashed line in Fig. 1) to be the best perturbative result (in the small-α limit) for *all* values of β^2 (except at resonances of energy E_n such that $E_n - E_0$ is a multiple of $\hbar\omega_{LO}$, a situation not considered in this paper). We also conclude from Fig. 1 that the approximate result defined by Eq. (7) is valid for $\beta^2 \leq 0.4$. In actual systems of interest, the polaronic corrections to the ground-state binding energy (according to Fig. 1) are given by $\Delta E/\hbar\omega_{LO}=0.13$ (GaAs), 1.0 (CdTe), and 0.05 (InSb). This translates into a relative shift of the ground-state binding energy $\Delta E/\beta^2$ by 21%, 40%, and 42%, respectively, for GaAs ($\beta^2=0.61$), CdTe ($\beta^2=2.47$), and InSb ($\beta^2=0.12$). These large calculated polaronic shifts of ground-state donor level energies in 2D systems are, however, misleading, since the calculated shift includes the polaronic binding energy, $\varepsilon_p=(\pi/2)\alpha$, which (being the shift in the nominal conduction-band edge) cannot be observed experimentally and, therefore, should be subtracted out from the theoretical result.[15] By doing that, we get an "observable" correction ΔE_p due to electron—LO-phonon interaction in the donor-level ground-state energy of a 2D system given by

$$\Delta E_p \equiv \Delta E - \varepsilon_p = \frac{\pi\alpha}{8}\beta^2 \equiv \left| \frac{m_p - m}{m} \right| \beta^2 = \frac{\Delta m_p}{m}\beta^2 . \quad (11)$$

Calculated values of $\Delta E_p/\beta^2$ for GaAs-, CdTe-, and InSb-based 2D systems are 3%, 16%, and 1%, respectively.

All our discussions so far have been based on strictly 2D systems with zero quantum-well width. Generalization to a finite-width system is easily done within the effective-mass approach [Eq. (11)]. In an earlier paper,[16] we have reported on our calculation of the polaronic effective mass in realistic 2D systems. From Eq. (11), we conclude that $\Delta E_p/\beta^2=\Delta m_p/m$ in the leading-order theory. In Fig. 2 we show our calculated $\Delta m_p/m\alpha$ for a *finite-width quantum-well* system as a function of the well width a (obviously the result,[16] being leading order in α, is valid only for a weak-coupling electron-phonon system). The result interpolates smoothly between the strictly 2D result[13] $\Delta m_p/m\alpha=\pi/8$, and the well-known 3D result $\Delta m_p/m\alpha=\frac{1}{6}$ as the well thickness increases from $a=0$ to large values of a. It should be noted that these calculations assumed an infinite square-well potential which cannot accurately describe very narrow quantum wells. When the subband energy becomes appreciable compared to the real well depths (below about 30 Å), other techniques are needed. From Fig. 2, we can obtain the polaronic shift of the donor-level ground-state energy of a quantum well of arbitrary thickness by using the formula $\Delta E_p/\beta^2=\Delta m_p/m =(\Delta m_p/am)\alpha$. For example, in a GaAs quantum-well system with $a=100$ Å, we get a polaronic shift of 1.6%, which is much smaller than the strictly 2D result of about 3%. Similarly, in a CdTe quantum well with $a=100$ Å, we get $\Delta E_p/\beta^2\approx10\%$. Figure 2 is the most important result of our paper because
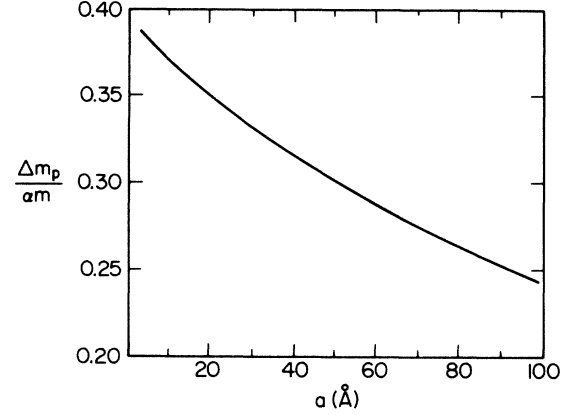

FIG. 2. The polaronic effective-mass correction Δm_p in units of am (where m is the band mass) is shown as a function of the quantum-well width (a). The result gives directly the effective binding-energy shift $\Delta E_p/\beta^2=(\Delta m_p/am)\alpha$ [see Eq. (11)] due to the electron—LO-phonon interaction.

it enables us to obtain directly the polaronic correction to binding energies in various realistic quantum-well structures without actually worrying about the details of the impurity-bound state calculation in the finite-size quantum wells. Our results should be approximately valid for all the known III-V and II-VI quasi-2D structures.

Since our systems of interest are weak-coupling ($\alpha < 1$) polar semiconductors, the above results based on perturbation theory suffice for our purpose. For larger α (>1), however, perturbation theory is not expected to be very good and we have carried out variational[9] and Feynman-path-integral[8] calculations of the binding-energy shift. The variational calculations are expected to be well valid for larger β^2 (as well as for larger α) where the electron is tightly bound to the impurity. The Feynman-path-integral calculation is expected to be fairly well valid for all α. In Fig. 3, we show our calculated shifts $\Delta E/\omega_{LO}$ to the ground-state donor-level energy as a function of the
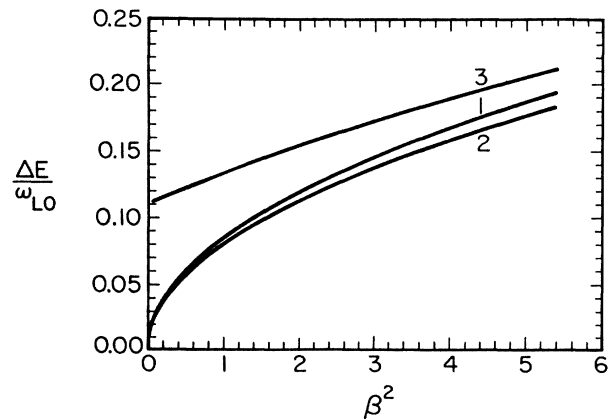

FIG. 3. Ground-state binding-energy shift ΔE shown as a function of 2D Rydberg β^2 (for $\alpha=0.07$) for three different approximations: (1) Coulombic variational calculation; (2) Gaussian variational calculation; (3) Feynman-path-integral calculation. The unit of energy is the LO-phonon energy.

unperturbed binding energy β^2 in a strictly 2D system due to three different theories: Curves 1 and 2 are both variational results calculated with Coulombic (exponential) and Gaussian variational wave functions, respectively, whereas curve 3 is the result of a Feynman calculation. All the results in Fig. 3 are for a fixed α (=0.07) corresponding to GaAs heterostructures. The variational calculations are standard[9] and use a linearly shifted phonon wave function,

$$\phi_L = \exp\left[\sum_{\mathbf{k}} F_{\mathbf{k}}(b_{\mathbf{k}}^\dagger - b_{\mathbf{k}})\right] |0\rangle ,$$

and a Coulombic (curve 1) or a Gaussian (curve 2) electronic wave function. The total energy is then minimized variationally and the calculation for the bound polaron problem follows rather closely our earlier work on the free 2D polaron.[17] In this case,

$$\Delta E = \langle \Psi | H - H_0 | \Psi \rangle , \tag{12}$$

where $|\Psi\rangle$ is the total ground-state wave function of the system. This approximation method is found to be better for the bound-polaron problem than the free-polaron[17] case for small α since the binding makes the localized wave function $|\Psi\rangle$ a good approximation for the electronic wave function. For $\beta^2 \to \infty$, curve 1, based on a Coulombic wave function, gives the best result, whereas the free-polaron limit ($\beta^2 \to 0$) is not correctly reproduced in either of the variational theories, since these are based on bound-state wave functions, whereas in the $\beta^2 \to 0$ limit the polaron is free. We should point out that the variational parameter in these calculations is β^{-1} which is basically the effective 2D-Bohr radius in polaron length units. Also, the Gaussian variational calculation is better for larger α (and, smaller β), whereas the Coulombic calculation is better for larger β (and smaller α). In the limit of small α and β, both the variational calculations fail since in that limit ($\alpha, \beta \to 0$), the polaron is basically free.

The path-integral calculation (curve 3 in Fig. 3) follows standard techniques and smoothly interpolates between the weak-coupling perturbative results at small β and the strong-coupling variational results at large β. A major problem with the path-integral result (and this applies to the Gaussian variational result as well) is that the $\alpha \to 0$ (or, equivalently, $\beta^2 \to \infty$) limit is wrong since it is based on a 2D Gaussian hydrogen atom with ground-state energy $E_G = \pi\beta^2/4$, rather than β^2, which is the exact result. Thus the binding energy based on the Feynman (or, the Gaussian variational calculation) is wrong by 20% in the weak-coupling ($\alpha \to 0$) limit. This problem can be fixed by using a Coulombic Feynman-path-integral formalism. We are, however, interested in the binding-energy *shift* due to the polar interaction and, therefore, the results presented here for ΔE measure the shift by defining $\Delta E = E(\alpha) - E(\alpha = 0)$, with $E(\alpha)$ and $E(0)$ calculated *within the same theory*. Thus the shifts ΔE for curves 2 and 3 in Fig. 3 are measured with respect to the Gaussian approximation to the ground state of the 2D hydrogen atom. A comparison of the path integral results with the perturbative results for *small* α and β^2 shows that the values of ΔE defined this way are quite good. In Fig. 4
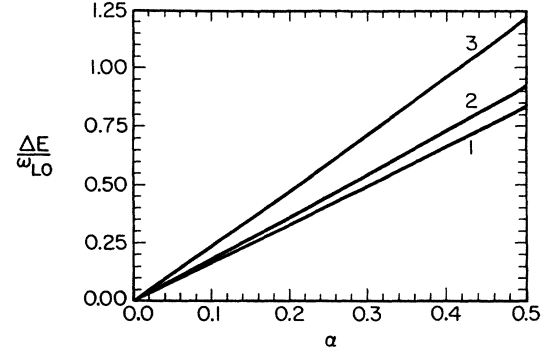

FIG. 4. Ground-state binding-energy shift ΔE shown as a function of the Fröhlich coupling constant α using the Feynman-path-integral technique for (1) $\beta^2 = 2.47$ (HgTe-CdTe), (2) $\beta^2 = 0.61$ (GaAs), (3) $\beta^2 = 0.12$ (InSb). The unit of energy is the LO-phonon energy.

we show the binding-energy shift ΔE as a function of the Fröhlich coupling constant α for three different values of the unperturbed binding energy $\beta^2 = 2.47$ (CdTe), 0.61 (GaAs); and 0.12 (InSb). Results are based on the Feynman-path-integral calculation and, for these values of α and β^2, perturbative results of Fig. 1 are almost the same. As stated earlier, we get polaron shifts $\Delta E/\beta^2$ of the order of 20% for GaAs ($\alpha = 0.07$) to 39% and 42% for CdTe ($\alpha = 0.4$) and InSb ($\alpha = 0.03$), respectively. When corrected for the shift in the band edge of the free polaron (as explained earlier), we again get 3% and 16% observable shifts in the ground-state energies of 2D hydrogen atoms in GaAs and CdTe, respectively.

So far we have only discussed the ground-state energy shift due to the Fröhlich interaction. One expects some shift in the excited-state energies as well, and, consequently, transition energies will be modified due to the electron-phonon interaction. Since transition energies can be directly measured spectroscopically,[18] it is of some interest to obtain the polaron correction to the unperturbed energy-level *differences*. We show these results in Fig. 5. Using the perturbation theory described in the beginning of this paper, we have carried out a calculation of the polaron corrections to the $2s$ and $2p$ levels of the 2D hydrogen atom in addition to the $1s$ level that we have described above (Figs. 1—4) in detail.

In Fig. 5, we show our calculated *corrections* to the low-lying excitation energies for donor levels $2s$ and $2p$, compared with the ground $1s$ level of a strictly 2D hydrogen atom. The solid line shows the polaronic correction to the s-level transition energy, whereas the dashed line shows the corresponding p-level result. We calculate E by perturbation theory where

$$E = [E_{2s}(\alpha,\beta) - E_{1s}(\alpha,\beta)]$$

$$\quad - [E_{2s}(\alpha = 0,\beta) - E_{1s}(\alpha = 0,\beta)]$$

(solid curve in Fig. 5), or (13)

$$E = [E_{2p}(\alpha,\beta) - E_{1s}(\alpha,\beta)]$$

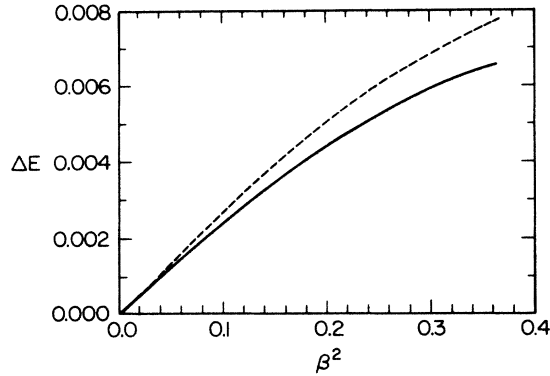$$\quad - [E_{2p}(\alpha = 0,\beta) - E_{1s}(\alpha = 0,\beta)]$$

FIG. 5. Phonon-induced shifts in the low-lying donor-level excitation energies are shown as a function of the 2D Rydberg β^2 (for $\alpha = 0.07$). Results for the s transition $(E_{2s}-E_{1s})$ and the p transition $(E_{2p}-E_{1s})$ are shown, respectively, by the solid and the dashed lines within the perturbation-theoretic calculation. The unit of energy is the LO-phonon energy. The difference between the solid and the dashed line in the figure is a measure of the phonon-induced Lamb shift in a two-dimensional system.
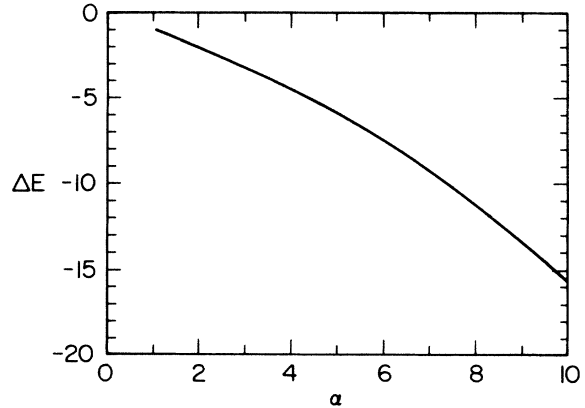

FIG. 6. Ground-state binding energy of a bound 2D polaron is shown as a function of the electron-phonon coupling α for a fixed value of the unperturbed Rydberg $(\beta^2 = 1)$. The calculation utilizes the Feynman-path-integral technique. The energy unit is the LO-phonon energy.

(dashed curve in Fig. 5). We emphasize that the perturbative results presented in Fig. 5 can be trusted only for $\beta^2 \lesssim 0.4$ (as was discussed earlier). We should point out that the unperturbed result for the excitation energy between the 1s and 2s (or 2p) level is $8\beta^2/9$ in our units.

Simple effective-mass arguments give

$$E = E_2 - E_1 = (\tfrac{8}{9}\beta^2)\frac{m_p}{m} , \tag{14}$$

and this result is *independent* of the azimuthal quantum number (i.e., the s and the p levels are shifted equally within the effective-mass theory). In the strictly 2D limit $m_p/m = \pi\alpha/8$, and hence $E_2 - E_1 = \tfrac{8}{9}\beta^2(\pi\alpha)/8$, which is approximately consistent with the results presented in Fig. 5. For a finite-size quasi-2D system, we conclude from Fig. 2 and Eq. (14) that polaronic corrections to transition energies will be of the order of 1—3 % in the GaAs system and 10—20 % in the CdTe system.

One very important aspect of Fig. 5 is that the small difference between the solid and the dashed lines is a measure of the phonon-induced Lamb shift in the 2D hydrogen atom. The accidental degeneracy between s and p levels in the unperturbed situation is lifted by the electron-phonon interaction, and the splitting between the two curves in Fig. 5 is a measure of this effect. This is the solid-state analog of the well-known Lamb shift effect in quantum electrodynamics. We conclude from Fig. 5 that Lamb shift for realistic systems is extremely small (less than 1% of the binding energies). We also point out that the effective-mass theory, while predicting fairly accurate values for the ground-state energy shift (Fig. 1), fails to give any Lamb shift (which is a higher-order effect) at all.

Finally, within the Feynman-path-integral technique, we investigate whether a 2D bound polaron exhibits a sharp self-trapping transition[17] as a function of the cou-

pling constant α. In Fig. 6 we show the calculated ground-state binding energy of a 2D *bound* polaron as a function of α and for a fixed value of $\beta = 1$. Since the energy is a smooth function of α (without any cusp), we conclude that there is *no* self-trapping transition[19] for the long-range polar interaction in a 2D impurity-bound electron. This is in agreement with our earlier work.[19]

III. CONCLUSION

In conclusion, we have calculated the electron—LO-phonon-interaction-induced corrections to the donor-level binding energies in two-dimensional semiconductor quantum wells. We find that the electron-phonon interaction increases the binding energy and the calculated shift in GaAs heterostructure is only about 1—3 % (depending on the quantum-well width). This is not yet an experimentally observable effect, since some of the experimental parameters are not known to better than 5%. On the other hand, in more polar materials (e.g., CdTe) the correction could be of the order of 10—20 %, which should be experimentally significant once experimental results on more polar heterostructures are available. We have also calculated the Lamb shifts due to electron-phonon interaction in these systems and find it to be negligibly small (less than 1% of the binding energies). Within a Feynman-path-integral calculation, we have also investigated the possibility of a sharp self-trapping transition in a bound 2D Fröhlich polaron. Our conclusion is that there is no such self-trapping transition for the long-range polar interaction.

ACKNOWLEDGMENTS

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[15]The dispersion of a free-conduction-band electron in the presence of polaron effects is given by $E_c(\mathbf{k}) = -\varepsilon_p + k^2/2m_p$ and thus the nominal band edge $E_c(k=0)$ shifts by ε_p in the presence of electron—LO-phonon interaction.

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