Strong-coupling theory of quasi-two-dimensional polarons

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A strong-coupling theory of the polaron is developed to study the electron-phonon interaction in quantum-well structures consisting of strong polar crystals. By introducing a two-parameter trial wave function to take care of the strong anisotropy, the Landau-Pekar theory is extended to investigate polaronic effects in a quantum well. It is shown that the theory yields correct results in both the two-dimensional and three-dimensional limits. The ground-state properties of quasi-two-dimensional polarons are calculated as functions of the well width for two choices of the coupling constant α.

I. INTRODUCTION

There has been a great deal of interest in the electronic properties of polar crystals of reduced dimensionality such as quantum wells and heterostructures. In particular, the interaction between electrons and the lattice vibration in such microsystems has been studied extensively, because the polaronic effects can strongly influence the optical and transport properties of the materials under consideration. Earlier works[1-4] discussed mainly the polaron properties near the surface. In the studies of interface polaronic effects, electron interaction with bulk longitudinal-optical (LO) phonon modes alone were first considered,[5-7] and the contribution of surface-optical (SO) phonon modes was also included later.[8,9]

More recently, polaronic properties in a quantum well have also received much attention[10-17] though the interface phonon effects may or may not be included in the calculations. In all these works, however, the weak-coupling limit of the electron-phonon interaction is assumed. This is all right for materials such as III-V compounds in which the coupling constant of the electron-phonon interaction as defined in the Fröhlich Hamiltonian is generally small.

Because of the rapid development of technology, the growth of heterostructures based on II-VI compounds by molecular-beam epitaxy has become more practical now. These new materials are increasingly attracting attention for their potential applications in optoelectronic devices. Other strongly polar materials such as ionic crystals that may also be of interest include thin films of alkali halides and Cu_2O. Since these crystals are generally more polar than III-V compounds, the electron-phonon coupling may become so strong that the weak-coupling scheme does not apply. Therefore it is of interest to study the strong-coupling theory of polarons in a quantum well consisting of materials of strongly polar crystals.

The strong coupling theory of polarons in the bulk was developed by Landau and Pekar[18] in the 1940s. An alternative approach was discussed by Feynman.[19] Both of these theories have been applied in recent years[20,21] to investigate strictly two-dimensional (2D) polarons as an approximate description of the quasi-two-dimensional (Q2D) case. The strong-coupling polaron confined in a harmonic-oscillator potential has also been discussed.[22]

In this paper, we study the ground-state properties of a strong-coupling polaron in an infinite quantum well of arbitrary width. Only the bulk LO phonon modes are considered for simplicity. From what we have learned in the case of weak-coupling free polarons in a slab of crystal,[11] the contribution of SO phonon modes is expected to be small and to become significant only for small widths. In more realistic quantum-well structures of III-V compounds, it is also known that the interface phonon modes can be ignored unless there exists an external magnetic field, which greatly enhances the interface phonon influence.[23]

In Sec. II, we choose the trial wave function with two parameters to be determined by the variational principle. The energy levels of the confined polaron are calculated in Sec. III as functions of the well width and the effective mass corresponding to each level is also found. In Sec. IV, we first show that in the limits of small and large well widths, our results approach those of 2D and 3D polarons, respectively. Numerical results for two choices of the coupling constant α are then plotted and discussed for a wide range of the well width.

II. THE TRIAL WAVE FUNCTION

We choose the geometry such that the xy plane is parallel to the interfaces and the origin is at the center of the well. The Hamiltonian for a system of a confined electron interacting with LO phonon modes in a quantum well of width L is, in the usual polaron unit $2m=\hbar=\omega_0=1$,

$$H=p^2+V(z)+\sum_{\mathbf{k}} a_{\mathbf{k}}^{\dagger}a_{\mathbf{k}}+\sum_{\mathbf{k}}[V_k^{*}a_{\mathbf{k}}^{\dagger}e^{-i\mathbf{k}\cdot\mathbf{r}}+V_k a_{\mathbf{k}}e^{i\mathbf{k}\cdot\mathbf{r}}] , \tag{1}$$

with the potential of confinement

$$V(z)=\begin{cases}0, & -\frac{1}{2}L\le z\le\frac{1}{2}L ,\\ \infty, & \text{otherwise} ,\end{cases} \tag{2a}$$

and the electron-phonon interaction energy

$$V_k=(4\pi a/Vk^2)^{1/2} \tag{2b}$$

in which we have defined the coupling constant with standard notation

$$\alpha=\frac{e^2}{2\hbar\omega_0}\left[\frac{2m\omega_0}{\hbar}\right]^{1/2}\left[\frac{1}{\varepsilon_\infty}-\frac{1}{\varepsilon_0}\right] \tag{2c}$$

and the crystal volume is V. Here, and throughout this paper, we adopt $\sqrt{\hbar/2m\omega_0}$ and $\hbar\omega_0$ as units for length and energy, respectively. The first two terms in (1) represent the Hamiltonian of the confined electron whose momentum and position are given by $\mathbf{p}=(p_x,p_y,p_z)$ and $\mathbf{r}=(\boldsymbol{\rho},z)=(x,y,z)$, respectively. The third term is the free phonon-field energy, in which the operator $a_{\mathbf{k}}^{\dagger}$ ($a_{\mathbf{k}}$) creates (annihilates) a phonon mode characterized by the wave vector $\mathbf{k}=(\boldsymbol{\kappa},q)$ and frequency ω_0. It should be noted that we have assumed dispersionless phonon modes and an infinite well just for simplicity.

Following Landau and Pekar,[18] we write the trial wave function

$$|\Psi\rangle=\Phi(\mathbf{r})U|0\rangle , \tag{3}$$

where $\Phi(\mathbf{r})$ depends only on the electron coordinates. The phonon vacuum state $|0\rangle$ is defined by $a_{\mathbf{k}}|0\rangle=0$, and the transformation U is a unitary displacement operator given by

$$U=\exp[\sum_{\mathbf{k}}(f_k a_{\mathbf{k}}^{\dagger}-f_k^{*}a_{\mathbf{k}})] , \tag{4}$$

where f_k is the variational function. The electron trial wave function is chosen as

$$\Phi(\mathbf{r})=\frac{\lambda}{\sqrt{\pi}}\varphi(z)e^{-\lambda^2\rho^2/2}\exp(i\mathbf{p}_0\cdot\mathbf{r}) , \tag{5}$$

with

$$\varphi(z)=\begin{cases}Ne^{-\mu^2z^2/2}\sin\left[l\pi\left[\dfrac{z}{L}+\dfrac{1}{2}\right]\right], & \\ \qquad l=1,2,3,\ldots, \quad |z|\le L/2 ,\\ 0, \quad |z|>L/2 ,\end{cases} \tag{6}$$

where λ and μ are the variational parameters introduced to reflect the anisotropic nature of the system. We note that one variational parameter is sufficient for the isotropic case of an n-dimensional problem where n is any integer.[24] Since the translational symmetry in the z direction is destroyed in a quantum-well structure, it is natural that the strong electron-phonon coupling in the z direction is different from that in the xy plane. Furthermore, a variational vector $\mathbf{p}_0=(p_{ox},p_{0y},p_{0z})$ has also been introduced in order to calculate the polaron effective mass. The normalization constant is given by

$$N=1/\{2L\int_0^{1/2}e^{-(\mu Lt)^2}\sin^2[l\pi(t+\tfrac{1}{2})]dt\}^{1/2} , \tag{7}$$

where we have made the variable change $z=Lt$ for convenience.

III. ENERGY LEVELS AND EFFECTIVE MASS

In order that the polaron energy and effective mass can be determined at the same time, we minimize the expectation value of the quantity $J=[H-\mathbf{u}\cdot(\mathbf{P}-\mathbf{P}_0)]$ in the state $|\Psi\rangle$. Thus, we have the variational problem

$$\delta J=\delta[\langle\Psi|H-\mathbf{u}\cdot(\mathbf{P}-\mathbf{P}_0)|\Psi\rangle]=0 , \tag{8}$$

where we have introduced the total momentum for the system

$$\mathbf{P}=\mathbf{p}+\sum_{\mathbf{k}}\mathbf{k}a_{\mathbf{k}}^{\dagger}a_{\mathbf{k}} \tag{9}$$

and its expectation value $\mathbf{P}_0$ in the state $|\Psi\rangle$, as we shall see later. The vector $\mathbf{u}$ plays the role of the Lagrange multiplier in Eq. (8), and can be identified as the polaron velocity, as it turns out. The unitary operator u transforms the phonon operators as follows.

$$U^{\dagger}a_{\mathbf{k}}^{\dagger}U=a_{\mathbf{k}}^{\dagger}+f_{\mathbf{k}}^{*} , \tag{10a}$$

$$U^{\dagger}a_{\mathbf{k}}U=a_{\mathbf{k}}+f_k . \tag{10b}$$

Inserting Eqs. (3), (4), and (9) in (8), we find the expectation value

$$\begin{aligned}J=\lambda^2+\mu^2[1-g_l(\mu)]+p_0^2+&\left[\frac{l\pi}{L}\right]^2\\&+\sum_{\mathbf{k}}(1-\mathbf{u}\cdot\mathbf{k})|f_{\mathbf{k}}|^2\\&+\sum_{\mathbf{k}}[V_{\mathbf{k}}^{*}f_k^{*}\langle\Phi(\mathbf{r})|e^{-i\mathbf{q}\cdot\mathbf{r}}|\Phi(\mathbf{r})\rangle+\text{H.c.}]\\&-\mathbf{u}\cdot\mathbf{p}_0+\mathbf{u}\cdot\mathbf{P}_0 ,\end{aligned} \tag{11}$$

where we have defined the integral

$$\begin{aligned}g_l=2N^2L\{\mu^2L^2\int_0^{1/2}t^2e^{-\mu^2L^2t^2}\sin^2[l\pi(t+1/2)]dt\\-l\pi\int_0^{1/2}te^{-\mu^2L^2t^2}\sin[2l\pi(t+1/2)]dt\} .\end{aligned} \tag{12}$$

Minimizing J with respect to $\mathbf{p}_0$ and f_k^{*} then leads to

$$f_k=-\frac{V_k^{*}}{l-\mathbf{u}\cdot\mathbf{k}}\langle\Phi(\mathbf{r})|e^{-i\mathbf{q}\cdot\mathbf{r}}|\Phi(\mathbf{r})\rangle \tag{13}$$

and

$$\mathbf{p}_0=\tfrac{1}{2}\mathbf{u}=m\mathbf{u}\ . \tag{14}$$

From Eqs. (13) and (14), we can calculate the expectation value of the Hamiltonian up to the second order in u,

$$\langle\Psi|H|\Psi\rangle=E_l+\tfrac{1}{2}m_{\parallel}^{*}u_{\parallel}^{2}+\tfrac{1}{2}m_z^{*}u_z^{2}\ , \tag{15}$$

and the expectation value of the total momentum

$$\langle\Psi|\mathbf{P}|\Psi\rangle=\mathbf{P}_0=m_{\parallel}^{*}\mathbf{u}_{\parallel}+m_z^{*}\mathbf{u}_z\ . \tag{16}$$

In Eq. (15), E_l denotes the polaron energy in the subband l and is given by

$$E_l=\lambda^2+\mu^2[1-g_l(\mu)]+\left[\frac{l\pi}{L}\right]^2-\frac{\alpha}{\pi}h_l(\lambda,\mu)\ , \tag{17}$$

where we have defined a new integral

$$h_l=2\int_0^{\infty}\kappa e^{-\kappa^2/2\lambda^2}d\kappa\int_0^{\infty}\frac{dq}{\kappa^2+q^2}\left\{2N^2L\int_0^{1/2}\sin^2[l\pi(t+\tfrac{1}{2})]\cos(qLt)e^{-\mu^2L^2t^2}dt\right\}^2\ . \tag{18}$$

The polaron effective mass is in general anisotropic in a quantum well and is given by

$$\frac{m_{\parallel}^{*}}{m}=\frac{4\alpha}{\pi}\int_0^{\infty}\kappa^3e^{-\kappa^2/2\lambda^2}d\kappa\int_0^{\infty}\frac{dq}{\kappa^2+q^2}\left\{2N^2L\int_0^{1/2}\sin^2[l\pi(t+\tfrac{1}{2})]\cos(qLt)e^{-\mu^2L^2t^2}dt\right\}^2\ , \tag{19a}$$

$$\frac{m_z^{*}}{m}=\frac{8\alpha}{\pi}\int_0^{\infty}\kappa e^{-\kappa^2/2\lambda^2}d\kappa\int_0^{\infty}\frac{q^2dq}{\kappa^2+q^2}\left\{2N^2L\int_0^{1/2}\sin^2[l\pi(t+\tfrac{1}{2})]\cos(qLt)e^{-\mu^2L^2t^2}dt\right\}^2\ . \tag{19b}$$

Equations (17) and (19) express the energy levels and effective masses of the polaron in terms of the variational parameters λ and μ, which are determined by minimizing the energy. Since these parameters depend explicitly on the coupling constant and the well width, they are calculated numerically later when we discuss the anisotropy of the problem. From Eqs. (15) and (16), it is also clearly seen that the Lagrange multiplier $\mathbf{u}$ represents indeed the polaron velocity $\mathbf{u}=(u_x,u_y,u_z)=(\mathbf{u}_{\parallel},u_z)$. The binding energy of the polaron for the lowest state in the subband l is therefore

$$E_b=\left[\frac{\pi}{L}\right]^2-E_l\ . \tag{20}$$

IV. LIMITING CASES

We have established a strong-coupling theory for quasi-two-dimensional polarons in a quantum well. By choosing a two-parameter trial wave function, energy levels and anisotropic effective mass are derived for the strongly coupled electron-phonon system. They are all expressed as functions of the well width L. In this section, we show that these results yield correct limits in both the 2D and 3D cases.

We consider the ground state ($l=1$). In the limit $L\rightarrow\infty$, the confinement in the z direction is released and hence the electron is essentially moving in the bulk. Consequently, we can set $\mu=\lambda$. Thus, we find from Eq. (17) the ground state

$$E_1=\frac{3}{2}\lambda^2-\sqrt{\frac{2}{\pi}}\alpha\lambda+\left[\frac{\pi}{L}\right]^2\ . \tag{21}$$

The effective mass now becomes isotropic and is given by

$$m_{\parallel}^{*}=m_z^{*}=m^{*}=\frac{2}{3}\left[\frac{2}{\pi}\right]^{1/2}\alpha\lambda^3\ . \tag{22}$$

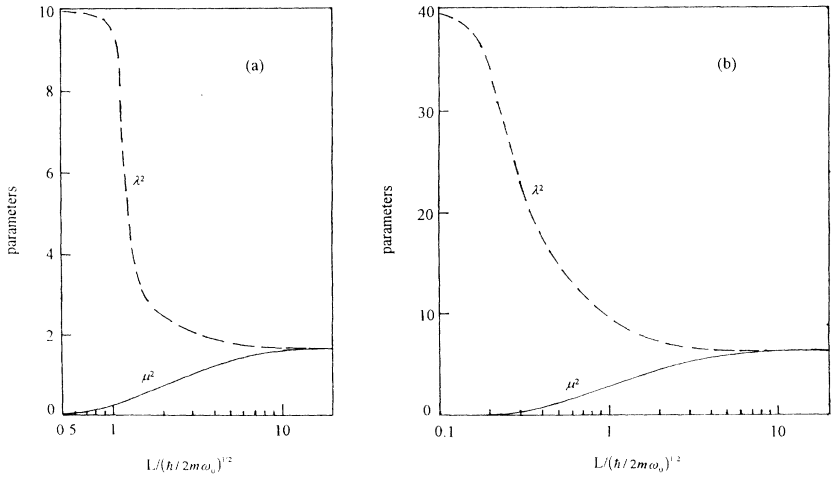

FIG. 1. Parameters λ_0^2 and μ_0^2 for the polaron ground state as functions of the well width L. The solid line represents μ^2 and the dashed line represents λ^2. (a) $\alpha=5$, (b) $\alpha=10$.

The parameter λ is determined by minimizing the energy E_1, and we find

$$\lambda_0=\frac{\alpha}{3}\left[\frac{2}{\pi}\right]^{1/2} . \tag{23}$$

Substituting Eq. (23) back into Eqs. (21) and (22), we obtain finally the polaron ground-state binding energy and effective mass in the 3D limit as

$$E_b=\left[\frac{\pi}{L}\right]^2-E_1=\frac{\alpha^2}{3\pi} , \tag{24}$$

$$\frac{m^*}{m}=\frac{16}{81\pi^2}\alpha^4 . \tag{25}$$

These results are in complete agreement with what can be found in the literature for 3D strong-coupling polaron theory.[24]

In the limit $L\to 0$, no motion in the z direction is possible and $u_z=0$. Following the same procedure as above, we find

$$E_1(\lambda)=\lambda^2+\mu^2\left[1+\frac{\pi}{4}\right]-\frac{\sqrt{2\pi}}{2}\alpha\lambda+\left[\frac{\pi}{L}\right]^2 , \tag{26}$$

$$m^*=\frac{\sqrt{2\pi}}{2}\alpha\lambda^3 . \tag{27}$$

Minimizing the energy yields the parameters

$$\mu_0=0, \quad \lambda_0=\frac{\sqrt{2\pi}}{4}\alpha . \tag{28}$$

Inserting Eq. (28) into (26) and (27), we find the ground-state binding energy and effective mass

$$E_b=\left[\frac{\pi}{L}\right]^2-E_1=\frac{\pi}{8}\alpha^2 , \tag{29}$$

$$\frac{m^*}{m}=\frac{\pi^2}{16}\alpha^4 . \tag{30}$$

Again, these results agree completely with what has been obtained in Ref. 24 for the 2D case.

TABLE I. Numerical values of the length and energy units for some selected materials.

Material	MnO	TiCl	Cu_2O	KI	KCl
$\sqrt{\hbar/2m\omega_0}$ (Å)	12.9	21.8	6.3	10.0	9.1
$\hbar\omega_0$ (meV)	90	21.5	53.28	18.11	25.46

V. RESULTS AND DISCUSSION

For quantum wells of arbitrary width, numerical calculation is inevitable. The procedure is outlined as follows. For a given sublevel l, we choose a particular α value. E_l in Eq. (17) is then minimized to determine the variational parameters λ and μ for a fixed L. Repeating this variational calculation for various L leads to λ_0 and μ_0 as functions of α and L.

We consider in this paper two choices of α, namely, $\alpha=5$ and 10 for $l=1$. We first apply the variational principle

$$\frac{\delta E_l}{\delta\lambda}=0, \quad \frac{\delta E_l}{\delta\mu}=0 , \tag{31}$$

which implies

$$2\lambda-\frac{\alpha}{\pi}\frac{\delta h_l}{\delta\lambda}=0 , \tag{32a}$$

$$2\mu[1-g_l(\mu)]-\mu^2\frac{\delta g_l}{\delta\mu}-\frac{\alpha}{\pi}\frac{\delta h_l}{\delta\mu}=0 . \tag{32b}$$

It is not interesting to reproduce the explicit forms as they follow by direct differentiation of Eqs. (12) and (18). Equations (32) are solved numerically for variational parameters λ_0 and μ_0. The parameters thus determined are plotted versus L for a given α in Fig. 1. The difference between the two curves serves as a measure of the anisotropy. It is observed that the problem is practically isotropic for large L, as it should be in the 3D limit. We recall that the unit for length is $\sqrt{\hbar/2m\omega_0}$ which depends strongly on the material. Its numerical value for some selected materials is listed in Table I. The anisotropy

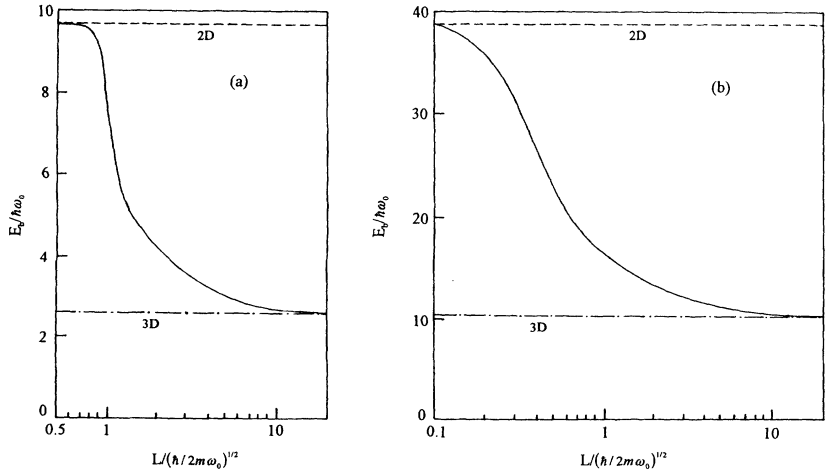

FIG. 2. Ground-state binding energy E_b of the confined polaron as a function of L (solid line). The corresponding polaron binding energies in the 2D and 3D cases are represented by dashed and dot-dashed lines, respectively. (a) $\alpha=5$, (b) $\alpha=10$.

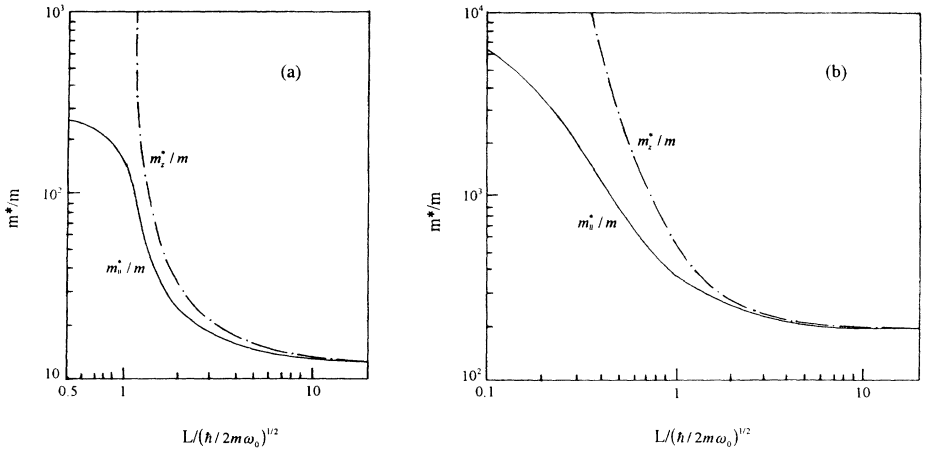

FIG. 3. Anisotropic effective masses of the polaron in its ground state. The solid line represents $m_\parallel^*/m$ and the dot-dashed line represents m_z^*/m. (a) $\alpha=5$, (b) $\alpha=10$.

starts to show up at about $L\sim10$, where the curves begin to split. The splitting increases rapidly as L becomes smaller than 1. When L decreases further, the system approaches the 2D limit, and $\mu_0^2\rightarrow0$ as expected. Figure 1 also indicates that the smaller the coupling constant α, the quicker the change of behavior from 3D to 2D.

The polaron binding energy E_b is plotted as a function of L in Fig. 2. The corresponding binding energies for 2D (dashed line) and 3D (dot-dashed line) are also shown for comparison purpose. The binding energy of a Q2D polaron is seen to approach the 3D value for $L\geq10$. It increases with decreasing well width, but the rate of change is much faster for weaker coupling ($\alpha=5$) than for stronger coupling. This is understandable because a stronger coupling implies a smaller size of the polaron. Thus, a well of width $L=1$ is practically 2D-like for polarons with $\alpha=5$, but remains 3D-like for polarons with $\alpha=10$. It is also of interest to note from Figs. 1 and 2 that there exists a region of dimensionality crossover. This is particularly clear for $\alpha=5$ for which the crossover region is $1\leq L\leq4$, corresponding to about 30 Å for such materials as MnO and ZnO.

In Fig. 3, we plot the anisotropic effective masses. The behavior of the parallel effective mass as the width changes is similar to that of the binding energy. The z-direction effective mass, on the other hand, increases monotonically as the well width decreases, and eventually blows up at small widths. The infinite m_z^*/m simply reflects the fact that the confinement becomes so strong that the polaron motion in the z direction is completely frozen.

It would be interesting to investigate the strong-coupling theory of polarons confined in quantum-well structures of finite barrier height. Then the contributions from surface or interface phonon modes should also be included in the consideration. This is now being carried out and results will be reported elsewhere in the future.

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[1]H. Ibach, Phys. Rev. Lett. **24**, 1416 (1970); **27**, 253 (1971).
[2]E. Evans and D. L. Mills, Solid State Commun. **11**, 1093 (1972); Phys. Rev. B **8**, 4404 (1973).
[3]J. Sak, Phys. Rev. B **6**, 3981 (1972).
[4]G. D. Mahan, Phys. Rev. B **5**, 739 (1972); R. S. Zheng, S. W. Gu, and D. L. Lin, Solid State Commun. **59**, 331 (1986).
[5]S. Das Sarma, Phys. Rev. B **27**, 2590 (1983).
[6]D. M. Larsen, Phys. Rev. B **30**, 4595 (1984).
[7]Wu Xiaoguang, F. M. Peeters, and J. T. Devreese, Phys. Rev. B **32**, 7964 (1985); **34**, 2621 (1986).
[8]M. H. Degani and O. Hipolito, Phys. Rev. B **35**, 7717 (1987).
[9]C. Y. Chen, T. Z. Ding, and D. L. Lin, Phys. Rev. B **35**, 4398 (1987); C. Y. Chen, D. L. Lin, and T. Z. Ding, *ibid.* **36**, 9816 (1987).
[10]R. Greene and K. K. Bajaj, Phys. Rev. B **31**, 913 (1985).
[11]X. X. Liang, S. W. Gu, and D. L. Lin, Phys. Rev. B **34**, 2807 (1986).
[12]C. Y. Chen, P. W. Jin, and S. Q. Zhang, J. Phys. Condens. Matter. **4**, 4483 (1992).
[13]F. Comas, C. Trallero-Giner, and R. Riera, Phys. Rev. B **39**, 5907 (1989).
[14]M. H. Degani and O. Hipolito, Surf. Sci. **196**, 459 (1988).
[15]D. L. Lin, R. Chen, and T. F. George, J. Phys. Condens. Matter. **3**, 4645 (1991).
[16]D. L. Lin, R. Chen, and T. F. George, Phys. Rev. B **43**, 9328 (1991); R. Chen, D. L. Lin, and T. F. George, Chin. J. Phys. (Taipei) **30**, 165 (1992).
[17]W. Xu, F. M. Peeters, and J. T. Devreese, Phys. Rev. B **48**, 1562 (1993).
[18]L. D. Landau and S. I. Pekar, Zh. Eksp. Teor. Fiz. **16**, 341 (1946).
[19]R. P. Feynman, Phys. Rev. **97**, 660 (1955).
[20]F. M. Peeters and J. T. Devreese, Phys. Rev. B **36**, 4442 (1987).
[21]S. Sil and A. Chatterjee, J. Phys. Condens. Matter. **3**, 9401 (1991); **5**, 1169 (1993).
[22]T. Yildirim and A. Ercelebi, J. Phys. Condens. Matter. **3**, 1271 (1991).
[23]R. Chen, D. L. Lin, M. Shukri, T. F. George, and C. Y. Chen, Phys. Rev. B **46**, 13 357 (1992).
[24]A. Chatterjee, Phys. Rev. B **41**, 1668 (1990).