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The polaronic shift of exciton binding energy in quantum dots with a degenerate valence band

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Abstract

The polaronic shifts of the electron, hole and exciton ground state energies are studied by taking into account the interaction between single particles and both bulk- and interface-type LO (longitudinal optical) phonons in spherical quantum dots with a degenerate valence band. Inclusion of the space confinement effect on the phonon spectrum causes decrease of the electron, hole and exciton lowest-energy polaronic shift. The polaronic shift of the exciton energy is relatively small due to the cancellation of the polaronic effects owing to the opposite charges of an electron and a hole. It is shown that in the strong-confinement regime the polaronic shift of the exciton ground state energy is caused by valence band degeneracy. Valence band degeneracy also causes the ‘interface-type’ part of the hole polaronic shift.

1. Introduction

In recent years considerable effort has been devoted to achieving an understanding of the unusual physical properties of quantum dots (QDs), because of their promise for application in device engineering. Three-dimensional carrier confinement gives rise to discrete energy levels with large spacings, provided that the QDs are sufficiently small. Therefore, QDs are often referred to as artificial atoms [1]. Realization of such QD systems in semiconductor heterostructures, and their integration with conventional electronic and optical devices, offers great potential for applications [2–4].

In QDs, not only the electron spectra but also the phonon modes become discrete due to the three-dimensional confinement. In [5], on the basis of the dielectric continuum approximation, expressions for the eigenfunctions corresponding to the bulk-type LO phonon and interface-type phonon modes have been derived, and the interaction Hamiltonian has been obtained. Resonant Raman scattering measurements [6] and far-infrared reflectance techniques [7] also verified experimentally the discrete character of the phonon spectrum in such structures.

During the past few decades the interaction of electrons and optical phonons in QDs has been the subject of a number of experimental and theoretical studies, as it strongly influences QD carrier relaxation and optical properties. In addition to numerous works on polaronic effects in quantum wells and wires, recently there have been a considerable number of theoretical studies on the same effects including the confinement problem in the QD system. Free polaron properties in QDs were studied earlier using variational techniques [5, 8–13] and within second-order perturbation theory [14–18]. The effect of the electron–phonon interaction on an electron bound to an impurity in a spherical QD is studied theoretically in [19–21]. Free and impurity-bound polaron states in the presence of the magnetic field are also studied [16, 22]. The effects of LO phonons on an exciton play an important role in the optical properties of QDs and have been investigated experimentally and theoretically [5, 9, 23–29].

Despite the great variety of works devoted to the polaronic problem in the QDs, we know of only one paper [30] where the electron–phonon interaction and polaron corrections are considered taking into account the valence band degeneracy but without including the effect of phonon confinement.

The purpose of our work is to study the energy of interaction of quasiparticles (electron, hole, exciton) with polar optical phonons in the strong-confinement regime in QDs, taking into account both the valence band degeneracy and the effect of spatial confinement on the phonon spectrum.

2. Theory

2.1. Energy spectra and wavefunctions of electrons, holes and excitons

Since the observation of discrete optical transitions in QDs, great efforts have been made to study the internal electronic structure of the confined electron–hole pair states. In particular, one is interested in the energies and symmetries of the energetically lowest pair states, as these states essentially determine the band edge absorption and luminescence. To describe the electronic structure of nanometre size QDs of the most popular semiconductors GaAs, InP, InSb, InAs, CdSe, CdTe, the envelope function approximation has been established [31–36].

We neglect the coupling between Γ_6 , Γ_7 and Γ_8 bands in the semiconductors mentioned above. The conduction and valence band states are built from these bulk bands in the framework of the envelope function approximation, for spherical QDs having a cubic lattice structure. The ground electron state is doubly degenerate with respect to its spin projection. The conduction band wavefunctions are given by

$$\psi_{S_z^{(e)}}^{(c)} = f^{(c)}(\mathbf{r}_e) u_{S_z^{(e)}}^{(c)}(\mathbf{r}_e), \quad (1)$$

where the $u_{S_z^{(e)}}^{(c)}(\mathbf{r}_e)$ are the Bloch functions at the bottom of the Γ_6 bulk band for $S_z^{(e)} = \pm 1/2$. If we assume that electrons are confined in a spherical potential well with infinitely high walls, then their energy spectrum and wavefunctions in the parabolic conduction band approximation are given by [37]

$$\varepsilon_{pst}^{(e)} = \frac{\hbar^2 \lambda_p^2(s)}{2m_e r_0^2}, \quad (2)$$

$$f_{pst}^{(c)}(\mathbf{r}_e) = \frac{\sqrt{2} j_s(\lambda_p(s) \frac{r_e}{r_0})}{r_0 j_{s+1}(\lambda_p(s))} Y_{st}(\theta_e, \varphi_e), \quad (3)$$

where m_e is the electron effective mass, r_0 is the radius of the QD, $j_s(x)$ is the spherical Bessel function of order s , $\lambda_p(s)$ is the root of this function and $Y_{st}(\theta, \varphi)$ are the spherical harmonics.

To consider the hole states in the spherical QD, we write the Luttinger Hamiltonian in the spherical approximation as follows [38]:

$$\hat{H} = \left(\gamma_1 + \frac{5}{2}\gamma \right) \frac{\hat{\mathbf{p}}^2}{2m_0} - \frac{\gamma}{m_0} (\hat{\mathbf{p}} \mathbf{J})^2, \quad (4)$$

where \mathbf{p} is the momentum and \mathbf{J} are the 4×4 matrices of the angular momentum $J = 3/2$; $\gamma > 0$ and γ_1 are the Luttinger parameters relevant to the light- and heavy-hole effective masses: $m_{lh} = m_0(\gamma_1 + 2\gamma)^{-1}$ and $m_{hh} = m_0(\gamma_1 - 2\gamma)^{-1}$, where m_0 is mass of the free electron.

The first quantum size level of holes is fourfold degenerate with respect to the projection M of its total momentum $\mathbf{F} = \mathbf{J} + \mathbf{L}$ ($F = 3/2$; $M = 3/2, 1/2, -1/2, -3/2$), where \mathbf{L} is the orbital momentum. The lowest valence band state is given by [34, 35]

$$\varepsilon_{3/2}^{(h)} = \frac{\hbar^2 \chi^2(\beta)}{2m_{hh}r_0^2}, \quad (5)$$

$$\psi_{3/2}^{(v)}(\mathbf{r}_h) = 2 \frac{A(\beta)}{r_0^{3/2}} \sum_{l=0,2} R_l \left(\frac{r_h}{r_0} \right) \sum_{m+\mu=3/2} \begin{pmatrix} 3/2 & l & 3/2 \\ \mu & m & -3/2 \end{pmatrix} Y_{lm}(\theta_h, \varphi_h) u_\mu, \quad (6)$$

where $\beta = m_{lh}/m_{hh}$, $\chi(\beta)$ is the first root of the equation

$$j_0(\chi) j_2(\chi\sqrt{\beta}) + j_2(\chi) j_0(\chi\sqrt{\beta}) = 0, \quad (7)$$

$\begin{pmatrix} i & k & l \\ m & n & p \end{pmatrix}$ are Wigner $3j$ symbols and u_μ are the Bloch functions of the fourfold-degenerate valence band Γ_8 [39].

The radial functions $R_l(x)$ are

$$R_2(x) = j_2(\chi x) + \frac{j_0(\chi)}{j_0(\chi\sqrt{\beta})} j_2(\chi x\sqrt{\beta}), \quad R_0(x) = j_0(\chi x) - \frac{j_0(\chi)}{j_0(\chi\sqrt{\beta})} j_0(\chi x\sqrt{\beta}), \quad (8)$$

where the constant $A(\beta)$ is determined by the normalization condition.

For the excitons we will employ the so-called strong-confinement approximation, which allows us to consider the electron-hole Coulomb interaction as a small perturbation in the single-particle Hamiltonian. In this approximation, the electron-hole pair ground state energy can be written as

$$\varepsilon_0^{(e-h)} = \varepsilon_{100}^{(e)} + \varepsilon_{3/2}^{(h)} - \int d\mathbf{r}_e \int d\mathbf{r}_h V_{Coulomb}(\mathbf{r}_e, \mathbf{r}_h) |\psi_0^{(e-h)}(\mathbf{r}_e, \mathbf{r}_h)|^2, \quad (9)$$

where

$$\psi_0^{(e-h)}(\mathbf{r}_e, \mathbf{r}_h) = f_{100}^{(e)}(\mathbf{r}_e) \psi_{3/2}^{(v)}(\mathbf{r}_h) \quad (10)$$

is the uncorrelated excitonic state wavefunction.

2.2. The interaction of a single particle and a polar optical phonon in a quantum dot

Let us consider a single particle (electron, hole, exciton) which is confined perfectly in a QD and interacts with LO phonons. The Hamiltonian of the system can be written as

$$H = H_i + H_{ph} + H_{int}, \quad (11)$$

where H_i is the single-particle Hamiltonian having eigenvalues for the electron, hole and exciton ground state given by (2), (5) and (9) respectively. The LO phonon Hamiltonian H_{ph} can be written as [5, 9, 40]

$$H_{ph} = \sum_{\alpha\sigma} \hbar\omega_{\alpha\sigma} a_{\alpha\sigma}^+ a_{\alpha\sigma}, \quad (12)$$

where $\sigma = 1$ (2) denotes the bulk-type (interface-type) LO phonon. The index α is given by $\alpha \equiv (n = 1, 2, \dots; l = 0, 1, \dots; m = 0, \pm 1, \dots, \pm l)$ for the bulk-type phonon and $\alpha \equiv (l = 1, 2, \dots; m = 0, \pm 1, \dots, \pm l)$ for the interface-type phonon, $a_{\alpha\sigma}^+$ ($a_{\alpha\sigma}$) is the creation (annihilation) operator of the $\alpha\sigma$ mode. The frequency for the bulk-type LO phonon $\omega_{\alpha 1}$ is equal to just the bulk LO phonon frequency. For the interface-type LO phonon we have

$$\omega_{\alpha 2} = \omega_l = \left[\frac{\kappa_{out} + (\kappa_{out} + \kappa_0)l}{\kappa_{out} + (\kappa_{out} + \kappa_\infty)l} \right]^{1/2} \omega_{TO}, \quad (13)$$

where κ_0 (κ_∞) is the static (high-frequency) dielectric constant of the QD and κ_{out} is that of the surrounding medium. H_{int} is the single-particle–phonon interaction Hamiltonian which can be written as:

(a) for the electron ($\gamma = 1$) and hole ($\gamma = 2$)

$$H_{int}^{e(h)} = (-1)^{\gamma+1} \left\{ \sum_{nlm} [V_l(q) j_l(qr_{e(h)}) Y_{lm}(\theta_{e(h)}, \varphi_{e(h)}) a_{\alpha 1} + \text{H.c.}] + \sum_{lm} \left[S_l \left(\frac{r_{e(h)}}{r_0} \right)^l Y_{lm}(\theta_{e(h)}, \varphi_{e(h)}) a_{\alpha 2} + \text{H.c.} \right] \right\}, \quad (14)$$

where

$$V_l(q) = - \left(\frac{4\pi\hbar\omega_{LO}e^2}{j_{l+1}(qr_0)r_0^3q^3\kappa^*} \right)^{1/2}, \quad (15)$$

$$S_l = - \frac{\sqrt{l}\kappa_\infty}{l\kappa_\infty + (l+1)\kappa_{out}} \omega_{LO} \left(\frac{2\pi\hbar e^2}{\omega_l r_0 \kappa^*} \right)^{1/2}, \quad q = \lambda_n(l)/r_0, \kappa^{*-1} = \kappa_\infty^{-1} - \kappa_0^{-1}; \quad (16)$$

(b) for the exciton

$$H_{int}^{(ex)} = \sum_{nlm} [V_l(q)(j_l(qr_e)Y_{lm}(\theta_e, \varphi_e) - j_l(qr_h)Y_{lm}(\theta_h, \varphi_h))a_{\alpha 1} + \text{H.c.}] + \sum_{lm} \left[S_l \left(\left(\frac{r_e}{r_0} \right)^l Y_{lm}(\theta_e, \varphi_e) - \left(\frac{r_h}{r_0} \right)^l Y_{lm}(\theta_h, \varphi_h) \right) a_{\alpha 2} + \text{H.c.} \right]. \quad (17)$$

In the strong-confinement regime, the energy of the Coulomb interaction between an electron and a hole as well as the energy of their interaction with polar optical phonons are smaller than the distance between the size-quantization levels in the QD. Therefore, averaging the single-particle–phonon system Hamiltonian (11) using wavefunctions (1), (6) and (10) for electrons, holes and excitons respectively, we obtain

$$\hat{H}_0 = \varepsilon + \sum_{\alpha\sigma} \hbar\omega_{\alpha\sigma} a_{\alpha\sigma}^+ a_{\alpha\sigma} + \sum_{qlm} (V_l \rho_{\alpha 1}^{(i)} a_{\alpha 1} + \text{H.c.}) + \sum_{lm} (S_l \rho_{\alpha 2}^{(i)} a_{\alpha 2} + \text{H.c.}), \quad (18)$$

where

$$\varepsilon = \varepsilon_{pst}^{(e)}, \quad \rho_{\alpha 1}^{(e)} = \int d\mathbf{r}_e j_l(qr_e) Y_{lm}(\theta_e, \varphi_e) |\psi_{S_\zeta^{(e)}}^{(c)}|^2, \quad (19)$$

$$\rho_{\alpha 2}^{(e)} = \int d\mathbf{r}_e \left(\frac{r_e}{r_0} \right)^l Y_{lm}(\theta_e, \varphi_e) |\psi_{S_\zeta^{(e)}}^{(c)}|^2$$

for electrons;

$$\varepsilon = \varepsilon_{3/2}^{(h)}, \quad \rho_{\alpha 1}^{(h)} = \int d\mathbf{r}_h j_l(qr_h) Y_{lm}(\theta_h, \varphi_h) |\psi_{3/2}^{(v)}|^2, \quad (20)$$

$$\rho_{\alpha 2}^{(h)} = \int d\mathbf{r}_h \left(\frac{r_h}{r_0} \right)^l Y_{lm}(\theta_h, \varphi_h) |\psi_{3/2}^{(v)}|^2$$

for holes;

$$\varepsilon = \varepsilon_0^{(e-h)}, \quad \rho_{\alpha 1}^{(ex)} = \rho_{\alpha 1}^{(e)} - \rho_{\alpha 1}^{(h)}, \quad \rho_{\alpha 2}^{(ex)} = \rho_{\alpha 2}^{(e)} - \rho_{\alpha 2}^{(h)} \quad (21)$$

for excitons.

2.3. The energy polaronic shift calculation

The intermediate-coupling method is used to treat the single-particle–phonon interaction. The Hamiltonian and the single-particle state $|\psi\rangle$ are transformed to $H_{eff} = U^{-1}H_0U$ and $|\psi_0\rangle = U^{-1}|\psi\rangle$ by performing the unitary transformation with the unitary operator

$$U = \exp \sum_{\alpha\sigma} (f_{\alpha\sigma} a_{\alpha\sigma} - f_{\alpha\sigma}^* a_{\alpha\sigma}^+), \quad (22)$$

where parameters $f_{\alpha\sigma}$, $f_{\alpha\sigma}^*$ can be obtained from the variational conditions

$$\frac{\partial}{\partial f_{\alpha\sigma}} \langle \psi_0 | H_{eff} | \psi_0 \rangle = 0, \quad \frac{\partial}{\partial f_{\alpha\sigma}^*} \langle \psi_0 | H_{eff} | \psi_0 \rangle = 0. \quad (23)$$

The trial function for the transformed state $|\psi_0\rangle$ is chosen as the product of the single-particle state $|\Phi\rangle$ and the zero-phonon state $|0\rangle$. After some calculations we can find

$$H_{eff} = \varepsilon + \sum_{\alpha\sigma} \hbar\omega_{\alpha\sigma} a_{\alpha\sigma}^+ a_{\alpha\sigma} - \sum_{nlm} \frac{|V_l|^2 |\rho_{\alpha 1}|^2}{\hbar\omega_{LO}} - \sum_{lm} \frac{|S_l|^2 |\rho_{\alpha 2}|^2}{\hbar\omega_l}. \quad (24)$$

The last two terms in (24) are the single-particle energy polaronic shifts ($\Delta\varepsilon_{pol1}^{(i)}$ and $\Delta\varepsilon_{pol2}^{(i)}$) in the QD due to the interaction of the electron ($i = e$), hole ($i = h$) and exciton ($i = ex$) with the bulk-type and interface-type phonons, respectively. After some calculations we can obtain

$$\Delta\varepsilon_{pol1}^{(e)} = -\frac{e^2}{2\kappa^*r_0} \left\{ 1 - \frac{\text{Si}(2\pi)}{\pi} + \frac{\text{Si}(4\pi)}{2\pi} \right\}, \quad (25)$$

$$\Delta\varepsilon_{pol2}^{(e)} = 0, \quad (26)$$

$$\begin{aligned} \Delta\varepsilon_{pol1}^{(h)} = & -\frac{e^2}{2\kappa^*r_0} A^2(\beta) \left\{ \frac{1}{\pi^2} \int_0^1 dx x P(x) \right. \\ & \times \left[(\pi - x) \int_0^x dy y^2 P(y) + x \int_x^1 dy y(\pi - y) P(y) \right] \\ & + \frac{2}{5} \int_0^1 dx x^4 Q(x) \left[\left(\frac{1}{x^5} - 1 \right) \int_0^x dy y^4 Q(y) \right. \\ & \left. \left. + \int_x^1 dy y^4 \left(\frac{1}{y^5} - 1 \right) Q(y) \right] \right\}, \quad (27) \end{aligned}$$

$$\Delta\varepsilon_{pol2}^{(h)} = -\frac{e^2}{2\kappa^*r_0} A^2(\beta) \frac{4}{5} \left(\frac{\sqrt{2}\varepsilon_\infty\omega_{LO}}{(2\varepsilon_\infty + 3\varepsilon_{out})\omega_2} \right)^2 \left| \int_0^1 dx x^4 Q(x) \right|^2, \quad (28)$$

$$\begin{aligned} \Delta\varepsilon_{pol1}^{(ex)} = & -\frac{e^2}{2\kappa^*r_0} \left\{ 1 - \frac{\text{Si}(2\pi)}{\pi} + \frac{\text{Si}(4\pi)}{2\pi} \right. \\ & - A(\beta) \int_0^1 dx x^2 P(x) F(x) + A^2(\beta) \int_0^1 dx x^2 P(x) \\ & \left. \times \left[\left(\frac{1}{x} - 1 \right) \int_0^x dy y^2 P(y) + \int_x^1 dy y^2 \left(\frac{1}{y} - 1 \right) P(y) \right] \right\} \end{aligned}$$

$$\begin{aligned}
& + \frac{2}{5} A^2(\beta) \int_0^1 dx x^4 Q(x) \\
& \times \left[\left(\frac{1}{x^5} - 1 \right) \int_0^x dy y^4 Q(y) + \int_x^1 dy y^4 \left(\frac{1}{y^5} - 1 \right) Q(y) \right] \Bigg\}, \quad (29)
\end{aligned}$$

$$\Delta \varepsilon_{pol2}^{(ex)} = \Delta \varepsilon_{pol2}^{(h)}, \quad (30)$$

where

$$P(x) = R_0^2(x) + R_2^2(x), \quad Q(x) = R_0(x)R_2(x), \quad (31)$$

$$\begin{aligned}
F(x) = \frac{1}{4\pi} [2\pi(x-1) - \text{Ci}(2\pi) + \text{Ci}(2\pi x) - \ln(x) - \sin(2\pi x)] \\
+ \left(\frac{1}{x} - 1 \right) \left(\frac{x}{2} - \frac{\sin(2\pi x)}{4\pi} \right). \quad (32)
\end{aligned}$$

$\text{Si}(x)$ and $\text{Ci}(x)$ are the sine and cosine integral functions, respectively.

3. Results and numerical calculations

According to relations (25)–(30), when the QDs radius decreases, the ground state energy polaronic shift of a particle (electron, hole, exciton) increases as r_0^{-1} . The polaronic shift of the ground state energy of the exciton for interface-type phonon interaction coincides with the hole energy polaronic shift. This is a consequence of the fact that the electron in the ground state interacts only with the bulk-type phonons. The hole and, therefore, the exciton in the ground state interact with interface-type phonons of frequency $\omega_2 = \omega_T \sqrt{(3\kappa_{out} + 2\kappa_0)/(3\kappa_{out} + 2\kappa_\infty)}$. This interaction takes place only when taking into account the valence band degeneracy.

The inclusion of the space confinement effect on the phonon spectrum causes decrease of the energy polaronic shift of the particles considered. Comparison of equation (25) with the corresponding result obtained for the electron in [30] shows that accounting for phonon confinement decreases the polaronic shift by a factor of about 2.3.

For the hole and, therefore, for exciton too, the valence band degeneracy leads to the dependence of the polaronic shift on the ratio of light-and heavy-hole masses. In figure 1 the solid curve shows the dependence of $\Delta \varepsilon_{pol}^{(h)} = \Delta \varepsilon_{pol1}^{(h)} + \Delta \varepsilon_{pol2}^{(h)}$ on β , and the dashed curve shows the result of [30] obtained without considering the phonon confinement. It is obvious that accounting for the phonon confinement leads to substantial decrease of the polaronic shift. Depending on the ratio of light-and heavy-hole masses, the result obtained in [30] is greater than our result by a factor of 3.6 (when $\beta = 0.16$) to 3.8 (when $\beta = 0.98$).

In figure 2, the solid curve indicates the dependence of $\Delta \varepsilon_{pol}^{(ex)} = \Delta \varepsilon_{pol1}^{(ex)} + \Delta \varepsilon_{pol2}^{(ex)}$ on β , and the dashed curve indicates the corresponding result without phonon confinement. As in previous cases, in this case, phonon confinement leads to decrease of the polaronic shift of the exciton ground state energy. This is especially obvious in the region $\beta < 0.4$. In this region, accounting for the phonon confinement causes decrease of the polaronic shift by on average a factor of 1.4.

The curves in figure 2 show that, in comparison with the electron and hole polaronic shifts, the polaronic shift for the exciton is smaller by an order of magnitude, both with and without inclusion of the phonon confinement effect. This is due to the compensation of the polaronic shifts of opposite charges of the electron and hole. It should be noted that the polaronic shift of the exciton is entirely absent regardless of the valence band degeneracy.

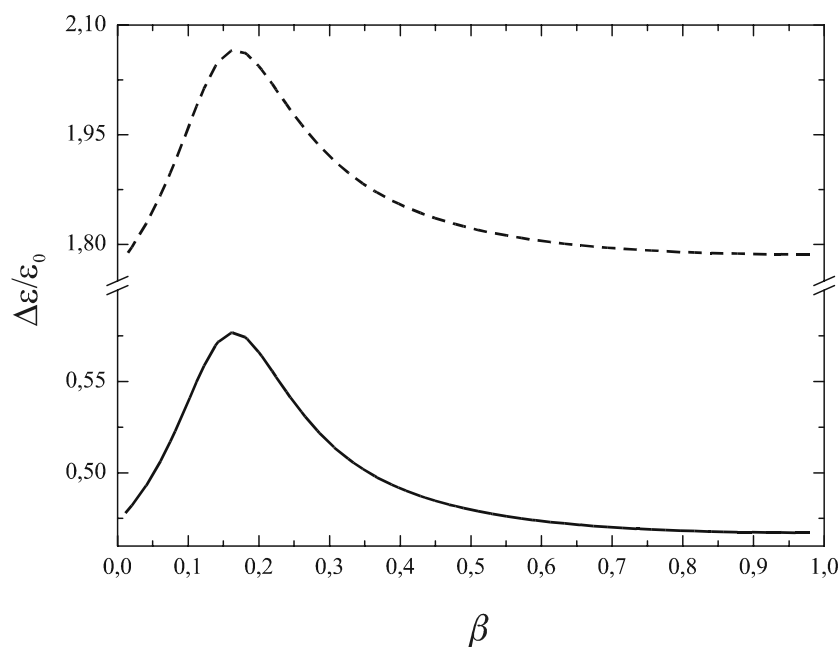


Figure 1. The dependence of the hole ground state energy polaronic shift on β with (solid curve) and without (dashed curve) the phonon confinement effect ($\epsilon_0 = -e^2/2\kappa^*r_0$).

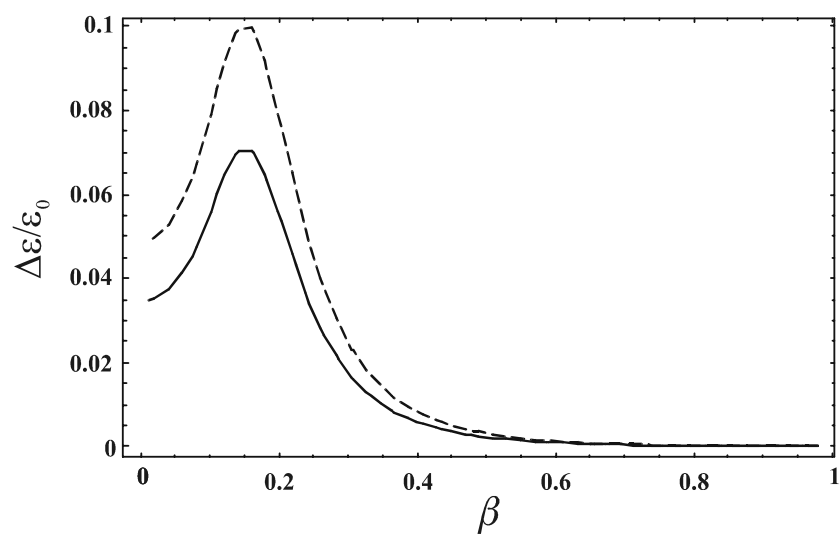


Figure 2. The dependence of the exciton ground state energy polaronic shift on β with (solid curve) and without (dashed curve) the phonon confinement effect ($\epsilon_0 = -e^2/2\kappa^*r_0$).

4. Conclusions

In this paper we have studied the interaction of single particles and polar optical phonons in QDs caused both by the valence band degeneracy and by the effect of spatial confinement on the phonon spectrum. Using the variation method, analytical results are obtained for polaronic

shifts of the ground state energy of an electron, a hole and an exciton in the strong-confinement regime. It is shown that the effect of interface-type phonons on the ground state energy is weak (for an electron it is entirely absent) and appears only with regard to valence band degeneracy. The exciton and the hole interact only with the interface-type phonons of frequency ω_2 . All particles in the ground state interact with bulk-type phonons. However, the polaronic shift owing to this interaction is smaller than the respective quantity obtained in the case where the phonon confinement effect is not included. For an electron, the phonon confinement leads to decrease of the polaronic shift by a factor 2.3. For a hole this factor can vary over the range from 3.6 to 3.8, depending on the ratio of light- and heavy-hole masses. The polaronic shift of the exciton is much smaller than those for the hole and the electron due to the cancellation of the polaronic effects owing to the opposite charges of an electron and a hole.

Acknowledgments

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