

Polaronic correction to the first excited electronic energy level in an anisotropic semiconductor quantum dot

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Abstract. Within the framework of second-order Rayleigh-Schrödinger perturbation theory, the polaronic correction to the first excited state energy of an electron in a quantum dot with anisotropic parabolic confinements is presented. Compared with isotropic confinements, anisotropic confinements will make the degeneracy of the excited states to be totally or partly lifted. On the basis of a three-dimensional Fröhlich's Hamiltonian with anisotropic confinements, the first excited state properties in two-dimensional quantum dots as well as quantum wells and wires can also be easily obtained by taking special limits. Calculations show that the first excited polaronic effect can be considerable in small quantum dots.

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1 Introduction

In polar crystalline semiconductors, the coupling between the electron and the longitudinal-optical (LO) phonon leads to the formation of a composite particle called the polaron. Polarons have an important effect on electronic and optical properties of the system. Recent developments in semiconductor fabrication technique have made it possible to obtain a system with reduced dimensionality, such as quantum wells, wires and dots. For a variety of new interesting electronic and optical properties in low-dimensional systems, polaron problems naturally attract great attention and have been widely investigated [1–18].

So far, most investigations on polarons are mainly focused on the polaronic correction to the ground state (GS) electronic level and quantum size effects on the interaction of the ground state electron and LO phonon [1–8]. The general result of accomplished investigations is that polaronic corrections are considerable in small quantum sizes and increase with the decreasing confinement length. Whereas, relatively much fewer investigations are available in the literature on the excited states, though, the excited states are important in optical absorption and emission which involve electron transitions.

Recently, Mukhopadhyay and Chatterjee [10] have investigated the first-excited state (ES) polaron energies in a three-dimensional (3D) and two-dimensional (2D) quantum dot (QD) with isotropic parabolic confinements by using second-order Rayleigh-Schrödinger perturbation

theory (RSPT), which is used for the small electron-LO phonon coupling constant and the electronic level spacing between the first excited and the ground state being substantially away from a real phonon-assisted transition. They found the excited state polaron exhibits new properties different from those for the ground state, e.g., as the confinement frequency approach the frequency of the LO phonon, the excited polaron will be unstable with respect to the emission of a phonon. Motivated by the work of Mukhopadhyay and Chatterjee, we extend their method to study the polaronic correction to the first excited electronic energy level in a QD with general parabolic confinements. The confining potential that we choose here is only axially symmetric (i.e., symmetrical in the xy -plane). The first excited state in isotropic 3D dots are three-fold degenerate, while in anisotropic 3D quantum dots (QDs) the degeneracy is totally or partly lifted. Anisotropic confined QDs will give some new properties compared with those for isotropic QDs. More interestingly, in the framework of 3D Fröhlich's Hamiltonian with anisotropic confinements, the excited polaronic correction in 2D QDs are easily obtained by taking special limits. In this anisotropic confinement framework, many properties of polarons can be easily illustrated such as why the polaronic correction in 2D QDs is larger than that in 3D QDs. The first excited polaronic corrections in bulk, quantum wells and wires are also obtained as 'by-products', which are found to be equal to those for their ground counterparts. This equality reflects that the energy levels of the unconfined electrons in bulk, quantum wells and wires are continuous.

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This article comprises the following. First we derive the expressions of the first excited polaronic correction in a general potential by using the second-order RSPT method. Then our numerical results are presented and discussed. Finally, a brief conclusion is drawn in our investigation.

2 Formulation

We assume the parabolic potential in the QD is only axially symmetric. Considering that bulk phonons play the most important role in the polaron effects and the contribution from the surface phonons is negligible [4], we assume that the electron interacts with only bulk phonons. This assumption was widely used for parabolic confinements [1–3, 6, 9, 10]. The Fröhlich's Hamiltonian of a polaron can be written as

$$H = -\frac{1}{2} \nabla_r^2 + \frac{1}{2} (\omega_\rho^2 \rho^2 + \omega_z^2 \mathbf{z}^2) + \sum_q a_q^+ a_q + \sum_q [\xi_q \exp(-i\mathbf{q} \cdot \mathbf{r}) a_q^+ + \text{h.c.}], \quad (1)$$

where all vectors are in three dimensions and the units have been chosen as $m_e = \hbar = \omega_{\text{LO}} = 1$ (Feynman units); m_e being the effective mass of the electron, and ω_{LO} the LO phonon frequency. $\mathbf{r} = (\rho, \mathbf{z})$, $\rho = (\mathbf{x}, \mathbf{y})$; $\omega_\rho = \omega_{\rho h}/\omega_{\text{LO}}$ and $\omega_z = \omega_{zh}/\omega_{\text{LO}}$, where $\omega_{\rho h}$ and ω_{zh} are the frequencies of the confining parabolic potential in the xy -plane and the direction \mathbf{z} , respectively; a_q^+ (a_q) is the creation (annihilation) operator for a LO phonon of wave vector q , and ξ_q is given by [7]

$$|\xi_q|^2 = \frac{2^{3/2}\pi}{Vq^2}\alpha, \quad (2)$$

where V is the volume of the three-dimensional crystal and α is the electron-phonon coupling constant.

Recently, Hameau et al. [11] show the electrons and the LO phonons in QDs are always in a strong coupling regime even for the material with relatively small α (e.g. for GaAs with $\alpha = 0.068$). The corresponding value of α seems to be much larger than (by a factor of 2, although a most recent result shows an increase only up to 25% [19]) than that in bulk due to the strain or other confinement effects. Otherwise, Lelong and Lin [20] also interpret the experimental results conducted by Hameau et al. [11] by using a perturbative approach with the same α as in bulk. In order to investigate the special polaron property in anisotropic QDs compared with that in isotropic QDs, we calculate the electron-LO phonon interaction energy by using the second-order RSPT method for $\alpha \ll 1$ with the same value of α as in bulk. The treatment is not very rigorous if the dot sizes are extremely small, but may still serve as a good approximation to obtain some of the most important properties of the electron-phonon interaction effects in QDs with anisotropic confinement. The second-order

RSPT correction to the electron energy in the first excited state is given by

$$\Delta E_1 = - \sum_m \sum_q \frac{|\langle \phi_m(\mathbf{r}) | \xi_q \exp(-i\mathbf{q} \cdot \mathbf{r}) | \phi_1(\mathbf{r}) \rangle|^2}{E_m - E_1 + 1}, \quad (3)$$

where

$$\left\{ -\frac{1}{2} \nabla_r^2 + \frac{1}{2} (\omega_\rho^2 \rho^2 + \omega_z^2 \mathbf{z}^2) \right\} \phi_m(\mathbf{r}) = E_m \phi_m(\mathbf{r}), \quad (4)$$

$$\begin{aligned} \phi_m(\mathbf{r}) = & \left(\frac{\omega_\rho \sqrt{\omega_z}}{\pi^{3/2} 2^{m_x+m_y+m_z} m_x! m_y! m_z!} \right)^{1/2} \\ & \times H_{m_x}(\sqrt{\omega_\rho}x) H_{m_y}(\sqrt{\omega_\rho}y) H_{m_z}(\sqrt{\omega_z}z) \\ & \times \exp \left[-\frac{1}{2} (\omega_\rho \rho^2 + \omega_z \mathbf{z}^2) \right], \quad (5) \end{aligned}$$

$$E_m = (m_x + m_y + 1)\omega_\rho + (m_z + 1/2)\omega_z. \quad (6)$$

$H_n(x)$ is the Hermite polynomial of order n . E_m is the energy of the unperturbed m -th state with parabolic confinements. For the first excited state, $E_1 = 2\omega_\rho + \omega_z/2$ for $\omega_\rho < \omega_z$ and $E_1 = \omega_\rho + 3\omega_z/2$ for $\omega_\rho > \omega_z$, respectively.

Using the transformations

$$\frac{1}{E_m - E_1 + 1} = \int_0^\infty \exp[-(E_m - E_1 + 1)t] dt, \quad \text{for } m \geq 1 \quad (7)$$

and the Slater sum rule for the Hermite polynomials

$$\begin{aligned} \sum_{j=0}^{\infty} \frac{1}{2^j j!} H_j(\lambda x) H_j(\lambda x') \exp \left[-\frac{1}{2} \lambda^2 (x^2 + x'^2) - 2jp \right] = \\ \frac{\exp(p)}{\sqrt{2 \sinh(2p)}} \exp \left\{ -\frac{1}{4} \lambda^2 [(x + x')^2 \right. \\ \left. \times \tanh(p) + (x - x')^2 \coth(p)] \right\}, \quad (8) \end{aligned}$$

one can perform the summation over m_x, m_y, m_z in equation (3) easily. Then using

$$\sum_q \frac{\exp[i\mathbf{q} \cdot (\mathbf{r} - \mathbf{r}')] }{q^2} = \frac{V}{4\pi} \frac{1}{|\mathbf{r} - \mathbf{r}'|}, \quad (9)$$

one can integrate over the electron position vectors \mathbf{r} and \mathbf{r}' by transforming these vectors into center-of-mass vector $\mathbf{u} = (\mathbf{r} + \mathbf{r}')/2$ and relative vectors $\mathbf{v} = (\mathbf{r} - \mathbf{r}')/2$. The final form depends on the relative values of ω_ρ and ω_z . We will proceed to discuss three different cases.

A. $\omega_z = \omega_\rho = \omega$

In this case, the first excited state is three-fold degenerate, and the second-order RSPT polaronic correction to the first excited state energy of an electron is obtained as follows:

$$\Delta E_1 = \frac{-\alpha}{6} \sqrt{\frac{\omega}{\pi}} \int_0^{+\infty} dt \exp[(\omega - 1)t] \times \left[\frac{5 \exp(-\omega t) + 1}{\sqrt{1 - \exp(-\omega t)}} - 1 \right] + \frac{\alpha}{6(\omega - 1)} \sqrt{\frac{\omega}{\pi}}, \quad (10)$$

where care has been taken to treat correctly the singularity originating from the $m = 0$ term of the sum over m in equation (3), which has been treated separately and appears as the last term of equation (10). Equation (10) is the same as the result of 3D QDs obtained by Mukhopadhyay and Chatterjee [10].

B. $\omega_z > \omega_\rho$

When the confinement in the direction z is stronger than that in the other two directions, the first excited state is two-fold degenerate related to the xy -plane confinement, and can be assumed as E_{1x} . The first excited polaronic correction is obtained as

$$\Delta E_1 = \frac{-\alpha}{4} \sqrt{\frac{\omega_z}{\pi}} \int_0^{+\infty} dt \exp[(\omega_\rho - 1)t] \times \left[\frac{\exp(-\omega_\rho t) \{ [1 - \exp(\omega_\rho t)] \sqrt{\zeta - 1} \}}{\sqrt{1 - \exp(-\omega_z t)} (\zeta - 1)^{3/2}} + \frac{[(3 + \exp(\omega_\rho t)) \zeta - 4] \arctan \sqrt{\zeta - 1}}{\sqrt{1 - \exp(-\omega_z t)} (\zeta - 1)^{3/2}} - \kappa \right] + \frac{\alpha}{4(\omega_\rho - 1)} \sqrt{\frac{\omega_z}{\pi}} \kappa, \quad (11)$$

where

$$\zeta = \frac{\omega_z [1 + \coth(\frac{1}{2} \omega_z t)]}{\omega_\rho [1 + \coth(\frac{1}{2} \omega_\rho t)]},$$

$$\kappa = \frac{1 - \frac{\omega_z}{\omega_\rho} + \frac{\omega_z}{\omega_\rho} \sqrt{\frac{\omega_z}{\omega_\rho} - 1} \arctan(\sqrt{\frac{\omega_z}{\omega_\rho} - 1})}{(\frac{\omega_z}{\omega_\rho} - 1)^2}.$$

From equation (11), equation (10) can be easily extracted if taking $\omega_z \rightarrow \omega_\rho$.

If the direction z is strongly confined, i.e. $\omega_z \rightarrow \infty$, we will show

$$\lim_{\omega_z \rightarrow \infty} \left(1 + \coth \frac{1}{2} \omega_z t \right) = 2,$$

$$\lim_{\omega_z \rightarrow \infty} \arctan \sqrt{\zeta - 1} = \frac{\pi}{2}.$$

Then we get

$$\Delta E_1 = \frac{-\alpha \sqrt{\pi \omega_\rho}}{8} \int_0^{+\infty} dt \exp[(\omega_\rho - 1)t] \times \left[\frac{(3 \exp(-\omega_\rho t) + 1)}{\sqrt{1 - \exp(-\omega_\rho t)}} - 1 \right] + \frac{\alpha \sqrt{\pi \omega_\rho}}{8(\omega_\rho - 1)}. \quad (12)$$

This is just the case of 2D isotropic QDs which is well-studied as an important low-dimensional quantum structure [10]. In equation (12), if we further take the limit $\omega_\rho \rightarrow 0$, we can get the first excited energy correction of pure 2D polarons: $\Delta E_1 = -(\pi/2)\alpha$, which is equal to the ground polaronic correction ΔE_0 [2]. The equality reflects the characteristic that the energy spectrum in the xy -plane of 2D unconfined systems is continuous.

As the confinement in the xy -plane is at the weak-confinement limit (i.e. $\omega_\rho \rightarrow 0$), equation (11) will yield the result for quantum wells with parabolic potentials. Then we can get

$$\lim_{\omega_\rho \rightarrow 0} \omega_\rho \left(1 + \coth \frac{1}{2} \omega_\rho t \right) = \frac{2}{t},$$

and

$$\Delta E_1 = -\alpha \sqrt{\frac{\omega_z}{\pi}} \int_0^{+\infty} dt \frac{e^{-t}}{\sqrt{1 - \exp(-\omega_z t)}} \times \frac{\arctan \sqrt{\zeta' - 1}}{\sqrt{\zeta' - 1}}, \quad (13)$$

where $\zeta' = \frac{1}{2} \omega_z t [1 + \coth(\frac{1}{2} \omega_z t)]$. ΔE_1 is equal to ΔE_0 [1], reflecting the characteristic that the energy spectrum in the z direction is continuous.

In equation (13), if further taking $\omega_z \rightarrow 0$, in which all the three directions are at the weak-confinement limit, we can get

$$\lim_{\omega_z \rightarrow 0} \frac{\sqrt{\omega_z}}{\sqrt{1 - \exp(-\omega_z t)}} = \frac{1}{\sqrt{t}},$$

$$\lim_{\omega_z \rightarrow 0} \frac{1}{2} \omega_z t \left[1 + \coth \left(\frac{1}{2} \omega_z t \right) \right] = 1,$$

$$\lim_{\zeta' \rightarrow 1} \frac{\arctan \sqrt{\zeta' - 1}}{\sqrt{\zeta' - 1}} = 1.$$

and $\Delta E_1 = -\alpha$. This form corresponds to the first excited energy correction of the free 3D polarons in the bulk limit [21], which equals to their ground energy correction for the continuous electron energy spectrum.

C. $\omega_z < \omega_\rho$

As the confinement in the direction z is weaker than those in the other two directions, the first excited state is E_{1z} and one-fold degenerate, and the form of the first excited

polaronic correction is

$$\begin{aligned} \Delta E_1 = & -\frac{\alpha}{4} \sqrt{\frac{\omega_z}{\pi}} \int_0^{+\infty} dt \exp[(\omega_z - 1)t] \\ & \times \left[\frac{e^{-\omega_z t} \left\{ 2(1 - e^{\omega_z t}) \sqrt{1 - \zeta} \right.}{(1 - \zeta)^{3/2} \sqrt{1 - \exp(-\omega_z t)}} \right. \\ & \left. + \frac{(1 + e^{\omega_z t} - 2\zeta) \ln \left(\frac{1 + \sqrt{1 - \zeta}}{1 - \sqrt{1 - \zeta}} \right)}{(1 - \zeta)^{3/2} \sqrt{1 - \exp(-\omega_z t)}} - \kappa' \right] \\ & + \frac{\alpha}{4(\omega_z - 1)} \sqrt{\frac{\omega_z}{\pi}} \kappa', \quad (14) \end{aligned}$$

where

$$\kappa' = \frac{\ln \left(\frac{1 + \sqrt{1 - \frac{\omega_z}{\omega_\rho}}}{1 - \sqrt{1 - \frac{\omega_z}{\omega_\rho}}} \right) - 2 \sqrt{1 - \frac{\omega_z}{\omega_\rho}}}{(1 - \frac{\omega_z}{\omega_\rho})^{3/2}}.$$

From equation (14), equation (10) can also be extracted if taking $\omega_z \rightarrow \omega_\rho$.

When $\omega_z \rightarrow 0$, the system then becomes quantum wires, which is one of the most important cases of low-dimensional quantum structures. We can get

$$\Delta E_1 = -\frac{\alpha}{2\sqrt{\pi}} \int_0^{+\infty} dt \frac{\exp(-t)}{\sqrt{t(1 - \zeta'')}} \ln \left(\frac{1 + \sqrt{1 - \zeta''}}{1 - \sqrt{1 - \zeta''}} \right), \quad (15)$$

where $\zeta'' = 2/(\omega_\rho t [1 + \coth(\frac{1}{2}\omega_\rho t)])$. For the continuous electron energy spectrum in the z direction, ΔE_1 will be equal to ΔE_0 which has been derived in reference [1].

Furthermore, in equation (15), if taking $\omega_\rho \rightarrow 0$, it corresponds to the case of free 3D polarons. We obtain

$$\lim_{\zeta'' \rightarrow 1} \frac{1}{\sqrt{1 - \zeta''}} \ln \left(\frac{1 + \sqrt{1 - \zeta''}}{1 - \sqrt{1 - \zeta''}} \right) = 2,$$

and $\Delta E_1 = -\alpha$, which also equals to the ground polaron energy shift of free 3D polarons.

From equations (10–15), we found the most several low-dimensional polaronic correction, including 3D, 2D anisotropic and isotropic QDs, quantum wells and wires as well as bulk limit, can be obtained on the basis of a 3D anisotropic Fröhlich's Hamiltonian.

3 Numerical results and discussion

In order to investigate the dependence of the polaron energy shift on the QD size conveniently, we define the dimensionless confinement length $l = 1/\sqrt{\omega}$, where $l = l_0/r_0$, $r_0 = [\hbar/(m_e \omega_{LO})]^{1/2}$, $l_0 = [\hbar/(m_e \omega)]^{1/2}$. Correspondingly, $l_\rho = 1/\sqrt{\omega_\rho}$ and $l_z = 1/\sqrt{\omega_z}$. Since the first excited polaronic corrections in quantum wells and wires as well as in the bulk limit are equal to those for their

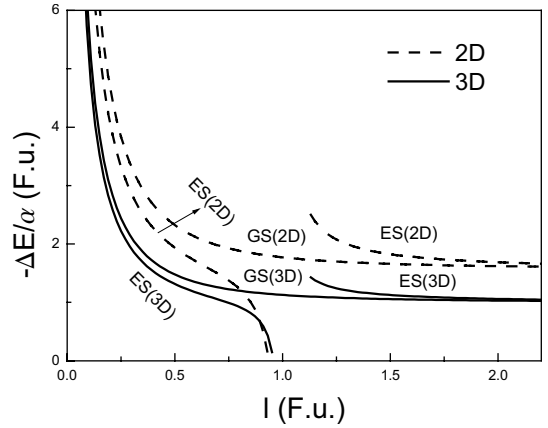


Fig. 1. Polaronic corrections $-\Delta E/\alpha$ (in Feynman units, F.u.) of the ground- and first- excited state versus the confinement length l (in F.u.) in isotropic 2D and 3D quantum dots.

ground state counterparts, which have been well studied [1], we will lay emphasis on the first excited polaronic correction of 3D and 2D QDs with equations (10–12) and (14).

Figure 1 shows $-\Delta E_1/\alpha$ for both 2D and 3D isotropic QDs as a function of the dimensionless confinement length l . The GS polaronic corrections $-\Delta E_0/\alpha$ are also plotted for comparison. Evidently there is a singularity at $l = 1$ of the plot corresponding to the first excited states both for 2D and 3D QDs. As $l > 1$ and $l < 1$, the ES polaronic corrections increase with the decreasing confinement length, so does the GS case. The polaronic corrections in 2D are larger than those in 3D both for the first excited and the ground state, which can be easily understood since 2D is the limit case in 3D as one of the dimensions is strongly confined.

Interestingly, in Figure 1, as $l > 1$, the ES polaronic correction are larger than, and tend to in the large confinement length, the GS polaronic correction both in 2D and in 3D QDs. With the increasing confinement length, the ES and GS polaronic correction identically tend to their 2D or 3D free polaronic GS data. While for $l < 1$, the ES polaronic correction are smaller than that for the GS one. Figure 2 presents the difference between the ES and the GS polaronic corrections $(-\Delta E_1/\alpha) - (-\Delta E_0/\alpha)$ versus the confinement length. As l is smaller than about 0.5 (far from the singularity), the difference become larger with the decreasing confinement length. It is noted that, near $l = 1$, the RSPT method is not rigorously valid for the appearance of the singularity and one should make other improved methods such as variational calculations [9].

For anisotropic 3D QDs, some new results different from isotropic cases are obtained, which are illustrated in Figure 3 and Figure 4. In Figure 3, we plot $(-\Delta E_1/\alpha)$ as a function of the dimensionless confinement length in the direction z with three fixed xy -plane confinements. As $l_\rho = 1.4$, there is no singularity for the first excited state, because when l_z changed from larger than to smaller than 1.4, the first excited state is changed from E_{1z} to E_{1xy} . Hence there will be a turning point at $l_z = 1.4$ for

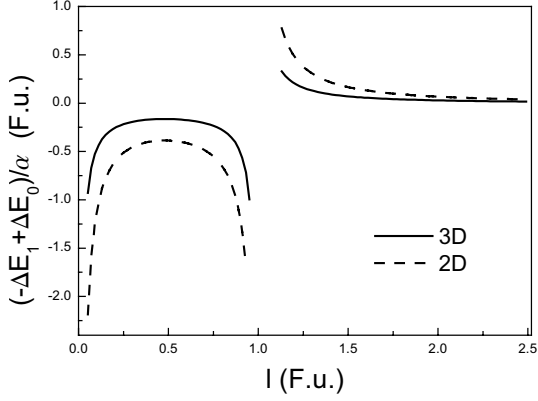


Fig. 2. The difference between the first excited polaronic correction $-\Delta E_1/\alpha$ (in Feynman units, F.u.) and the ground correction $-\Delta E_0/\alpha$ (in F.u.) as a function of the confinement length l (in F.u.).

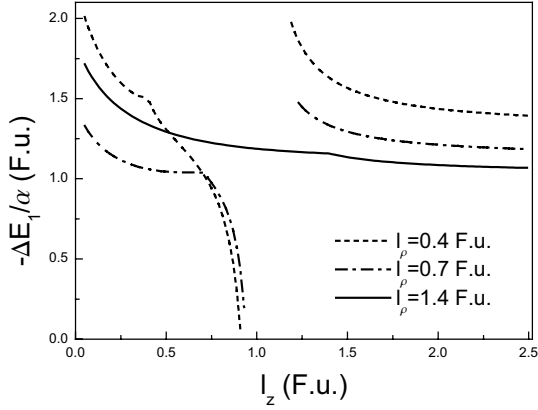


Fig. 3. The first excited polaronic correction $-\Delta E_1/\alpha$ as a function of the confinement length in the direction z with the fixed confinement length in the xy -plane. Both the energy and the confinement length are denoted in Feynman units (F.u.).

$l_\rho = 1.4$ curve, so do the other two cases at $l_z = 0.4$ for $l_\rho = 0.4$ and $l_z = 0.7$ for $l_\rho = 0.7$, respectively. For $l_\rho = 0.4$ and $l_\rho = 0.7$, the singularity still exists. Similar results are also shown in Figure 4, corresponding to the plot of the fixed $l_z = 1.4$, no singularity points exist; while for $l_z = 0.4$ and $l_z = 0.7$, singularity exists with a turning point at $l_\rho = 0.4$ and $l_\rho = 0.7$, respectively. It is noted that, the plot for $l_\rho = 1.4$ is higher than that for $l_\rho = 0.7$ in Figure 3 when $l_z < 1$, and the plot for $l_z = 1.4$ is higher than both of those for $l_z = 0.4$ and 0.7 when $l_\rho < 1$ in Figure 4. Both of these results are irregular and disobey the rule that the smaller QDs have larger corrections. This irregularity originates from the singularity, which can be illustrated by comparing the polaronic corrections near the singularity in Figure 1.

Finally, in some realistic cases, e.g. for GaAs, $\alpha = 0.068$, $m_e = 0.066 m_0$ (m_0 is the free electron mass). the energy Feynman units $\hbar\omega_{LO} = 36.7$ meV, the confinement length Feynman units $[\hbar/(m_e\omega_{LO})]^{1/2} = 5.57$ nm, when the confinement length l_z and l_ρ of anisotropic QDs are 2.0 nm and 6.5 nm, respectively, the first ES polaronic

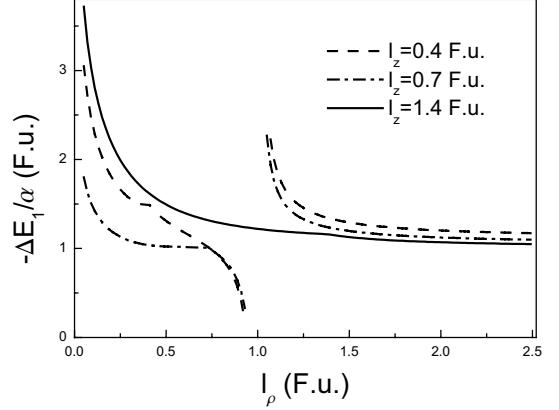


Fig. 4. The first excited polaronic correction $-\Delta E_1/\alpha$ as a function of the confinement length in the xy -plane with the fixed confinement length in the z direction. Both the energy and the confinement length are denoted in Feynman units (F.u.).

correction is 4.1 meV. Compared to bulk value 2.5 meV, this first ES polaronic correction is enhanced much. It is considerable compared to the unperturbed energy of the first excited state (which is 196 meV), and the unperturbed energy of the ground state (which is 169 meV). We can also find it is the same order of the transition energy between the ground and the first excited state (which is 27 meV), and of the exciton binding energy (< 5 meV [22]). The electron-LO phonon interaction should be taken into account especially when one considers the fine structure of the energy level.

4 Conclusions

We have calculated the effect of the electron-LO phonon interaction on the first excited electronic energy level in an anisotropic parabolic QD by using the second-order RSPT method. In the framework of an anisotropic 3D Fröhlich's Hamiltonian, the excited state properties in 2D QDs, quantum wells and wires are naturally obtained by taking special limits. The polarons in anisotropic QDs have some new properties compared with those for isotropic QDs, e.g. there exists a turning point in the plot for the first excited polaronic correction, and singularity may not exist in some cases while there always exists a singularity point for isotropic QDs. Moreover, numerical calculation shows that the polaronic correction to the first excited electronic energy can be considerable in the strong confinement region.

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