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Original article

Effect of electron-phonon interaction on the first excited energy level of a Gaussian GaAs quantum dot

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ABSTRACT The effect of electron-phonon interaction on the first excited state of a three-dimensional polar semiconductor quantum dot with Gaussian confinement is studied using the second-order Rayleigh-Schrödinger perturbation theory. An analytical expression for the first excited state polaronic correction is obtained under a plausible approximation. It is shown that this energy depends both on the strength and range of the Gaussian potential. Finally our theory is applied to a GaAs quantum dot and it is shown that the polaronic effect to the first excited level can be significantly large if the size of the dot is small. Since the information of the excited states is important for the study of decoherence phenomena, our results could be useful for quantum information processing.

KEYWORDS electron-phonon interaction; first excited state; GaAs quantum dot

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1. Introduction

The subject of quantum dot (QD) has been extensively studied both theoretically [1–3] and experimentally [4,5] over the last four decades and a wealth of data on the electronic, thermodynamics optical and magnetic properties of QDs have piled up in the literature. One of the essential information that is required to carry out a theoretical investigation for the electronic and several other properties of a QD is the nature of the potential that is responsible for the confinement of the electrons in the QD. Several initial investigations have considered the confinement potential as a square well. According to this potential, the force experienced by the particles inside a QD is zero which is of course not true. Thus, the square-well potential is an over-simplified approximation of a realistic QD. Later, it has been shown based on the generalized Kohn theorem and the magneto-optical experiments that the confinement potential in a QD is essentially harmonic in character [6, 7]. This triggered a large number of theoretical studies in this area [8–10] as the parabolic potential can be handled much more easily. Some relatively recent experimental observations have advocated that the QD confinement potential does not truly conform to the parabolic form but in reality has an anharmonic character. Motivated by these new observations, Adamowsky and collaborators [11] have considered an attractive Gaussian potential as a model for the QD confinement potential to investigate certain properties of a QD. It turns out that a Gaussian potential is a much better model for quantum confinement in a QD than the parabolic potential or the square-well potential. The Gaussian potential mimics the behaviour of a harmonic potential near the minimum and therefore it does satisfy the generalized Kohn's theorem for low-lying states [6]. Because of its finite depth, the Gaussian potential can take care of realistic phenomena like ionization and tunneling processes which are not possible with a parabolic potential. Also, since this potential is smooth at the dot boundary, it is mathematically more convenient to handle [12, 13]. Another advantage of the Gaussian potential over the power law anharmonic potentials is that the latter can lead to divergences at larger distances.

Since in polar semiconductor QDs, the scale of the electron-longitudinal-optical (LO)-phonon interaction energy is of the same order as that of the confinement energy and the repulsive Coulomb energy, the electron-LO-phonon interaction is expected to influence the electronic properties equally as the other potentials do [1–3, 8–10, 13]. Indeed, the electron-phonon interaction has been shown to produce pronounced effects on the various properties of a polar Semiconductor QD. Most studies in this context have been essentially confined to the ground state (GS) [12, 13] and the investigations on the

excited states [14] have been only few and far between. Also the research studies have been mostly limited to square well and parabolic confining potentials [7–9, 15].

Chatterjee and collaborators [13] have carried out extensive studies on the GS polaronic properties of a Gaussian QD (GQD) using several approximate methods. The purpose of the present paper is to explore the nature of the polaronic correction to the first excited state (ES) of a one-electron GQD. Since the contribution of the electron-phonon interaction to the polaronic correction according to the first-order Rayleigh-Schrodinger perturbation theory (RSPT) vanishes, we will use the second-order RSPT in this work to obtain the lowest-order polaronic effect [14–18]. The knowledge of the excited states would be important for the study of optical properties of a QD system. We will formulate our theory for a general N-dimensional (ND) QD and finally get results for a 3D QD.

2. Main results

Let us consider a system of an electron that moves in an ND GQD and interacts with its LO phonons of dispersionless frequency ω_0. The system under consideration can be modeled by the modified Frohlich Hamiltonian in the Feynman dimensionless units as [8, 19]

$$H = \frac{\mathbf{p}^2}{2} - V_o e^{-\frac{r^2}{2R^2}} + \sum_{\mathbf{q}} b^\dagger_{\mathbf{q}} b_{\mathbf{q}} + \sum_{\mathbf{q}} \left(\xi_q \, e^{-i\mathbf{q}.\mathbf{r}} \, b^\dagger_{\mathbf{q}} + \ h.c.\right), \tag{1}$$

where all the vectors are N dimensional, $\mathbf{r}$ $(\mathbf{p})$ is the electron position (momentum) operator, V_o (R) is the depth (range) of the Gaussian confinement potential, $b^\dagger_{\mathbf{q}}$ $(b_{\mathbf{q}})$ is the creation (annihilation) operator the LO phonons of wave vector $\mathbf{q}$ and energy $\hbar\omega_o$ and ξ_q is the electron-phonon interaction coefficient which is given by [1, 5, 8]

$$\left|\xi_{\overrightarrow{q}}\right|^2 = \left[\frac{\Gamma\left(\frac{(N-1)}{2}\right) \, 2^{\left(N-\frac{3}{2}\right)} \, \pi^{\frac{(N-1)}{2}}}{v_N \, q^{N-1}}\right] \alpha, \tag{2}$$

where α is the dimensionless electron-phonon coupling constant which depends on the material parameters and v_N is the dimensionless volume of the N-dimensional QD. In 3D, $\left|\xi_{\overrightarrow{q}}\right|^2$ is given by the usual expression:

$$\left|\xi_{\overrightarrow{q}}\right|^2 = \left(\frac{2\sqrt{2}\pi}{v_N q^2}\right) \alpha \tag{3}$$

We assume that at least for low-lying states, it is possible to write the Gaussian potential as a sum of a harmonic potential and a small perturbative potential [20, 22]. So we rewrite Eq. (1) as

$$H = \frac{\mathbf{p}^2}{2} + \frac{1}{2}\widetilde{\omega}_h^2 r^2 - \ V_o \ + \sum_{\mathbf{q}} b^\dagger_{\mathbf{q}} b_{\mathbf{q}} \ + \lambda\left[-V_o\left(e^{\frac{-r^2}{2R^2}} - 1\right) - \frac{1}{2}\widetilde{\omega}_h^2 r^2\right] + \sum_{\overrightarrow{q}} \left(\xi_{\overrightarrow{q}} e^{-i\overrightarrow{q}.\overrightarrow{r}} b^\dagger_{\overrightarrow{q}} + \ h.c.\right), \tag{4}$$

where we have added and subtracted a parabolic potential and introduced a parameter λ. For $\lambda = 0$, the confinement potential becomes parabolic while for $\lambda = 1$, Eq. (4) describes a Gaussian confinement potential. We choose: $\widetilde{\omega}_h = \sqrt{V_o}/R$ so that the lowest-order perturbative term for the confinement potential becomes quartic in (r/R). Let us now write the Hamiltonian (4) as

$$H = H_0 + H_1 + H_2, \tag{5}$$

where

$$H_0 = \frac{\mathbf{p}^2}{2} + \frac{1}{2}\widetilde{\omega}_h^2 r^2 - \ V_o \ + \sum_{\mathbf{q}} b^\dagger_{\mathbf{q}} b_{\mathbf{q}} \, , \tag{6}$$

$$H_1 = \lambda\left[-V_o\left(e^{\frac{-r^2}{2R^2}} - 1\right) - \frac{1}{2}\widetilde{\omega}_h^2 r^2\right], \tag{7}$$

$$H_2 = \sum_{\overrightarrow{q}} \left(\xi_{\overrightarrow{q}} e^{-i\overrightarrow{q}.\overrightarrow{r}} b^\dagger_{\overrightarrow{q}} + \ h.c.\right), \tag{8}$$

where H_0 is exactly soluble, the harmonic oscillator part having the GS wave function $\phi_0^{ND}(\mathbf{r})$ and H_1 and H_2 can be treated as perturbations. We approximate the λ-term by its average w.r.t. $\phi_0^{ND}(\mathbf{r})$. Thus, we write:

$$\lambda\left[-\frac{V_o}{r^2}\left(e^{\frac{-r^2}{2R^2}} - 1\right) - \frac{1}{2}\widetilde{\omega}_h^2\right] r^2 \approx \ -\lambda\Big\langle \ \phi_0^{ND}(\mathbf{r}) \Big| \left[\frac{1}{2}\widetilde{\omega}_h^2 + \frac{V_o}{r^2}\left(e^{-\frac{r^2}{2R^2}} - 1\right)\right] \Big| \phi_0^{ND}(\mathbf{r}) \Big\rangle \, r^2 =$$

$$= \ \lambda V_o\left[\frac{2\widetilde{\omega}_h}{N-2} - \frac{1}{R^2} - \frac{4R^2\widetilde{\omega}_h^{N/2}}{(N-2)\left(2R^2\widetilde{\omega}_h + 1\right)^{(N-2)/2}}\right] r^2. \tag{9}$$

Substituting Eq. (9) in (5), we have an effective parabolic QD problem given by the effective Hamiltonian:

$$H_{eff} = \ \frac{\mathbf{p}^2}{2} + \frac{1}{2}\omega_h^2 r^2 - V_o \ + \sum_{\mathbf{q}} b^\dagger_{\mathbf{q}} b_{\mathbf{q}} + H_2 \ = H_{har} - V_o \ + \sum_{\mathbf{q}} b^\dagger_{\mathbf{q}} b_{\mathbf{q}} + H_2 = \ H_o^{eff} + H_2, \tag{10}$$

where

$$H_o^{eff} = H_{har} - V_o \ + \sum_{\mathbf{q}} b_{\mathbf{q}}^{\dagger} b_{\mathbf{q}}, \tag{11}$$

$$H_{har} = \frac{\mathbf{p}^2}{2} + \frac{1}{2}\omega_h^2 r^2, \quad \omega_h = \left[(1-\lambda)\,\widetilde{\omega}_h^2 + \frac{4\lambda V_o \widetilde{\omega}_h}{(N-2)} \left(1 - \left\{ \left(1 + \frac{\widetilde{\omega}_h}{2V_o} \right) \frac{N}{2} - 1 \right\}^{-1} \right) \right]^{1/2}. \tag{12}$$

We shall consider H_o^{eff} as an unperturbed Hamiltonian and H_2 as the perturbation. The eigenstate of H_o^{eff} can be written as $\left|\phi_j^{ND}\right\rangle \bigotimes |n\rangle$, where $\left|\phi_j^{ND}\right\rangle$ satisfies the Schrödinger equation

$$H_{har}\phi_j^{ND}(r) = E_j^{ND}\phi_j^{ND}(r), \tag{13}$$

with

$$\phi_j^{ND}(r) = \left(\frac{{\omega_h}^{N/2}}{\pi^{N/2} 2^{j_1+j_2+\cdots+j_N} (j_1! j_2! \ldots j_N!)} \right)^{1/2} \times H_{j_1}(\sqrt{\omega_h} x_1) H_{j_2}(\sqrt{\omega_h} x_2) \ldots . H_{j_N}(\sqrt{\omega_h} x_N)\, e^{-\frac{1}{2}\omega_h r^2}, \tag{14}$$

$H_{j_i}(\sqrt{\omega_h} x_i)$ being the Hermite polynomial and

$$E_j^{ND} = \left(j_1 + j_2 + \cdots + j_N + \frac{1}{2}N \right) \omega_h, \tag{15}$$

and $|\, n\rangle = \prod_{\mathbf{q}} |\, n_{\mathbf{q}}\rangle$, where $n_{\mathbf{q}} = 0,\ 1,\ 2,\ 3\ \ldots$etc., is the eigen state of $H_{ph} = \sum_{\mathbf{q}} b_{\mathbf{q}}^{\dagger} b_{\mathbf{q}}$ belonging to the eigen value n i.e.,

$$H_{ph}\,|n\rangle \ = \sum_{\mathbf{q}} b_{\mathbf{q}}^{\dagger} b_{\mathbf{q}}\,|n\rangle \ = \ \sum_{q} n_q\,|n\rangle \ = n\,|n\rangle \ ,$$

where $|0\rangle = \prod_{\mathbf{q}} |\, 0_{\mathbf{q}}\rangle$, $\ b_{\mathbf{q}}|\, 0_{\mathbf{q}}\rangle = 0$ for all $\mathbf{q}$ and $\sum_q n_q = n$. As we have already mentioned, the lowest-order perturbative contribution from the electron-phonon interaction H_2 comes from the second-order RSPT. We are here interested in calculating the polaronic correction to the first ES energy of H_o^{eff} due to H_2 [3, 13].

The second-order RSPT correction of the electron-phonon interaction H_2 to the first ES energy of H_o^{eff} is given by the expression [14, 15]:

$$\Delta E_1^{ND} = -\sum_{jn} \frac{\left| \left\langle \phi_j^{ND} \right| \langle n \mid H_2 \mid 0 \rangle \left| \phi_1^{ND} \right\rangle \right|^2}{E_j^{ND} - E_1^{ND} + \ n}, \tag{16}$$

which can be easily simplified to

$$\Delta E_1^{ND} = \sum_{j} \sum_{\mathbf{q}} \frac{\left| \left\langle \phi_j^{ND} \mid \xi_q e^{-iq.r} \mid \phi_1^{ND} \right\rangle \right|^2}{E_j^{ND} - E_1^{ND} + 1} \tag{17}$$

Applying the transformation

$$\frac{1}{E_j^{ND} - E_1^{ND} + 1} = \int_0^{\infty} e^{-\left(E_j^{ND} - E_1^{ND} + 1\right)t} dt \tag{18}$$

and the sum rule for the Hermite Polynomials

$$\sum_n \frac{1}{2^n n!} H_n(\lambda x)\, H_n(\lambda x^{\iota}) \times \exp\left[-\frac{\lambda^2}{2}\left(x^2 + x^{\iota 2} \right) - 2np \right] \ =$$

$$= \frac{e^p}{\sqrt{2\sinh(2p)}} \exp\left\{ -\frac{1}{4}\lambda^2 \left[(x + x^{\iota})^2 \tanh(p) \right] + (x - x^{\iota})^2 \coth(p) \right\}, \tag{19}$$

we obtain

$$\Delta \mathrm{E}_1^{\mathrm{ND}} = -\frac{\alpha}{8\sqrt{\omega_{\mathrm{h}}}} \frac{\Gamma\left((\mathrm{N}-1)/2\right)}{\Gamma\left(\mathrm{N}/2+1\right)} \times \left[(2N-1)\, B\left(\frac{1}{\omega_h}, \frac{1}{2} \right) + B\left((\frac{1}{\omega_h} - 1), \frac{1}{2} \right) \right], \tag{20}$$

$B(x, y)$ being the beta function. Finally, in terms of simpler functions, the first ES polaron energy is given in second-order RSPT as

$$E_{1,per}^{ND} = \frac{N+2}{2}\omega_h - \frac{\alpha\sqrt{\pi}}{2l} \frac{\Gamma\left(\frac{\mathrm{N}-1}{2}\right)}{\Gamma\left(\frac{N}{2}\right)} \frac{\Gamma\left(\frac{1}{\omega_h}+1\right)}{\Gamma\left(\frac{1}{\omega_h}+\frac{1}{2}\right)} \times \left(1 + \frac{\omega_h}{4N\left(1-\omega_h\right)} \right). \tag{21}$$

which can be calculated numerically to give the first ES energy of a GQD with polaronic interaction. One may note that the present approximation leads to a divergent result in 2D and therefore it cannot be applied to a 2D QD. This however is not a matter of concern here since we are interested in a 3D QD in this work, as mentioned earlier. For the sake of

completeness, we would like to mention that a slightly different approximation can be made to obtain results for a 2D GQD. For example, Eq. (9) can alternatively be approximated as

$$\lambda\left[-\frac{V_o}{r^2}\left(e^{\frac{-r^2}{2R^2}}-1\right)-\frac{1}{2}\widetilde{\omega}_h^2\right]r^2 \approx -\lambda\Big\langle\ \phi_0^{ND}(\mathbf{r})\ \Big|\ \left[\frac{1}{2}\widetilde{\omega}_h^2+\frac{V_o}{r^2}\left(e^{-\frac{r^2}{2R^2}}-1\right)\right]\ \Big|\phi_0^{ND}(\mathbf{r})\ \Big\rangle\ r^2 \approx$$

$$\approx -\lambda\left[\frac{1}{2}\widetilde{\omega}_h^2+\frac{V_o}{\left\langle\phi_0^{ND}(\mathbf{r})\ \middle|\ r^2\ \middle|\ \phi_0^{ND}(\mathbf{r})\right\rangle}\left(\left\langle\phi_0^{ND}(\mathbf{r})\ \middle|\ e^{-\frac{r^2}{2R^2}}\ \middle|\ \phi_0^{ND}(\mathbf{r})\right\rangle-1\right)\right]r^2, \tag{22}$$

which gives

$$\omega_h=\left[(1-\lambda)\,\widetilde{\omega}_h^2+2\lambda V_0\widetilde{\omega}_h\left\{1-2\widetilde{\omega}_h R^2\left(1+2\widetilde{\omega}_h R^2\right)^{-1}\right\}\right]^{1/2}. \tag{23}$$

One can easily see that the approximation made in Eq. (9) is supposed to give better results in all dimensions except in 2D. Therefore, we use Eq. (21) in this work, as we are interested here in a 3D GQD.

We are interested in applying our theory to a realistic material. Since GaAs QD is a useful QD for technological applications, we apply in this work our theory to a GaAs QD with Gaussian confinement. For GaAs, we have considered $m = 0.066\ m_0$, $\alpha = 0.068$ and $\hbar\omega_0 = 36.7$ meV [3,21]. Fig. 1. shows the behaviour of the ES polaronic correction $-\triangle E$ (in meV) with respect to the effective QD size R (in nm) for GQD of GaAs for different strengths of the confinement potential (V_0) [22]. One may note that in the effective parabolic problem, the energy of the first excited state electron plus the zero phonon energy is degenerate with the electron GS energy plus the one LO-phonon energy. Because of this degeneracy, an electron in the first ES of the effective QD problem is unstable to the emission of an LO phonon. This instability is expected to show up in the first ES polaronic correction in the form of a singularity. Indeed, the singularity structure is clearly visible in Fig. 1. For different potential strengths V_0, the singularity in the ES polaronic correction occurs at different values of the range R. As V_0 increases, the value of R at which the singularity appears also increases. Thus, the singularity point can be tuned by tuning the potential. In general (except around the singularity), the polaronic correction increases as the QD size decreases. The figure also shows that, as the confinement length is sufficiently reduced, the polaronic correction may become substantially large. For large values of R, the electron- phonon interaction corrections appear to be independent of R, as one would naturally expect in the case of bulk material. Therefore, we may conclude that the effect of electron- phonon interaction on the first ES of a GQD is really important when the size of the size of the GQD is small.

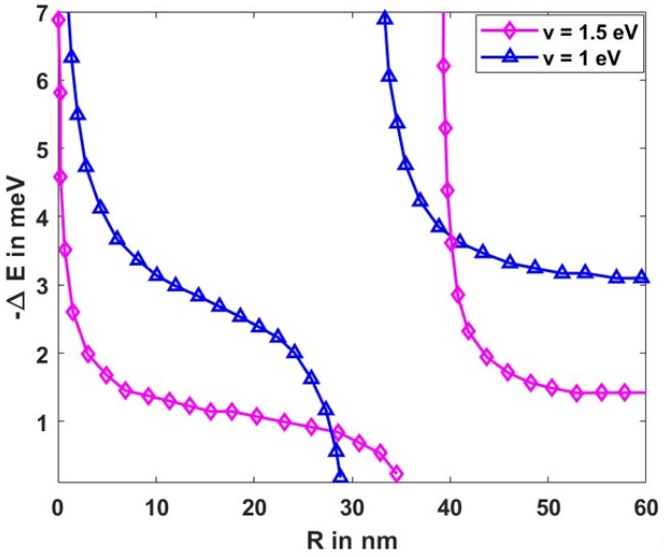

FIG. 1. Polaronic correction ΔE (in meV) to the first ES energy of an electron in GQD of GaAs with respect to R (in nm)

3. Conclusion

In conclusion, we have studied the effect of electron-LO-phonon interaction on the first ES energy level of an electron in a 3D GQD using second order RSPT. We have obtained an analytical expression for the first ES polaronic energy under a plausible approximation. Since we are interested in a realistic material, we have applied our results to a GaAs polar semiconductor QD. We have shown how the polaronic correction to the first ES energy of an electron in a GQD of GaAs behaves as a function of the effective confinement length for two values of the potential strengths. The energy curves show the singularity behaviour that corresponds to the instability of the first ES electron to the emission of an LO phonon.

Our results show that the polaronic effect to the electron in the ES of a GaAs GQD can be significantly large if the size of the QD is small. Since the information of the energies and wave functions of the excited state is important for the study of decoherence phenomena particularly the decoherence time, our results could be useful for quantum information processing.

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