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Polaron exciton in spherical quantum dot

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Abstract. The electron and hole polaron energies are found in a quantum dot made of the materials with high ionicity. It is shown that the energy of hole polaron is larger then the energy of electron polaron due to the degeneration of the valence band. Polaron energies increase with decreasing of the quantum dot radius. In the interband optical transitions, polaron effects are partly compensated, because electron and hole create the polarization potential wells with opposite signs in the process of optical transition. It is shown that there is no total compensation when the degeneration of the valence band structure is taken into account. Therefore the interband transitions are accompanied by the polarization of the medium. The polarization leads to the intensive phonon replicas of the electron transition and to the large Stokes shift of absorption and emission light.

Localization of charge particles in quantum dot results in considerable increase in electrostatic energy of particle interaction. Since longitudinal optical phonons in ionic crystals have also electrostatic nature, the enhancement of the electron–phonon interaction in nanostructures occurs. As a result, polaron effects increase also [1, 2].

If the polaron binding energy is less than the energy of size quantization in the dot, one has the strong confinement regime [3] when

$$\frac{a_0}{R} \gg 1. \tag{1}$$

Here a_0 is the polaron radius and R is the radius of spherical quantum dot.

The binding energies of electron and hole polarons and polaron exciton can be found by adiabatic approach based on parameter (1).

In zero approximation, the electron wave function and energy are defined by Schrödinger equation

$$\hat{H}_{\mathrm{e}}\Psi_{\mathrm{e}}^{(ln)} = \left[-\frac{\hbar^2}{2m_{\mathrm{e}}}\nabla^2 + V_{\mathrm{e}}(\mathbf{r})\right]\Psi_{\mathrm{e}}^{(ln)} = E_{\mathrm{e}}\Psi_{\mathrm{e}}^{(ln)}, \tag{2}$$

where $V_{\mathrm{e}}(r)$ is the quantum dot potential energy for electron, l is the electron orbital quantum number and n is the radial quantum number. Since the energy in Eq. (2) does not depend on magnetic quantum number m, we do not label the wave function with m for simplicity.

The description of the hole is based on the Luttinger Hamiltonian

$$\hat{H}_{\mathrm{h}}\Psi_{\mathrm{h}}^{(FN)} = \left[\left(\gamma_1 + \frac{5}{2}\gamma\right)\frac{\hat{p}^2}{2m_0} - \frac{\gamma}{m_0}(\hat{p}\hat{j}) + V_{\mathrm{h}}(\mathbf{r})\right]\Psi_{\mathrm{h}}^{(FN)} = E_{\mathrm{h}}\Psi_{\mathrm{h}}^{(FN)}. \tag{3}$$

Here $V_{\mathrm{h}}(\mathbf{r})$ is quantum dot potential energy for hole,

$$\gamma_1 = \frac{m_0}{2}\left(\frac{1}{m_l} + \frac{1}{m_{\mathrm{h}}}\right), \qquad \gamma = \frac{m_0}{4}\left(\frac{1}{m_l} - \frac{1}{m_{\mathrm{h}}}\right),$$

m_l and m_h being light and heavy hole masses. The quantity j is the effective "spin" of the hole which is equal to 3/2 for typical semiconductors. [Spin–orbit interaction is not taken into account]. The hole wave functions are classified according to total angular momentum $\hat{F} = \hat{l} + \hat{j}$, where $\hat{l}$ is the orbital momentum of the hole. The quantity N is the radial quantum number.

Polaron states of the electron and the hole are described by the following Schrödinger equation

$$\left[\hat{H}_{\mathrm{e,h}} + \sum_q \hbar\omega_q a_q^+ a_q + e\sqrt{\frac{2\pi\hbar}{V\varepsilon}} \sum_q \sqrt{\omega_q}\frac{1}{q}\left(a_q e^{i\mathbf{qr}} + a_q^+ e^{-i\mathbf{qr}}\right)\right]\Psi_{\mathrm{e,h}} = E\Psi_{\mathrm{e,h}}. \quad (4)$$

Here a_q, a_q^+ are the annihilation and creation phonon operators, $\varepsilon^{-1} = \varepsilon_0^{-1} - \varepsilon_\infty^{-1}$ is the optical dielectric permittivity, ω_q is the frequency of longitudinal optical phonon. The second term in Eq. (4) represents the phonon field and the third term is the electron–phonon interaction with longitudinal optical phonons. We consider the electron–phonon interaction to be strong. All three terms in Hamiltonian Eq. (4) have the same order of magnitude (Pekar polaron).

Adiabatic parameter Eq. (1) allows to make an average of Eq. (4) over the fast motion of the electron or hole in the quantum dot. One has for the electron Hamiltonian

$$\hat{H}_{\mathrm{e}}^{(ln)} = E_{ln} + \sum_q \hbar\omega_q a_q^+ a_q + e\sqrt{\frac{2\pi\hbar}{V\varepsilon}} \sum_q \frac{\sqrt{\omega_q}}{q}\left[\rho^{(ln)}(q) a_q + \rho^{(ln)} * (q) a_q^+\right], \quad (5)$$

where $\rho^{(ln)}(q) = \int e^{i\mathbf{qr}}\left[\Psi^{(ln)}(\mathbf{r})\right]^2 d^3r$ is electron density. The unitarian transformation

$$U_{\mathrm{e}}^{(ln)} = \exp\left[\sum_q \frac{e}{q}\sqrt{\frac{2\pi}{V\varepsilon\hbar\omega_q}}\left(\rho^{(ln)}(q) a_q^+ - \rho^{(ln)} * (q) a_q\right)\right], \quad (6)$$

allows to make the diagonalization of matrix $\hat{H}_{\mathrm{e}}$ for Eq. (5)

$$\hat{H}_{\mathrm{e}}^{(ln)} = E_{ln} - \frac{2\pi e^2}{V\varepsilon}\sum_q \frac{\left|\rho^{(ln)}(q)\right|^2}{q^2} + \sum_q \hbar\omega_q a_q^+ a_q. \quad (7)$$

Second term in Eq. (7) is the polaron renormalization of the electron energy

$$\triangle E_{\mathrm{e}}^{(ln)} = -\frac{2\pi e^2}{V\varepsilon}\sum_q \frac{\left|\rho^{(ln)}(q)\right|^2}{q^2}. \quad (8)$$

Further calculations require the knowledge of electronic wave functions $\Psi^{(ln)}(\mathbf{r})$. They were obtained by Al. Efros and A. Efros [3]

$$\Psi_{\mathrm{e}}^{(ln)} = \frac{J_l[C_n(l)r/R]Y_{lm}(\theta,\phi)}{\sqrt{J_{l+1}[C_n(l)]}}, \quad (9)$$

where $C_n(l)$ is nth root of lth spherical Bessel function $J_l(x)$, $Y_{lm}(\theta,\phi)$ is spherical function. Substituting the wave function Eq. (9) for spherical states with $l = 0$ in Eq. (8) results in

$$\triangle E_{\mathrm{e}}^{(0n)} = -\frac{e^2}{2\varepsilon R}2\left(1 - \frac{Si(2n\pi)}{2n\pi} + \frac{Si(4n\pi)}{4n\pi}\right), \quad (10)$$

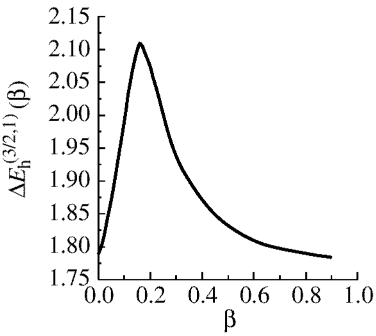

Fig. 1. The dependence of hole polaron energy on the mass ratio $\beta = m_l/m_{\rm h}$.

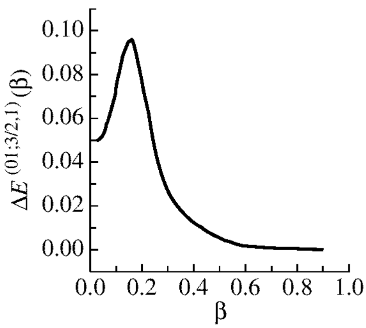

Fig. 2. The dependence of exciton polaron energy on the mass ratio $\beta = m_l/m_{\rm h}$.

where $Si(x)$ is the integral sinus.

The polarization energy of the hole polaron $\triangle E_{\rm h}^{(FN)}$ is calculated in a similar way from Eqs. (3) and (4). The wave function of the hole in the spherical dot is taken from [4]. The dependence of the hole ground state energy om the ratio of the light and heavy hole masses, $\beta = m_l/m_{\rm h}$, is shown in Fig. 1.

The electron and the hole create in the interband optical transition their own potential polarization wells of opposite sign. Nevertheless, the degeneration of the hole band prevent the compensation of polaron effects. There appear the polarization quasiparticle which is called *polaron exciton* [5].

Under condition of strong confinement Eq. (1), the Coulomb interaction of the electron and the hole and their interaction with polar optical phonons are small with respect to the energy of size quantization. The wave function of the electron–hole pair is reduced to the product

$$\Psi(\mathbf{r}_{\rm e}, \mathbf{r}_{\rm h}) = \Psi_{\rm e}^{(ln)}(\mathbf{r}_{\rm e})\Psi_{\rm h}^{(FN)}(\mathbf{r}_{\rm h}), \tag{11}$$

where wave functions $\Psi_{\rm e}^{(ln)}(\mathbf{r}_{\rm e})$ and $\Psi_{\rm h}^{(FN)}(\mathbf{r}_{\rm h})$ are defined by Eqs. (2) and (3), respectively.

Taking an average over the fast motion of the electron and the hole in a quantum dot and using the wave functions from [3, 4] one can find the polaron exciton energy

$$\triangle\, E^{(ln,FN)} = -\frac{e^2}{2\varepsilon R} B^{(ln,FN)}(\beta), \tag{12}$$

where dimensionless coefficients $B^{(ln,FN)}(\beta)$ depends on the ratio of light and heavy hole masses β. This dependence for optical transition between the electron ground state $(l = 0, n = 1)$ and the hole ground state $(F = 3/2, N = 1)$ is shown in Fig. 2. It is follows from Eq. (12) that exciton polaron energy $\triangle E^{(ln,FN)}$ decreases with R.

Polaron exciton in quantum dots manifest itself in multiple phonon replicas of the same intensity as electronic zero–phonon line and in the strong Stokes shift between the absorption and emission lines. Both effects were observed experimentally for A^2B^6 quantum dots in glassy matrix [6, 7].

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