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The spectral distribution of polaron exciton line in quantum dot

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Abstract. The intensity of polaron exciton absorption in spherical quantum dot is calculated. The localized electrons and holes are shown to interact with long wavelength phonons with wave vectors $q \approx 1/R$, R being the radius of the dot. The broadening of different phonon replicas are shown to be less then their separation

Compounds II–VI are good semiconductor system suitable for fabrication of blue–green lasers [1]. Since II–VI materials have high ionicity, it is natural to expect the existence of electron and hole polaron states [2].

The electron and hole polaron effect in nanostructures has been considered in [3, 4] using Kane model and Luttinger Hamiltonian approximation. It is shown that hole polaron polarization effect differ from electron polarization effect because of the degeneration of hole bands.

The paper deals with the theory of optical transition in ionic crystal were initial and final states of the interband transitions are electron and the hole polarons, respectively. The Coulomb interaction of the electron and the hole is taken into account in optical transition. It is accepted that the binding energy of polaron exciton is smaller then the size quantization energies of both the electron and the hole. It means that the polaron exciton radius $R_{\rm ex}$ is larger then the quantum dot radius R (strong confinement regime):

$$\frac{R_{\rm exc}}{R} \gg 1. \tag{1}$$

When the light is absorbed, there is a photon in the initial state, there are neither electrons nor holes yet. The wave function of the initial state is approximated by

$$\Psi^{(i)} = \delta(\mathbf{r}_{\rm e} - \mathbf{r}_{\rm h})\chi^{(i)}_{\rm lattice}. \tag{2}$$

Here $\mathbf{r}_{\rm e}$, $\mathbf{r}_{\rm h}$ are radius vectors of the electron and the hole, respectively, $\chi^{(i)}_{\rm lattice}$ is the lattice vibrations wave function. In the final state of the optical transition, there are the electron and the hole which interact one with another and with polar optical phonons. The wave function of the exciton final state in the strong confinement limit Eq. (1) is

$$\Psi^{(f)} = \psi_{\rm e}(\mathbf{r}_{\rm e})\psi_{\rm h}(\mathbf{r}_{\rm h})\chi^{(f)}_{\rm lattice}. \tag{3}$$

Since the quantum dot is considered here as a macroscopic conglomerate, the electron wave function $\psi_{\rm e}(\mathbf{r}_{\rm e}) = \psi_n(\mathbf{r}_{\rm e})u_c(\mathbf{r}_{\rm e})$ is the product of convolution $\psi_n(\mathbf{r}_{\rm e})$ by the electron modulating Bloch wave function $u_c(\mathbf{r}_{\rm e})$. The function $\psi_{\rm h}(\mathbf{r}_{\rm h}) = \psi_N(\mathbf{r}_{\rm h})u_v(\mathbf{r}_{\rm h})$ is the hole wave function where $\psi_N(\mathbf{r}_{\rm h})$ is the solution of Schrödinger equation with Luttinger Hamiltonian and $u_v(\mathbf{r}_{\rm h})$ is the hole Bloch modulating wave function. The vibrational wave functions $\chi^{(f)}_{\rm lattice}$ correspond to normal phonon modes with shifted equilibrium positions with respect to $\chi^{(i)}_{\rm lattice}$, because the different levels of the electron and the hole size quantization create different polarizations of the medium.

The rate of the absorption of a photon in the dipole approximation

$$W_{nN}^{abs} = \frac{2\pi}{\hbar} \mid P_{cv} \mid^2 \mid I_{nN} \mid^2 \sum_f W_{if} \delta(E_{nN} - E_f - \hbar\omega). \tag{4}$$

Here P_{cv} is the interband transition matrix element over Bloch modulating function, I_{nN} is the overlapping integral of the electron and the hole wave functions

$$I_{nN} = \int d^3 r \psi_n(\mathbf{r}) \psi_N(\mathbf{r}) \tag{5}$$

and

$$W_{if} = \left| \int dx_{q1} dx_{q2} \ldots \chi^{(i)}_{\text{lattice}}(x_{q1} x_{q2} \ldots) \chi^{(f)}_{\text{lattice}}((x_{q1} - s_{q1})(x_{q2} - s_{q2}) \ldots) \right|^2 \tag{6}$$

is the overlapping integral of lattice vibration wave functions. The polarization shift of the normal mode equilibrium positions s_q has been calculated in [4]. Both the electron and the hole polarization effects contribute to the shift

$$s_q = \frac{e}{\omega(q) q} [\rho_N(q) - \rho_n(q)] \sqrt{\frac{2\pi}{M_0 V \varepsilon}}. \tag{7}$$

Here M_0 is the oscillator mass, V is the volume of the crystal, ε is the optical dielectric constant, $\omega(q)$ is the phonon frequency with the wave vector q,

$$\rho_n(q) = \int d^3 \mathbf{r}_e e^{i\mathbf{q}\mathbf{r}_e} \psi_n^2(\mathbf{r}_e), \tag{8}$$

$$\rho_N(q) = \int d^3 \mathbf{r}_h e^{i\mathbf{q}\mathbf{r}_h} \psi_N^2(\mathbf{r}_h), \tag{9}$$

are the electron and the hole densities.

Equation (6) can be rewritten in the form of the product of the transition rates w_q for each normal mode of lattice vibrations:

$$W_{if} = \prod_q w_q \tag{10}$$

where q numerates normal modes of the lattice vibrations and

$$w_q = \left| \int dx_q \chi^{(i)}_{\text{lattice}}(x_q) \chi^{(f)}_{\text{lattice}}(x_q - s_q) \right|^2. \tag{11}$$

Since equilibrium positions in integral (11) are shifted, there is the finite probability of transition from initial to final state with emission of arbitrary number of phonons.

The rate of the transition from the ground state of oscillator into the excited state with the emission of K_q phonons of q-th normal mode with the frequency $\omega(q)$ is equal to [4]

$$w_q(K_q) = \frac{\left(\mid s_q \mid^2 \frac{M_0 \omega(q)}{\hbar} \right)^{K_q}}{2^{K_q} (K_q)!} \exp\left[-\frac{1}{2} \mid s_q \mid^2 \frac{M_0 \omega(q)}{\hbar} \right]. \tag{12}$$

The equation (12) is the Poisson distribution of spectral lines corresponding to different number of emitted phonons with the frequency $\omega(q)$. Each spectral line is not broadened.

The vibrational spectrum of the crystal consists of many normal modes with different $\omega(q)$. The total set of normal modes contributes to the probability Eq. (4).

The transition rate Eq. (4) contains in the lowest dipole approximation the succession of multi-phonon transitions (peaks) each of which corresponds to the different number K of emitted phonons. The number K is obtained by the selection from the total set of normal modes only those which satisfy the condition

$$K = \sum_q K_q. \tag{13}$$

Each phonon replica K is broadened due to the dispersion of optical branches. The quantity K takes the values $K = 0, 1, 2, 3, \ldots$ where $K = 0$ is the zero-phonon line, $K = 1$ is the process with the participation of the single phonon, $K = 2$ is one with participation of two phonons and so on.

It is necessary to find condition under which the polaron exciton optical spectrum still has the form of well seen succession of lines corresponding to multi-phonon replicas. The electron density $\rho(q)$ is not zero inside of the quantum dot only. Therefore, electrons and holes interact with long wavelength polar optical phonons with the wave vector $q \approx 1/R \ll \pi/a$, a being the lattice parameter. In terms of K, the rate of polaron exciton absorption takes the form:

$$W_{nN}^{\text{abs}} = \frac{2\pi}{\hbar} \mid P_{cv} \mid^2 \mid I_{nN} \mid^2 \sum_K \sum_{q_1 q_2 \ldots q_K} \left(\frac{4\pi e^2}{\varepsilon V}\right)^K W_K(q_1, q_2, \ldots q_K)$$
$$\times \delta(E_{nN} + \hbar\omega(q_1) + \hbar\omega(q_2) + \ldots + \hbar\omega(q_K) - \hbar\omega), \tag{14}$$

where

$$W_K(q_1 \ldots q_K) = \frac{\mid [\rho_N(q_1) - \rho_n(q_1)][\rho_N(q_2) - \rho_n(q_2)] \ldots [\rho_N(q_K) - \rho_n(q_K)] \mid^2}{\hbar\omega(q_1)\hbar\omega(q_2) \ldots \hbar\omega(q_K) q_1^2 q_2^2 \ldots q_K^2}$$
$$\times \exp\left[-\frac{S_0}{2}\right]. \tag{15}$$

Here S_0 is equal to

$$S_0 = \sum_q \frac{4\pi e^2}{\varepsilon V} \mid \rho_N(q) - \rho_n(q) \mid^2 \frac{1}{\hbar\omega(q) q^2} \tag{16}$$

Notice that the summation index q in Eq. (16) means the phonon wave vector and the lable of the optical branch. When $K = 0$, we find from Eq. (14) the intensity of the zero-phonon line

$$W_0 = \exp\left[-\frac{S_0}{2}\right], \tag{17}$$

which is not broadened. When $K = 1$, the intensity of the one-phonon replica $I_1(q)$ has been calculated numerically for the model phonon spectrum $\omega(q) = \omega_0 - \alpha q^2$, where

$$\alpha = \left.\frac{\partial^2 \omega(q)}{\partial q^2}\right|_{q \leq 1/R}.$$

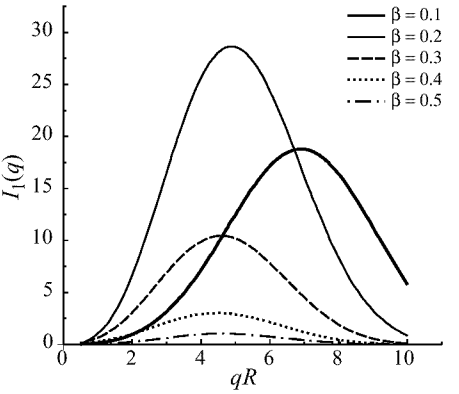

Fig. 1. The spectral distribution of polaron exciton lines of the first phonon replica for the different values of light and heavy hole masses $\beta = m_l/m_h$.

The wave functions $\psi_n(\mathbf{r}_e)$ and $\psi_N(\mathbf{r}_h)$ are taken to be solutions of Schrödinger equation for the spherical quantum dot with infinite walls [5, 6]. The result of calculations is shown in Fig. 1 for different values of the ratio of the light and heavy hole masses β. The broadening of the one-phonon replica is shown to be less then the separation of peaks.

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