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Optical spectra of quantum dots: A non-adiabatic approach

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Abstract

The presence of accompanying resonances to the longitudinal optical phonon satellites in the optical spectra of semiconductor quantum dots is confirmed theoretically by a non-adiabatic approach of the optical absorption. The theory is applied to simulate features of the optical spectra of small spherical GaAs/AlAs quantum dots. The intensity and the spectral position of the accompanying resonances are influenced by both optically active and dark levels and increase to measurable values with temperature.

Experimentally, in the optical spectra of quantum dots (QDs), besides the longitudinal optical (LO) phonons satellites, accompanying resonances have been observed as well (see, e.g. [1,2]). Theoretical analyses within the adiabatic approximation for defect-free QDs yield Huang-Rhys (HR) factors which are one to four orders of magnitude smaller than observation [3,4]. Also, a red shift with temperature of the 0LO phonon line in the photoluminescence spectra of QDs is observed by experiment [5].

To offer an explanation to the mentioned experimental observations, in this Letter, we consider a non-adiabatic approach of the optical absorption. Non-adiabatic treatments, necessary when the electron–hole pair (EHP) level spacing is comparable to the LO phonon energy, have recently been proposed [6–9]. Unlike the study from [6], our approach is able to predict the resonances accompanying the LO satellites. Moreover, technically, instead of evaluating the intensity lines from the oscillator strength using the eigenstates of the EHP–phonon Hamiltonian with a Fröhlich coupling at temperature $T = 0$ K, as in [8], the temperature effect in the absorption spectra is obtained by averaging over the equilibrium phononic states. Our treatment, in principle, is not limited to a certain number of electronic levels. Unlike the analysis from [9], where two electronic levels are considered, the theory is applied to find the absorption coefficient for three EHP levels coupled by phononic modes. Firstly, a general expression of the linear absorption coefficient is derived by applying a semiclassical theory of the radiation-matter interaction to the EHP–LO phonons system. Secondly, we consider a spherical geometry, the effective mass approximation and pure EHP (the case of small QDs, where the Coulomb electron–hole interaction may be neglected [10]). The Fröhlich coupling of the EHP–phonon system is chosen. The charge separation of the electron and hole appears as a result of using finite confinement potentials for the QD. The selection rules are derived. Thirdly, with the calculated EHP–phonon coupling matrix elements (CME), the absorption spectra are simulated for GaAs/AlAs QDs.

The Hamiltonian of the EHP–phononic reservoir is described by an extension of the Huang-Rhys model of F centers

$$H = H_{\mathrm{EHP}} + H_{\mathrm{ph}} + H_{\mathrm{EHP-ph}} \tag{1}$$

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with $H_{\rm EHP} \equiv \sum_f E_f B_f^+ B_f$, $H_{\rm ph} \equiv \sum_{\mathbf{q}} \hbar\omega_{\mathbf{q}} b_{\mathbf{q}}^+ b_{\mathbf{q}}$, $H_{\rm EHP\text{–}ph} \equiv \sum_{\mathbf{q},f,f'} M_{\mathbf{q}}^{ff'} B_f^+ B_{f'}(b_{\mathbf{q}} + b_{-\mathbf{q}}^+)$, and B_f^+ (B_f), $b_{\mathbf{q}}^+$ ($b_{\mathbf{q}}$) are the bosonic creation (annihilation) operators of the EHP and phonons, respectively, $M_{\mathbf{q}}^{ff'} \equiv \langle f|M_{\mathbf{q}}|f'\rangle$ the CME, $\omega_{\mathbf{q}}$ the frequency of the phononic mode with wave vector $\mathbf{q}$, and $E_f(|f\rangle)$ the EHP eigenvalues (eigenstates). The radiation field is modeled as a single mode of linearly polarized plane wave. Within the semiclassical theory, in the limit of the linear response theory and long-wave approximation, the absorption coefficient can be written as [11]

$$\alpha(\omega) = \frac{2\pi e^2}{ncm_0^2\hbar\omega V_0} \sum_{f,f'\neq 0} \left[P_{0f} P_{f'0} \int_{-\infty}^{\infty} \mathrm{d}t \exp[\mathrm{i}(\omega - \omega_f)t] \times \langle 0|B_f \left\langle \hat{T} \exp\left[-\frac{\mathrm{i}}{\hbar} \int_0^t \mathrm{d}t_1 \tilde{V}(t_1) \right] \right\rangle_0 B_{f'}^+ |0\rangle \right], \quad (2)$$

where $\hat{T}$ is the time-ordered operator, $\tilde{V}(t) = \exp(\mathrm{i}tH_0/\hbar) H_{\rm EHP\text{–}ph} \exp(-\mathrm{i}tH_0/\hbar)$, $\langle \hat{T} \exp[-\frac{\mathrm{i}}{\hbar}\int_0^t \mathrm{d}t_1 \tilde{V}(t_1)]\rangle_0 \equiv \langle U(t)\rangle_0$ and $\langle \ldots \rangle_0$ denotes an average over the phononic system at thermal equilibrium, $|0\rangle$ the EHP vacuum state, $H_0 \equiv H_{\rm EHP} + H_{\rm ph}$, V_0 the volume of absorptive system, c the speed of light in vacuum, n the refractive index, m_0, e the mass and the charge of electron, ε and ω, the polarization vector and the frequency of light wave, and $P_{0f} \equiv \langle 0|(\varepsilon \cdot \mathbf{P})|f\rangle$, $\mathbf{P} \equiv \sum_i \mathbf{p}_i$ the total electronic momentum (with $\mathbf{p}_i$ the electron momentum). For the linear phononic coupling in Eq. (1), the cumulant expansion method gives the result that all cumulants higher than the second order vanish. Thus, by using the bosonic commutation rules one obtains

$$\langle U(t)\rangle_0 = \exp\left[-\frac{1}{\hbar^2} \sum_{\mathbf{q},i_1,j_1,i_2,j_2} M_{\mathbf{q}}^{i_1 j_1} M_{-\mathbf{q}}^{i_2 j_2} B_{i_1}^+ B_{j_1} B_{i_2}^+ B_{j_2} \times \int_0^t \mathrm{d}t_1 \int_0^{t_1} \mathrm{d}t_2 \exp(\mathrm{i}t_1\omega_{i_1 j_1}) \exp(\mathrm{i}t_2\omega_{i_2 j_2}) D^0 \times (\mathbf{q}, t_1 - t_2) \right] \equiv \exp(S), \quad (3)$$

where $D^0(\mathbf{q}, t_1 - t_2) \equiv [\bar{N}_{\mathbf{q}} \exp(\mathrm{i}\omega_{\mathbf{q}}(t_1 - t_2)) + (\bar{N}_{\mathbf{q}} + 1) \exp(-\mathrm{i}\omega_{\mathbf{q}}(t_1 - t_2))]$, $\omega_{ij} = (E_i - E_j)/\hbar$, and $\bar{N}_{\mathbf{q}}$ is the thermally averaged phononic occupation number of the $\mathbf{q}$ mode; the integral is not time-ordered. Using the series expansion of the exponential in Eq. (3), we find

$$\langle 0|B_f S^n B_{f'}^+|0\rangle = \sum_{i_3,i_5,\ldots,i_{2n-1}} \sum_{i_2} (f i_2 i_2 i_3) \sum_{i_4} (i_3 i_4 i_4 i_5) \cdots \sum_{i_{2n}} (i_{2n-1} i_{2n} i_{2n} f') \overset{\text{cutting}}{\rightarrow} \left[\sum_i (f i i f') \right]^n, \quad (4)$$

where $\sum_{k'} (i_k i_{k'} i_{k'} i_{p'}) \equiv \sum_{\mathbf{q},k'} [M_{\mathbf{q}}^{kk'} M_{-\mathbf{q}}^{k'p'} \int_0^t \mathrm{d}t_1 \int_0^{t_1} \mathrm{d}t_2 \exp(\mathrm{i}t_1\omega_{kk'}) \exp(\mathrm{i}t_2\omega_{k'p'}) D^0(\mathbf{q}, t_1 - t_2)] \equiv \sum_{\mathbf{q},k'} [M_{\mathbf{q}}^{kk'} M_{-\mathbf{q}}^{k'p'} I(\mathbf{q}, t, k, k', k', p')]$. A partial ressumation (PR) of $\langle 0|B_f S^n B_{f'}^+|0\rangle$ is obtained by the 'cutting' procedure suggested in Eq. (4). Hereafter, we consider a dispersionless phononic bath (Einstein model) of frequency ω_0 and introduce notations as follows: $I(\mathbf{q}, t, k, k' p, p') \rightarrow I(t, k, k', p, p')$, $\bar{N}_{\mathbf{q}} \rightarrow \bar{N}$, $g_{kk'pp'} \equiv \sum_{\mathbf{q}} [M_{\mathbf{q}}^{kk'} M_{-\mathbf{q}}^{pp'} / (\hbar\omega_0)^2]$ ($g_{pppp} \equiv g_p$ is the HR factor), $G_{piis} \equiv g_{piis} \omega_0^2 I(t, p, i, i, s)$. The PR yields

$$\alpha(\omega) = \frac{2\pi e^2}{ncm_0^2\hbar\omega V_0} \sum_{p,s\neq 0} \left[P_{0p} P_{s0} \int_{-\infty}^{\infty} \mathrm{d}t \exp[\mathrm{i}(\omega - \omega_p)t] \times \exp\left(-\sum_{i\neq 0} G_{piis} \right) \right]. \quad (5)$$

Unlike the case presented in [6], the CME does not have to respect the restriction, $\max|M_{\mathbf{q}}^{ff'}| \ll 1$, which may be difficult to fulfill for large $|g_{kk'pp'}|$ as long as few LO phononic modes are strongly coupled with the EHPs see, e.g. [9]. If the off-diagonal coupling terms in Eq. (1) are disregarded then Eq. (5) is exact and it *recovers* the adiabatic limit (the Franck–Condon progression)

$$\alpha^{\rm ad}(\omega) = \frac{4\pi^2 e^2}{ncm_0^2\hbar\omega V_0} \sum_{f\neq 0} \left\{ |P_{0f}|^2 \exp\left[-g_f(2\bar{N} + 1) \right] \times \sum_{n\neq 0}^{\infty} I_n\left(2g_f \sqrt{\bar{N}(\bar{N} + 1)} \right) \exp(n\beta\hbar\omega_0/2)\delta \times \left(\omega - \omega_f + \Delta_f^{\rm ad} - n\omega_0 \right) \right\}, \quad (6)$$

where I_n are the modified Bessel functions, and $\Delta_f^{\rm ad} = \omega_0 \sum_{\mathbf{q}} (|M_{\mathbf{q}}^{ff}|^2 \hbar^{-2} \omega_0^{-2}) \equiv \omega_0 g_f$ is the self-energy. The relative intensity of absorption lines is given by the coefficients of the Dirac delta functions.

The one-EHP energy spectrum has been computed for spherical GaAs microcrystallites embedded in AlAs matrix in [11]. The optical selection rules are dictated by the matrix element $\langle 0|\mathbf{P}|f\rangle$. With an appropriate definition of the momentum $\mathbf{P}$ [12], one obtains $\langle \varphi_{ab}|\mathbf{P}|0\rangle = \mathbf{p}_{cv}^0 \delta_{l_e l_h} \delta_{m_e m_h} \int_0^{\infty} \mathrm{d}r r^2 R_{n_e l_e}(r) R_{n_h l_h}(r) \equiv \mathbf{p}_{cv}^0 \delta_{m_e m_h} A_{n_e n_h l}$, with $l_e = l_h = l$. The index $a(b)$ holds for the set of quantum numbers n_e, l_e, m_e (n_h, l_h, m_h) and $\varphi_{nlm}(\mathbf{r}) = R_{nl}(r) Y_{lm}(\theta, \varphi)$, where $R_{nl}(r)$ is the normalized radial function (spherical or modified spherical Bessel functions) and $Y_{lm}(\theta, \varphi)$ the spherical harmonics (see [11]). Thus, the optical selection rule allows optical transitions between states with $l_e = l_h$ and $m_e = m_h$. The quantity $P_{0f} P_{f'0} = |\mathbf{p}_{cv}^0|^2 3^{-1} A_{n_e n_h l} A_{n'_e n'_h l'} \delta_{m_e m_h} \delta_{m'_e m'_h}$ in Eq. (2) is averaged over polarization directions. The Fröhlich coupling is written for dispersionless bulk LO phonons (for a QD with high geometrical symmetry, the interface modes are usually weak [13]). Then the CME reads

$$M_{\mathbf{q}}^{ff'} \to M_{\mathbf{q}}^{ab;a'b'} = V_0^{-1/2} f_0 q^{-1} \left[\delta_{bb'} \int_0^\infty \mathrm{d}r\, r^2 R_{n_e l_e}(r) R_{n'_e l'_e}(r) Q_{l'_e m'_e}^{l_e m_e}(q,r) - \delta_{aa'} \int_0^\infty \mathrm{d}r\, r^2 R_{n_h l_h}(r) R_{n'_h l'_h}(r) Q_{l'_h m'_h}^{l_h m_h}(q,r) \right], \quad (7)$$

where

$$Q_{l'm'}^{lm}(q,r) \equiv \int \mathrm{d}\Omega\, Y_{lm}^*(\Omega) Y_{l'm'}(\Omega) \exp(\mathrm{i}\mathbf{q}\mathbf{r}) = \sqrt{\frac{2l'+1}{2l+1}} \delta_{mm'} \sum_{\substack{l''=|l-l'| \\ l''+l'+l\to\text{even}}}^{l+l'} i^{l''}(2l''+1) \times \langle l''l'00|l0\rangle \langle l''l'0m'|lm\rangle j_{l''}(qr)$$

with $j_l(x)$ the spherical Bessel functions, $\langle l''l'm''m'|lm\rangle$ the Clebsh–Gordan coefficients, and f_0 the Fröhlich coupling constant. Thus, the states $\varphi_{ab}, \varphi_{a'b'}$ are not coupled by phonons if both $a \neq a'$ and $b \neq b'$.

Applying the theory, we consider a rather rarefied distribution of energy levels, as is generally the case in small spherical GaAs/AlAs QDs [11]. Large values of $|g_{kk'pp'}|$ obtained for such spherical-shaped nanostructures less polar than CdSe, e.g., are the result of charge separation induced by the finite confinement potential (not considered for the CdSe QDs embedded in glass in [6]). For such large values of $|g_{kk'pp'}|$ and consequently, for large HR factors, the present theoretical approach is useful.

Thus, following [11] for a QD with radius $R_0 = 20$ Å, there are only two levels, both optically active, that we denote as $1 \equiv \mathsf{A}_1 \to (1,0,0,1,0,0)$ and $2 \equiv \mathsf{A}_2 \to (1,0,\ 0,2,0,0)$; their separation is approximately $11\hbar\omega_0$ (the energy of LO phonons is considered as $\hbar\omega_0 = 36.2$ meV). On the other hand, for $R_0 = 32$ Å, between the lowest two optical levels A_1 and A_2 (with separation of approximately $5.3\hbar\omega_0$), there is a dark level, namely, $3 \equiv \mathsf{D}_1 \to (1,0,0,1,1,m_h)$ situated at approximately $1.86\hbar\omega_0$ from A_1. For D_1 one considers $m_h = 0$, the only CME given by Eq. (7) that does not vanish. The other levels are situated far enough not to influence significantly the spectra. Considering only two optical levels, the absorption spectrum in Eq. (2) is written as

$$\alpha(\omega) = \frac{2\pi e^2 |\mathbf{p}_{cv}^0|^2}{3ncm_0^2\hbar\omega V_0} \sum_{i=1,2} \left\{ \int_{-\infty}^{\infty} \mathrm{d}t \exp[\mathrm{i}(\omega-\omega_i)t] \times \left[A^2_{n_e^{(i)} n_h^{(i)} l^{(i)}} \sum_{f=1,2} \langle i|\langle U(t)\rangle_0|i\rangle + A_{n_e^{(1)} n_h^{(1)} l^{(1)}} A_{n_e^{(2)} n_h^{(2)} l^{(2)}} \times \sum \langle i|\langle U(t)\rangle_0|f\rangle \right] \right\}. \quad (8)$$

For $R_0 = 20$ Å, the first term in the integrand of Eq. (8) may be approximated by using Eq. (5) (approximation (i)). Moreover, the second (mixing) term of the integrand of Eq. (8), responsible for the phonon mixing of different optically active states, is negligible (approximation (ii)). The neglected first-order term in approximation (i), namely, $(G_{1112} + G_{1222})(G_{2111} + G_{2221})$, yields linear combinations of exponentials of the form $\exp[(n\omega_{21} + m\omega_0)t]$, where n, m are integers. The coefficients of these exponentials are small quantities related to ω_0/ω_{21} or $\omega_0/(\omega_{21} \pm \omega_0)$ with power two or higher, thus the accuracy of approximation (i) is very good. To justify approximation (ii), let us consider expansion of $\langle 1|\langle U(t)\rangle_0|2\rangle$, for instance. The neglected first-order term obtained in the expansion is $G_{1112} + G_{1222}$.

It is also a linear combination of exponentials with the same form as above, which also have small coefficients. In addition, the contribution of the overlap integrals increases the accuracy of approximation (ii) ($|A_{120}/A_{110}| = 0.34$). Thus, from Eq. (8), we find an accurate approximation of the absorption spectrum, which for the lines centered on A_1 reads

$$\alpha_1(\omega) = \frac{2\pi e^2 |\mathbf{p}_{cv}^0|^2}{3ncm_0^2\hbar\omega V_0} A_{110}^2 \exp(-\Lambda_1) \times \sum_{p=-\infty}^{\infty} \sum_{k,r=0}^{\infty} \left[I_p\left(2g_1\sqrt{\bar{N}(\bar{N}+1)}\right) \left(\frac{\bar{N} g_{1221}\beta^2}{k!}\right)^k \times \left(\frac{(\bar{N}+1)g_{1221}\gamma^2}{r!}\right)^r \exp\left(\frac{-p\hbar\omega_0}{2k_BT}\right) \times \delta[\omega - \omega_1 + \Lambda_1 + p\omega_0 - k(\omega_{21}-\omega_0) - r(\omega_{21}+\omega_0)] \right], \quad (9)$$

where $\Lambda_1 = g_1(2\bar{N}+1) + g_{1221}[\bar{N}\beta^2 + (\bar{N}+1)\gamma^2]$, $\Delta_1 = \omega_0[g_1 + g_{1221}(\bar{N}\beta + (\bar{N}+1)\gamma)]$ is the *self-energy*, $\beta \equiv \omega_0/(\omega_{21}-\omega_0)$, and $\gamma \equiv \omega_0/(\omega_{21}+\omega_0)$. The non-adiabatic corrections are, as expected from the large inter-level energy, not significant and the non-adiabatic spectrum plotted with Eq. (9) is practically identical with the adiabatic spectrum obtained with Eq. (6) ($g_1 = 0.117$, $g_{1221} = 0.314$). The spectra, not plotted here show the ordinary phononic satellites.

For $R_0 = 32$ Å, considering the two optical levels A_1 and A_2, the same reasons as those employed for the case of $R_0 = 20$ Å also validate accurately approximations (i) and (ii) ($|A_{120}/A_{110}| = 0.21$, and β and γ are also smaller than one). On the other hand, the effect of the dark level D_1 is roughly estimated [14] by considering the PR procedure implied by Eq. (5). Thus, the contribution of the optical and dark levels to the absorption centered on line A_1 is described by an equation similar to Eq. (9), written to account for the dark level too

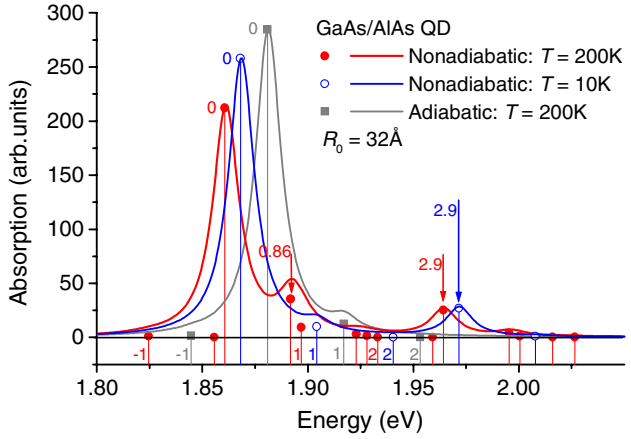

Fig. 1. The simulated absorption spectra of GaAs/AlAs nanocrystal QDs for different temperatures. The adiabatic spectrum obtained with Eq. (6) has no temperature-induced shift and its maxima are not significantly changed with temperature. The non-adiabatic spectra are obtained with Eq. (10). The following quantities obtained within the adopted QD model have been used: $E_1 = 1.8822$ eV, $E_2 = 2.0738$ eV, $E_3 = 1.9496$ eV, $g_1 = 0.039$, $g_{1221} = 0.234$, and $g_{1331} = 0.904$. The stronger accompanying resonances are marked by arrows. The energy of some resonances are indicated by factors which multiply the LO phonon energy; they are placed to the left of the lines or arrows.

$$\begin{aligned}\alpha_1(\omega) = {} & \frac{2\pi e^2|\mathbf{p}_{cv}^0|^2}{3ncm_0^2\hbar\omega V_0} A_{110}^2 \exp(-\Lambda_1) \\ & \times \sum_{p=-\infty}^{\infty} \sum_{k,r=0}^{\infty} \sum_{s,t=0}^{\infty} \left[I_p\left(2g_1\sqrt{\bar{N}(\bar{N}+1)}\right) \right. \\ & \times \left(\frac{\bar{N}g_{1221}\beta^2}{k!}\right)^k \left(\frac{(\bar{N}+1)g_{1221}\gamma^2}{r!}\right)^r \left(\frac{\bar{N}g_{1331}\bar{\beta}^2}{s!}\right)^s \\ & \times \left(\frac{(\bar{N}+1)g_{1331}\bar{\gamma}^2}{t!}\right)^t \exp\left(\frac{-p\hbar\omega_0}{2k_BT}\right) \\ & \times \delta[\omega - \omega_1 + \Delta_1 + p\omega_0 - k(\omega_{21} - \omega_0) \\ & \left. - r(\omega_{21} + \omega_0) - s(\omega_{31} - \omega_0) - t(\omega_{31} + \omega_0)] \right] \end{aligned} \tag{10}$$

with $\bar{\beta} \equiv \omega_0/(\omega_{31} - \omega_0)$ and $\bar{\gamma} \equiv \omega_0/(\omega_{31} + \omega_0)$. This non-adiabaticity effect as given by Eq. (10) is shown in Fig. 1, where the absorption spectra at different temperatures are plotted. The temperature dependence of the spectra, weak in the case of adiabatic treatment, becomes important now. Thus, decrease of intensity (by 37%) and red shift (from 1.87 to 1.85 eV) of the 0PL lines are obtained when temperature increases from 10 to 300 K. This agrees with the behavior observed experimentally for CdTe QDs [5]. On the other hand, the *simulated* HR factors (after we dressed the lines by Lorentzians with a finite width of 15 meV to simulate the EHP-acoustic phonons interaction) reach values larger by two orders of magnitude than those of the bulk phase (0.0079 in [15]). A similar behavior is reported, e.g., in [6] for spherical CdSe QDs and for small self-assembled InAs/GaAs QDs in [16]. Thus, by the non-adiabatic activated channel at +0.86LO, the simulated HR factor obtained as the ratio of the *dressed* line intensities for this accompanying resonance increases from 0.084 at $T = 10$ K to 0.23 at $T = 200$ K. The usual Franck–Condon progression is obtained by the adiabatic treatment (see the gray line in Fig. 1). On the other hand, the non-adiabaticity effect manifests by strong resonances at 2.9LO (see Fig. 1), close to the third LO phonon replica as reported by some experiments see, e.g. [17]. The validity of the cutting procedure for more polar semiconductors (CdSe QDs, e.g.) and consideration of the electron–acoustical phonons interaction (which must be taken into account when a theoretical fit of the experiment is the goal) are going to be discussed in another context.

In conclusion, in accordance with the experimental observation, the non-adiabatic approach predicts: (i) accompanying resonances to the LO phonon satellites in the optical spectra of QDs; (ii) a red shift of the 0LO phonon lines and increased intensities of the accompanying resonances with temperature in the absorption spectra of QDs.

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