

On Exciton Decoherence in Quantum Dots

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The effects resulting due to dressing of an exciton with phonons are analyzed as the source of unavoidable decoherence of orbital degrees of freedom in quantum dots. The dressing with longitudinal optical phonons results in energetic shift of order of a few meV even of the ground state of exciton in a state-of-the-art InAs/GaAs dot and the mediating role of longitudinal acoustical phonons is essential in this process. The characteristic time needed for dressing of the exciton with optical phonons is of a picosecond order. That time can be regarded as the lower limit for decoherence for optically driven quantum gates employing self-assembled quantum dot structures.

KEY WORDS: quantum dots; exciton–phonon interaction; polaron; decoherence.

1. INTRODUCTION

We consider an exciton, i.e. an electron-hole pair created in a quantum dot (QD) by extremely short optical pulse (Borri *et al.*, 2001; Kikkawa and Awschalom, 1988). The rapidly created bare exciton is further dressing with phonons, which results in arising a composite quasiparticle being the coherent mixture of the electron-hole pair and phonons. Its energy is lower than the bare exciton energy. In the case of interaction with only optical phonons this composite particle can be called exciton–polaron in analogy to the Frolich electron–polaron. In a realistic model much weaker deformation effects responsible for the interaction with acoustical phonons are included.

The observation of the exciton dressing with phonons is possible when the dipole coupling with the electromagnetic field is sufficiently strong to preserve a nonadiabatic (i.e. rapid) exciton creation (cf. 0.2 ps laser pulse in Borri *et al.* (2001)) much faster than dressing with phonons (of ps scale as we will show below). We consider a weakly polar medium for QDs (GaAs) in which the interaction of

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electrons with longitudinal optical (LO) phonons is dominating in terms of the influence on the quasiparticle energy shift. Much weaker interaction with gap-less longitudinal acoustic (LA) phonons plays an important role in kinematics of the dressing process, being a channel of energy transfer from exciton. To determine the limit of decoherence time of the optical excitations in QDs, this LA channel for dressing exciton with LO/LA phonons must be included.

We describe and analyze the dressing and the relaxation of exciton problems in detail for typical self-assembled strain induced InAs/GaAs QDs (Jacak, 1998) within the Green function formulation. We model the QD by the parabolic confinement potential.

2. PHONON DRESSING DESCRIPTION

To investigate the time evolution of the lowest exciton state, we consider the Hamiltonian describing a single electron and a hole interacting with phonons

$$\begin{aligned} \mathcal{H} = & \mathcal{H}_e(\mathbf{r}_e) + \mathcal{H}_h(\mathbf{r}_h) - \frac{e^2}{\epsilon_0 |\mathbf{r}_e - \mathbf{r}_h|} + \sum_{\mathbf{k}s} \hbar \omega_{\mathbf{k}s} b_{\mathbf{k}s}^\dagger b_{\mathbf{k}s} - \frac{e}{N^{1/2}} \left(\frac{2\pi \hbar \Omega}{v \tilde{\epsilon}} \right)^{1/2} \\ & \times \sum_{\mathbf{k}} \frac{1}{k} (b_{\mathbf{k}o} + b_{-\mathbf{k}o}^\dagger) (e^{i\mathbf{k}\mathbf{r}_e} - e^{i\mathbf{k}\mathbf{r}_h}) - \frac{1}{N^{1/2}} \left(\frac{\hbar}{2Mc_a} \right)^{1/2} \\ & \times \sum_{\mathbf{k}} k^{1/2} (b_{\mathbf{k}a} + b_{-\mathbf{k}a}^\dagger) (\sigma_e e^{i\mathbf{k}\mathbf{r}_e} - \sigma_h e^{i\mathbf{k}\mathbf{r}_h}). \end{aligned} \quad (1)$$

Here $b_{\mathbf{k}s}^\dagger$ ($b_{\mathbf{k}s}$) denote the phonon creation (annihilation) operators, ($s = o$ for LO, $s = a$ for LA), Ω denotes the frequency of the zero wave vector LO phonons, c_a —the LA phonon frequency, $\sigma_{e,h}$ —deformation constants for the electron and hole, M —the mass of ions in the elementary cell, v —the elementary cell volume, N —the number of cells in crystal, $\tilde{\epsilon} = (1/\epsilon_\infty - 1/\epsilon_0)^{-1}$ —the effective dielectric constant. The electron (hole) part of the Hamiltonian

$$\mathcal{H}_i(r_i) = -\frac{\hbar^2}{2m_i^*} \nabla_i^2 + \frac{1}{2} m_i^* (\omega_0^i)^2 r_{\perp i}^2 + \frac{1}{2} m_i^* (\omega_z^i)^2 z_i^2 \quad (2)$$

(where $i = e, h$, $r_{\perp i}^2 = x_i^2 + y_i^2$) contains the vertical and horizontal confinement potentials. There are characteristic confinement sizes $l_i = \sqrt{\frac{\hbar}{m_i^* \omega_0^i}}$, $l_{iz} = \sqrt{\frac{\hbar}{m_i^* \omega_z^i}}$ ($\omega_0^i \ll \omega_z^i$). Finding the eigenfunctions $\Phi_n(\mathbf{r}_e, \mathbf{r}_h)$ of the exciton Hamiltonian

$$\mathcal{H}_{ex} = \mathcal{H}_e(\mathbf{r}_e) + \mathcal{H}_h(\mathbf{r}_h) - \frac{e^2}{\epsilon |\mathbf{r}_e - \mathbf{r}_h|}, \quad (3)$$

we write the Hamiltonian (1) in the second quantization representation in the form of the sum of unperturbed and interaction parts

$$\begin{aligned} \mathcal{H} = & \sum_n \hbar E_n a_n^\dagger a_n + \sum_{k,s} \hbar \omega_{ks} b_{ks}^\dagger b_{ks} \\ & + \frac{1}{N^{1/2}} \sum_{n_1, n_2, \mathbf{k}, s} F_s(n_1, n_2, \mathbf{k}) a_{n_1}^\dagger a_{n_2} (b_{\mathbf{k}o} + b_{-\mathbf{k}o}^\dagger), \end{aligned} \quad (4)$$

where

$$\begin{aligned} F_o(n_1, n_2, \mathbf{k}) = & -\frac{e}{k} \left(\frac{2\pi \hbar \Omega}{v \tilde{\epsilon}} \right)^{1/2} \\ & \times \int \Phi_{n_1}^*(\mathbf{r}_e, \mathbf{r}_h) (e^{i\mathbf{k}\mathbf{r}_e} - e^{i\mathbf{k}\mathbf{r}_h}) \Phi_{n_2}(\mathbf{r}_e, \mathbf{r}_h) d^3 \mathbf{r}_e d^3 \mathbf{r}_h. \\ F_a(n_1, n_2, \mathbf{k}) = & -\left(\frac{\hbar k}{2Mc_a} \right)^{1/2} \\ & \times \int \Phi_{n_1}^*(\mathbf{r}_e, \mathbf{r}_h) (\sigma_e e^{i\mathbf{k}\mathbf{r}_e} - \sigma_h e^{i\mathbf{k}\mathbf{r}_h}) \Phi_{n_2}(\mathbf{r}_e, \mathbf{r}_h) d^3 \mathbf{r}_e d^3 \mathbf{r}_h. \end{aligned} \quad (5)$$

The linear response of the exciton to the electromagnetic field is described by using the retarded Green function of the electric current operators

$$\mathbf{j}(t) \sim \sum_n \mathbf{d}_n [a_n(t) + a_n^\dagger(t)], \quad (6)$$

here \mathbf{d}_n denotes an effective dipole moment of the exciton (Elliott, 1957; Mahan, 2000). The spectral density of the linear response function is determined by the causal one-particle functions $G_{nn'}(t) = -i \langle T \{ a_n(t) a_{n'}^\dagger \} \rangle$ (where the average is taken with Hamiltonian (4)) which we evaluate solving the Dyson equation. We calculate the spectral function $A(\omega) \equiv -2\text{Im}G_{00}^r(\omega) = -2\text{Im}G_{00}(\omega + i0^+)$ and its time course $A(t) = (2\pi)^{-1} \int_{-\infty}^{\infty} A(\omega) e^{-i\omega t} d\omega$. To determine the one-particle causal function, we evaluate the components of the mass operator using standard method of the solution of the Green function equations of motion

$$\left(i\hbar \frac{d}{dt} - E_n \right) G_{nn'}(t) - \frac{1}{N^{1/2}} \sum_{n_1, \mathbf{k}, s} F_s(n, n_1, \mathbf{k}) R_{n_1 n'}(\mathbf{k}s, t) = \delta(t) \delta_{nn'}, \quad (7)$$

where

$$R_{n_1 n'}(\mathbf{k}s, t) = \frac{i}{\hbar} \langle T \{ a_{n_1}(t) [b_{\mathbf{k}s}(t) + b_{-\mathbf{k}s}^\dagger(t)] a_{n'}^\dagger(0) \} \rangle, \quad (8)$$

with additional condition $\langle \tilde{\varphi}_{\mathbf{k}s}(t) \rangle \equiv \langle b_{\mathbf{k}s}(t) + b_{-\mathbf{k}s}^\dagger(t) \rangle = 0$ (Engelsberg and Schrieffer, 1963; Martin and Schwinger, 1959). Its Fourier transform takes the

form of the Dyson equation

$$G_{nn'}(\omega) = G_{nn_1}^{(0)}(\omega) + \sum_{n_1 n_2} G_{nn_1}^{(0)}(\omega) \Sigma_{n_1 n_2}(\omega) G_{n_2 n'}(\omega). \tag{9}$$

Neglecting the off diagonal Green functions, one finds the diagonal elements of the mass operator

$$\begin{aligned} \Sigma_{nn}(\omega) &= \frac{1}{N} \sum_{\mathbf{k}_s} \sum_{n_1} F_s(n, n_1, \mathbf{k}) F_s(n_1, n, -\mathbf{k}) \\ &\times \left\{ \frac{\Gamma_{n_1, n, -\mathbf{k}_s}(\omega - \omega_{\mathbf{k}_s}, -\omega_{\mathbf{k}_s}) [1 + n(\hbar\omega_{\mathbf{k}_s})]}{\hbar(\omega - \omega_{\mathbf{k}_s}) - E_{n_1} - \Sigma_{n_1 n_1}(\omega - \omega_{\mathbf{k}_s})} \right. \\ &\left. + \frac{\Gamma_{n_1, n, \mathbf{k}_s}(\omega + \omega_{\mathbf{k}_s}, \omega_{\mathbf{k}_s}) n(\hbar\omega_{\mathbf{k}_s})}{\hbar(\omega + \omega_{\mathbf{k}_s}) - E_{n_1} - \Sigma_{n_1 n_1}(\omega + \omega_{\mathbf{k}_s})} \right\}. \end{aligned} \tag{10}$$

Here $n(x) = (e^{x/T} - 1)^{-1}$, and $\Gamma_{nn', \mathbf{k}_s}(\omega_1, \omega_2)$ denotes the Fourier transform of a vertex function

$$\Gamma_{nn', \mathbf{k}_s}(t, t') \equiv - \frac{\delta G_{nn'}^{-1}(t)}{F_s(n, n', \mathbf{k}) \delta \tilde{\varphi}_{\mathbf{k}_s}(t')}. \tag{11}$$

satisfying $\Gamma_{nn, \mathbf{k}_s}(\omega, \omega_1) \approx 1 - \hbar^{-1} d\Sigma_{nn}/d\omega$ when consider terms of the lowest order in \mathbf{k} . Let us denote real and imaginary part of the mass operator, respectively, $\text{Re} \Sigma_{00}(\omega) \equiv \Delta(\omega)$, $\text{Im} \Sigma_{00}(\omega) \equiv -\gamma(\omega)$.

We may solve the mass operator equation (10) with the assumption that the dressed exciton is a durable composite particle which is undamped (Suna, 1964). It corresponds to the strictly solvable one level model of (Krummheuer *et al.*, 2002). We expect in this case that the exciton Green function has a real pole at frequency corresponding to the exciton–polaron energy. In the frequency region in vicinity of this pole, the imaginary part of the mass operator can be neglected compared to the real part, which allows us to write the self-consistent equation for $\Delta(\omega)$ (assuming $\Delta(\omega) \simeq -\Delta = \text{const}$)

$$-\Delta \simeq \frac{\hbar}{N} \sum_{\mathbf{k}} J_{00}^o(\mathbf{k}) \left\{ \frac{[1 + n(\hbar\omega_{\mathbf{k}_o})]}{E_0 - \tilde{E}_n - \hbar\omega_{\mathbf{k}_o}} + \frac{n(\hbar\omega_{\mathbf{k}_o})}{E_0 - \tilde{E}_n + \hbar\omega_{\mathbf{k}_o}} \right\} \tag{12}$$

which may be solved exactly ($\tilde{E}_n = E_0 - \Delta$, $J_{nn'}^s(\mathbf{k}) = F_s(n, n', \mathbf{k})$). The energy shift due to the interaction with LA phonons is negligible. One can calculate the form factors

$$\begin{aligned} J_{00}^o(\mathbf{k}) &= \frac{\pi e^2 \hbar \Omega}{18 \nu \tilde{\epsilon}} (L_e^2 - L_h^2)^2 e^{l_{\perp} k_{\perp}^2 / 2 - l_z k_z^2 / 2}, \\ J_{00}^a(\mathbf{k}) &= \frac{\hbar k}{2M c_a} (\sigma_e - \sigma_h)^2 e^{l_{\perp} k_{\perp}^2 / 2 - l_z k_z^2 / 2}, \end{aligned} \tag{13}$$

(here $l = l_e \approx l_h, l_z = l_{ze} \approx l_{zh}, L_e = \sqrt{\frac{\hbar}{m_e^* \omega_m}}, L_h = \sqrt{\frac{\hbar}{m_h^* \omega_m}}$, and $\omega_m = \sqrt{\frac{m_e^*}{m} (\omega_0^e)^2 + \frac{m_h^*}{m} (\omega_0^h)^2}$, $m = m_e^* + m_h^*$) (Davydov, 1976).

However, we intend to describe the dressed exciton as a damped quasiparticle at nonzero temperature range. When include many exciton energy levels, the damping is a consequence of the inclusion of phonon dispersion which is responsible for the interaction between the dressed exciton and the phonon subsystem (Davydov and Pestryakov, 1972). We solve (10) in two iteration steps in the vicinity of $\hbar\omega = E_0$, taking in the first approximation the vertex functions $\Gamma_{n_1 n', \mathbf{k}s}(\omega, \omega_1) = 1$, as done in (Moskalenko *et al.*, 1968). Starting from $\Delta_{(0)}(\omega) = 0, \gamma_{(0)}(\omega) = 0^+$ and assuming that $\Delta(\omega) \simeq \text{const}$, we find in the first iteration step

$$\Delta_{(1)}(\omega) \simeq \Delta_{(1)}(E_0/\hbar) = -\Delta' = \frac{\hbar}{N} \sum_{\mathbf{k}} \frac{b_n J_{0n}^o(\mathbf{k})}{E_0 - E_n - \hbar\omega_{\mathbf{k}o}},$$

$$\gamma_{(1)}(\omega) = \frac{\hbar\pi}{N} \sum_{\mathbf{k}} J_{00}^a(\mathbf{k}) \{ [1 + n(\hbar\omega_{\mathbf{k}a})] \delta(\hbar\omega - E_0 - \hbar\omega_{\mathbf{k}a}) + n(\hbar\omega_{\mathbf{k}a}) \delta(\hbar\omega - E_0 + \hbar\omega_{\mathbf{k}a}) \}. \tag{14}$$

Here b_n denotes the n th level degeneracy rank ($b_n = 1, 2$). The optical phonon contribution to $\gamma(\omega)$, is significant for $\hbar\omega \cong E_n \pm \hbar\Omega$ only. We include the LO phonon influence on $\gamma(\omega)$, introducing an effective constant to $J_{00}^a(\mathbf{k})$, which does not influence the time of fast dressing with phonons but it determines the relaxation time at nonzero T . This constant will be estimated from the sum rule $A(t = 0)| = 1$ for $T = 0$. In the second step, the relation $\Delta_{(1)}(\omega) \gg \gamma_{(1)}(\omega)$ enables one to neglect the $\gamma_{(1)}(\omega)$ in the denominators of (10). It leads to

$$\Delta_{(2)}(\omega) \cong \Delta_{(2)}(E_0/\hbar) = -\Delta'' \simeq \Delta_{(2)}(\tilde{E}'_0/\hbar) + \hbar^{-1} \frac{d\Delta_{(2)}}{d\omega} \Big|_{\omega=\tilde{E}'_0/\hbar} \Delta'$$

$$= -\Delta' Z^{-1}, \gamma_{(2)}(\omega) = \gamma_{(1)}(\omega + \Delta'/\hbar), \tag{15}$$

where $\tilde{E}'_n = E_n - \Delta', \Delta_{(2)}(\tilde{E}'_0/\hbar) = \frac{\hbar}{N} \sum_{\mathbf{k}} \frac{b_n J_{0n}^o(\mathbf{k})}{\tilde{E}'_0 - \tilde{E}'_n - \hbar\omega_{\mathbf{k}o}} = -\Delta'$. Continuing the iteration process we would arrive to a self-consistent equation for Δ similar to (12), however, after infinite number of the approximations. We find

$$\gamma_{(2)}(x) = Z^{-1} \alpha x^3 e^{-\beta x^2} f(x) \{ -n(-x)\theta(-x) + [1 + n(x)]\theta(x) \} \tag{16}$$

where $x = \hbar\omega - E_0 + \Delta', f(x) = \sum_{n=0}^{\infty} \frac{[-\beta x^2 (l_v^2/l_h^2 - 1)]^n}{(2n+1)n!}$, Z denotes the vertex function renormalization factor ($Z = |1 - \hbar^{-1} d\Delta/d\omega|^{-1}$). We write the spectral function in the form

$$A(\omega) = Z \frac{\hbar 2\tau^{-1}(\omega)}{[\hbar\omega - E_0 + Z^{-1}\Delta']^2 + \tau^{-2}(\omega)}, \tag{17}$$

where $\tau^{-1}(\omega) = Z\gamma(\omega)$ (Langreth and Kadanoff, 1964; Mahan, 1966). Since at zero temperature, the renormalization factor may be calculated directly

$$Z \approx 1 + \frac{\hbar}{N} \sum_{\mathbf{k}} \frac{b_0 J_{0n}^o(\mathbf{k})}{(E_0 - E_n - \hbar\omega_{\mathbf{k}o})^2}, \quad (18)$$

the sum rule $|A(t=0)| = 1$ enables us to estimate the effective damping constant α . At higher temperatures, when the phonon dispersion effects are strong we evaluate Z using the same condition ($|A(t=0)| = 1$).

3. RESULTS AND CONCLUSIONS

We have evaluated the interaction form factors $J_{nm}^s(\mathbf{k})$ taking the function of the noninteracting electron and hole system as the first approximation for the exciton wave function $\Phi_n(\mathbf{r}_e, \mathbf{r}_h) = \Psi_{n_e m_e}^{(e)}(\mathbf{r}_e) \Psi_{00}^{(h)}(\mathbf{r}_e)$, where

$$\begin{aligned} \Psi_{nm}^{(i)}(\mathbf{r}) &= \psi_{\perp nm}^{(i)}(r_{\perp}, \varphi) \phi^{(i)}(z) = \frac{N_{nm}^{(i)}}{(2\pi)^{1/2}} \left(\frac{r_{\perp}^2}{l_i^2} \right)^{|m|/2} e^{-\frac{r_{\perp}^2}{2l_i^2}} L_n^{|m|} \left(\frac{r_{\perp}^2}{l_i^2} \right) \\ &\times e^{im\varphi} \frac{1}{\pi^{1/2} l_{iz}} e^{-\frac{z^2}{2l_{iz}^2}}, \quad N_{nm}^{(i)} = \frac{1}{\pi^{1/2} l_i} \left(\frac{n!}{(n+|m|)!} \right) \end{aligned} \quad (19)$$

Since $\omega_0^i \ll \omega_z^i$, we consider only the first level for the quantization in z -axis direction. A fact that an effective mass of a hole is large in comparison to the electron effective mass, enables one to neglect the hole levels different than the

Table I. Exciton Energy Levels E_n and Corresponding Integral Form Factors J_{0n}^o Multiplied by n th Level Degeneracy Rank b_n

$l_e = 3 \text{ nm}$		$l_e = 6 \text{ nm}$	
$E_n [\text{meV}]$	$b_n J_{0n}^o [\text{meV}^2]$	$E_n [\text{meV}]$	$b_n J_{0n}^o [\text{meV}^2]$
88.9	13.1	7.87	12.5
113.3	2×39.0	15.7	2×18.8
134.0	36.0	23.3	21.5
135.8	2×16.9	23.8	2×9.9
152.0	2×18.4	29.6	2×10.4
155.6	2×5.6	30.8	2×3.5
167.9	18.2	34.8	7.4
169.4	2×10.7	35.5	2×4.7
183.8	2×13.0	39.5	2×5.4
199.0	11.5	44.2	4.0

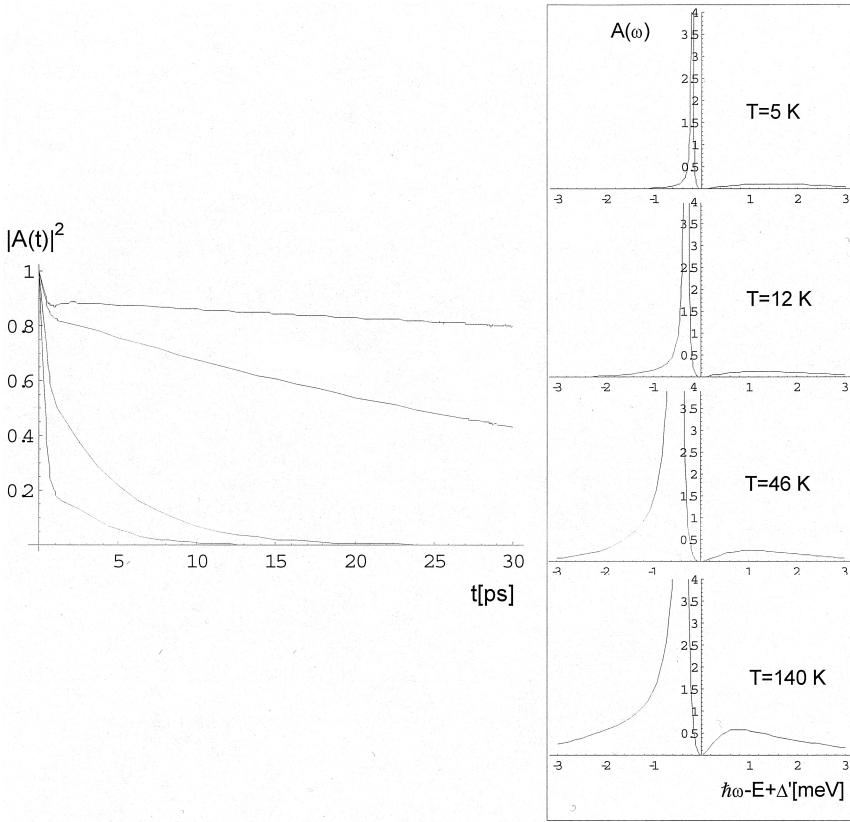


Fig. 1. Time course of the response function and spectral intensity for $l_e = 3$ nm, $l_z/l_e = 1/3$.

lowest one (of $n_h = 0, m_h = 0$). Defining the functions

$$\begin{aligned}
 I_{n'n''}^{(i)}(\mathbf{k}) &= \int_0^\infty \int_0^{2\pi} \psi_{n''}^{(i)*}(r_\perp, \varphi) e^{ik_\perp r_\perp \cos(\varphi)} \psi_{n'}^{(i)}(r_\perp, \varphi) dr_\perp d\varphi \\
 &\times \int_{-\infty}^\infty \phi^{(i)*}(z) e^{ik_z z} \phi^{(i)}(z) dz = I_{\perp n'n''}^{(i)}(k_\perp) e^{-2(k_z l_z/2)^2}, \quad (20)
 \end{aligned}$$

where the index n replaces both n_e, m_e quantum numbers, we write the form factors in the form

$$\begin{aligned}
 J_{nn'}^o(\mathbf{k}) &= \frac{2\pi e^2 \hbar \Omega}{\nu \tilde{\epsilon} k^2} \left[I_{nn'}^{(e)}(\mathbf{k}) - I_{nn'}^{(h)}(\mathbf{k}) \right]^2, \\
 J_{nn'}^a(\mathbf{k}) &= \frac{\hbar k}{2Mc_d} \left[\sigma_e I_{nn'}^{(e)}(\mathbf{k}) - \sigma_h I_{nn'}^{(h)}(\mathbf{k}) \right]^2. \quad (21)
 \end{aligned}$$

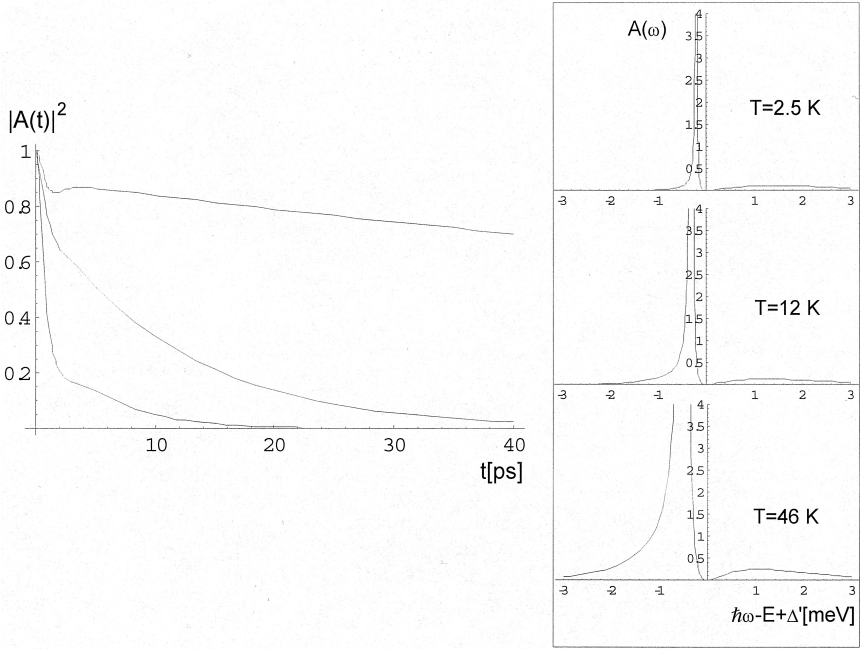


Fig. 2. Time course of the response function and spectral intensity for $l_e = 6$ nm, $l_z/l_e = 1/3$.

We perform the calculations of the energy shift including ten lowest energy levels, for the two confinement sizes $l_e = 6$ nm $\approx l_h$ and $l_e = 3$ nm $\approx l_h$. The following material parameters suitable to the InAs/GaAs QD have been used $m_e^* = 0.067m_0$, $m_h^* = 0.38m_0$, $\epsilon_0 = 12.9$, $\epsilon_\infty = 10.9$, $\hbar\Omega = 36.4$ meV, $c_a = 4.8 \times 10^3$ m/s. The integral form factors of the exciton-LO phonon interaction $J_{0n}^o = (\hbar/N) \sum_k J_{0n}^o(\mathbf{k})$ multiplied by the level degeneracy rank corresponding to the exciton energies E_n are presented in Table I.

The inclusion of many exciton levels influences the dressed exciton energy shift. It is responsible for its substantial increase (from 0.5 meV found in one level approximation to 3.6 meV when include ten levels for $l_e = 3$ nm, and from 0.35 meV to 2.9 meV for $l_e = 6$ nm). The other material constants $\sigma_e = 6.7$ eV, $\sigma_e = 2.7$ eV, $\rho = 5.36$ g/cm³ (Adachi, 1985) are useful for estimations.

The spectral density and its inverse Fourier transform calculated including many exciton levels and including realistic asymmetry of the QD ($l_z/l_e = 1/3$) are plotted in Figs. 1 and 2. Generally, the LA channel of dressing gives for the typical QDs the *picosecond scale of dressing*. Inclusion of the LO channel does not modify significantly the overall LO and LA dressing kinetics in comparison to LA channel solely (Takagahara, 1999). Thus the LA channel of dressing gives

the limit for adiabatic creation of exciton–polaron in InAs/GaAs QD. The slope of the time courses of spectral functions is related to damping of the dressed exciton at nonzero temperature. The relaxation time is strongly influenced by the size of QD.

Let us emphasize that our results for time dependence of the response functions (Figs. 1 and 2) coincide with the experimental results by Borri *et al.* (2001).

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