

Unavoidable decoherence in semiconductor quantum dots

L. Jacak, P. Machnikowski, W. Jacak, M. Krzyżosiak
*Institute of Physics, Wrocław University of Technology,
Wybrzeże Wyspiańskiego 27, 50-370 Wrocław, Poland*

The phonon-induced decoherence of orbital degrees of freedom in quantum dots (QDs) is studied in order to verify the feasibility of quantum information processing (QIP) in the QD technology. A source of an unavoidable partial leakage of the information from a QD is the dressing of quickly (sub-ps) excited carriers (electrons/excitons) in QDs mostly with inertial crystal deformation modes (LA phonons). This ps-time process scales as the QD size divided by the LA phonon velocity. For a polar medium (e.g. self-assembled GaAs/InAs dots) polarization effects (LO phonons) also contribute to dephasing however with a longer time scale (up to 100 ps) proportional to the square of the QD diameter. The anharmonicity effects (e.g. LO-TA in GaAs) enhance the efficiency of this channel again to the order of a few ps—inconveniently located between the possible sub-ps operation time and the ns exciton recombination time. Relatively slow (of a ps scale) dressing of electrons with phonon clouds is also a source of the temporal inefficiency of Pauli blocking (in spin-charge conversion schemes) since a quickly excited electron differs from an electron stored in a QD and thus already dressed with phonons. In a polar medium an additional confinement-induced enhancement of the interaction of carriers with optical phonons is also predicted (strong enhancement of the effective Fröhlich constant in a QD, strengthening polaronic effects). In so-called magnetic QDs, i.e. dots placed in a diluted magnetic semiconductor medium (promising for the coherent spin control) the role of phonons is played by magnons. The dressing of a localized spin with magnons also results in an inconvenient time scale of dephasing (of the order of 100 ps, similarly as for LO phonons due to the quadratic form of the magnon dispersion).

I. INTRODUCTION

Recent progress in coherent quantum control¹ gives rise to the interest in the possibility of QIP within the solid state technology employing orbital^{2–5} or spin^{6–9} degrees of freedom in QDs. In spite of the still growing accuracy both in QD manufacturing and control techniques, including the observation of Rabi oscillations^{3,10–14} and the demonstration of entanglement between states of interacting dots¹⁵, a complete implementation of a quantum gate on QDs has not been achieved so far. Since fault-tolerant quantum schemes require the decoherence per quantum gate to be below the threshold of 10^{-5} (DiVincenzo conditions¹⁶), the most important problem emerging within the challenging field of QD-based quantum computing is how to overcome the phonon decoherence in optically driven QD gates (orbital degrees of freedom) or how to accelerate single-qubit spin operations in the magnetic-field-driven ones. An estimation of the decoherence in state-of-the-art QD systems is thus of major importance. Since QDs are embedded in a surrounding crystal structure, the crucial role in the decoherence (relaxation and dephasing) of orbital degrees of freedom is played by phonons. Phonons are also important in the decoherence of the spin in QDs, not only due to the spin-orbit interaction but also due to Hund-like rules in multi-electron dots¹⁷ and in spin-charge conversion schemes.

The dipole-type interaction couples the electric field of a laser pulse only with charge degrees of freedom which can be thus directly manipulated in a controlled way even on the time-scale down to femtoseconds¹⁸. Due to the lattice inertia, a fast charged carrier excitation leaves the lattice in its initial state leading to the creation of a bare electron-hole pair. On the other hand, it is known that eigenstates of an interacting carrier-phonon system correspond to a composite quasiparticle: the electron-hole pair accompanied by a coherent phonon cloud (lattice polarization or deformation)^{19,20}. The phonon cloud corresponds to the energy-minimizing state of the lattice and is therefore characterized by a negative energy shift (red-shift) with respect to the original bare exciton energy. The excess energy is being transferred from the QD region to the rest of the crystal (to the phonon subsystem) and locally in the dot it is creating a dressed exciton being a coherent composition of the bare exciton and a local phonon cloud. If only longitudinal optical (LO) phonons are considered, this composite particle can be called exciton-polaron in analogy to the Fröhlich electron-polaron. The corresponding energy red-shift in a typical QD is of the order of a few meV^{21,22} for dressing with LO phonons. Dressing with acoustic phonons, due to deformation effects (longitudinal acoustic, LA) and piezoelectric effects (transversal acoustic, TA) results in an usually much smaller energy shift, even by a few orders of magnitude. However, acoustic phonons mostly contribute to the overall dephasing.

Dressing with phonons appears to be a relatively slow process—of a ps scale as we will show below—and turns out to be governed mainly by phonon dispersions and the dot size. The characteristic dressing time depends on the QD size (the scale of confinement), and it can be approximated as the ratio of the dot size and the phonon velocity. It corresponds to the time needed for the transfer of the excess energy from the QD region to the surrounding crystal.

In a polar material (here we consider the weakly polar GaAs as the most typical medium for QDs) with dominating

coupling of electrons with LO phonons, this polarization interaction produces almost all energy red-shift of the composite quasiparticle. The deformation interaction of the exciton with LA phonons is of a few orders of magnitude weaker (in the GaAs case) and thus results in a negligibly small additional energy shift (similarly to TA phonons). Despite of the small energy shift, dephasing due to LA phonons is pronounced. The gapless, almost linear (near to the Γ point) and wide dispersion of LA phonons allows for an effective (many LA phonons contribute) and quick channel of the deformation energy transfer from the step-by-step dressing exciton to the phonon sea.

The transfer of the excess polarization energy from the dot region to the surrounding medium allowing the creation of an exciton-polaron by means of LO phonons is considerably distinct from the LA channel. Firstly, because of the presence of a relatively wide gap in the dispersion of LO phonons (~ 36 meV in GaAs, i.e. much larger than the energy shift of the dressed particle which is of the order of a few meV) the process of dressing via the LO channel cannot be interpreted as elementary phonon excitations unlike in the case of gapless LA phonons. For the LO channel we deal rather with a coherent multiparticle process. The dressing is a redistribution of the original instant bare exciton energy over the whole system consisting of the dressed exciton (only few LO phonon modes couple to the exciton²³) and the rest of LO phonons. The initial bare exciton state is not a stationary state of the total system including phonons. The kinetics of dressing corresponds to the time-evolution of this nonstationary state. Since the exciton-phonon system is closed, the average energy is conserved, even though at different instants of time the mean energy is distinctly shared between particular parts of the system. LO phonons coherently contribute to the averaged energy, according to the total system wave function which is however not of a separable structure. Secondly, the dispersion of LO phonons is narrow (especially near to the Γ point) and of an almost parabolic form (this region of small k is the most important one due to the bottle-neck effect for QDs similarly as for the LA channel). It results in a longer dressing time via the LO channel (for not extremely small dots) in comparison to the LA channel since the group velocity of LO phonons is considerably smaller than that of LA phonons. For LO phonons this velocity scales as the inverse of the dot size which gives an additional factor for an estimation of the dressing-time scale, as it is proportional to the square of the dot diameter. The dressing with LO phonons is however aided by the LO-TA anharmonicity, the quickest anharmonic channel in GaAs, which allows for a polarization energy transfer to the TA phonon sea. In this case the characteristic time can be estimated as the dot size divided by TA phonon velocity and multiplied by anharmonicity and exciton-LO phonon coupling constants.

The shortest timescale of all dressing processes (LA channel in GaAs medium at least) can be treated as a limit for the unavoidable decoherence (dephasing) of rapidly created exciton states in QDs (also of the ground state), strongly limiting optically driven QIP in nanoscopic systems. Longer processes as those via LO phonons (even aided by LO-TA) are also inconvenient since they additionally confine adiabatical switching regime to a slower range.

These problems are described and analyzed below in detail for a typical state-of-the-art strain induced InAs/GaAs QD within the Green function approach^{24,25}. The QD is modelled by a parabolic confinement potential¹⁷ (distinct for electrons and holes). Other shapes of QD confining potential (square well or Gaussian) does not significantly modify the results.

II. PHONON DRESSING OF AN EXCITON IN A QD

In order to investigate the time evolution of the nonstationary state corresponding to a rapidly created QD exciton (in the limit—instantly excited exciton; in practice sub-ps time-scale process) we consider the Hamiltonian describing a single exciton interacting with phonons

$$H = \sum_n E_n a_n^\dagger a_n + \sum_{\mathbf{q},s} \hbar \omega_s(\mathbf{q}) c_{\mathbf{q},s}^\dagger c_{\mathbf{q},s} + \frac{1}{\sqrt{N}} \sum_{\mathbf{q},n_1,n_2,s} F_s(n_1, n_2, \mathbf{q}) a_{n_1}^\dagger a_{n_2} (c_{\mathbf{q},s} + c_{-\mathbf{q},s}^\dagger), \quad (1)$$

where the interaction with LO ($s = o$) and LA ($s = a$) phonons is described by functions

$$F_o(n_1, n_2, \mathbf{q}) = -\frac{e}{q} \sqrt{\frac{2\pi\hbar\Omega}{v\tilde{\epsilon}}} \int \Phi_{n_1}^*(\mathbf{R}_e, \mathbf{R}_h) (e^{i\mathbf{q}\cdot\mathbf{R}_e} - e^{i\mathbf{q}\cdot\mathbf{R}_h}) \Phi_{n_2}(\mathbf{R}_e, \mathbf{R}_h) d^3\mathbf{R}_e d^3\mathbf{R}_h \quad (2)$$

and

$$F_a(n_1, n_2, \mathbf{q}) = -\sqrt{\frac{\hbar q}{2MC_a}} \int \Phi_{n_1}^*(\mathbf{R}_e, \mathbf{R}_h) (\sigma_e e^{i\mathbf{q}\cdot\mathbf{R}_e} - \sigma_h e^{i\mathbf{q}\cdot\mathbf{R}_h}) \Phi_{n_2}(\mathbf{R}_e, \mathbf{R}_h) d^3\mathbf{R}_e d^3\mathbf{R}_h. \quad (3)$$

Here $c_{\mathbf{q},s}^{(\dagger)}$ is the bosonic annihilation (creation) operator for a LO or a LA phonon with the quasi-momentum \mathbf{q} and with the frequency $\omega_o = \Omega_{\mathbf{q}} \simeq \Omega - \beta q^2$ (Ω denotes the gap of LO phonons at the Γ point) and $\omega_a = C_a q$, where C_a is the sound velocity for LA phonons, M —the mass of ions in the elementary cell, $\sigma_{e(h)}$ —the deformation constant for

electrons (holes), v —the volume of the elementary cell, N —the number of cells in the crystal, $\tilde{\epsilon} = (1/\epsilon_\infty - 1/\epsilon_0)^{-1}$ —the effective dielectric constant $\mathbf{R}_{e(h)}$ is the position of the electron (hole), $\Phi_n(\mathbf{R}_e, \mathbf{R}_h)$ is the exciton wave function and $a_n^{(\dagger)}$ —the annihilation (creation) operator of the exciton.

We will consider the exciton single-particle causal Green function

$$G_c(n_1, n_2, t) = -\frac{i}{\hbar} \langle T \{ a_{n_1}(t) a_{n_2}^\dagger(0) \} \rangle.$$

The symbol $\langle \dots \rangle$ denotes the temperature dependent averaging with respect to phonon degrees of freedom and the vacuum of the exciton (cf. Ref. 26). It corresponds to the case when the grand canonical averaging sector without exciton—the vacuum, is energetically distant (by the order of 1 eV) from the next sectors.

For $t \geq 0$ this Green function up to a constant factor coincides with the correlation function $\langle a_{n_1}(t) a_{n_2}^\dagger(0) \rangle$, modulus of which gives a measure of the fidelity of the time dependent state (for $n_1 = n_2$, in particular of the *ground* state for $n_1 = n_2 = 0$), since it corresponds to the overlap of the state at time t with this state at the initial moment $t = 0$. The Fourier transform of the correlation function $I_{n_1, n_2}(\omega) = \int_{-\infty}^{\infty} \langle a_{n_1}(t) a_{n_2}^\dagger(0) \rangle e^{i\omega t} dt$ is usually called the spectral density^{24,25}, and it can be expressed by the imaginary part of the causal Green function

$$\text{Im} G_c(n_1, n_2, \omega) = -\frac{1}{2\hbar} \left(1 + e^{-\frac{\hbar\omega}{k_B T}} \right)^{-1} I(n_1, n_2, \omega), \quad (4)$$

where $G_c(n_1, n_2, \omega) = \int_{-\infty}^{\infty} G_c(n_1, n_2, t) e^{i\omega t} dt$. Moreover

$$\text{Im} G_r(n_1, n_2, \omega) = -\frac{1}{2\hbar} \left(1 - e^{-\frac{\hbar\omega}{k_B T}} \right)^{-1} I(n_1, n_2, \omega),$$

where $G_r(n_1, n_2, t) = -\frac{i}{\hbar} \Theta(t) \langle [a_{n_1}(t), a_{n_2}^\dagger(0)]_- \rangle = \frac{1}{2\pi} \int_{-\infty}^{\infty} G_r(n_1, n_2, \omega) e^{-i\omega t} d\omega$ is the commutation retarded Green function which describes the linear dielectric response to a e-m wave coupled to the exciton²⁷. In our case, of the instant creation of the exciton, the time-dependent e-m signal is assumed as $\delta(t)$.

The causal Green function satisfies the equation of motion

$$i\hbar \frac{d}{dt} G_c(n_1, n_2, t) = \delta(t) \delta_{n_1, n_2} - \frac{i}{\hbar} \langle T \{ [a_{n_1}(t), H(t)]_- a_{n_2}^\dagger(0) \} \rangle, \quad (5)$$

which can be rewritten as a Dyson-type equation

$$(\hbar\omega - E_{n_1}) G_c(n_1, n_2, \omega) - \sum_{n_3} M_{n_1, n_3}(\omega) G_c(n_3, n_2, \omega) = \delta_{n_1, n_2}, \quad (6)$$

with the mass operator

$$M_{n_1, n_5}(\omega) = \frac{i}{2\pi\sqrt{N}} \sum_{n_3, \mathbf{k}, \mathbf{s}} F_s(n_1, n_3, \mathbf{k}) \times \sum_{n_4, \mathbf{k}'', \mathbf{s}''} \int d\omega_4 G_c(n_3, n_4, \omega + \omega_4) \Gamma(n_4, n_5, \omega + \omega_4, \mathbf{k}'', \mathbf{s}'', \omega_4) \mathbf{D}_c(\mathbf{k}'', \mathbf{s}'', \mathbf{k}, \mathbf{s}, \omega_4), \quad (7)$$

where Γ is the appropriate vertex function^{24,25,28} and

$$D_c(\mathbf{k}, \mathbf{s}, \mathbf{k}', \mathbf{s}', t) = -\frac{i}{\hbar} \langle T \{ [c_{\mathbf{k}, \mathbf{s}}(t) + c_{-\mathbf{k}, \mathbf{s}}^\dagger(t)] [c_{\mathbf{k}', \mathbf{s}'}(0) + c_{-\mathbf{k}', \mathbf{s}'}^\dagger(0)] \} \rangle$$

is the phonon causal Green function. For a weak exciton-phonon coupling, the phonon function D can be replaced by the free phonon function D^0 .

Since $F_s(n_1, n_2, \mathbf{k}) \sim g_s$, where g_s is the exciton-phonon coupling constant, then $|M_{n_1, n_3}(\omega)|^2 \sim g_s^2$. For $g_s \ll 1$, with the accuracy up to g_s^3 , we have from Eq. (6)

$$G_c(n, n, \omega) = \frac{1}{\hbar\omega - E_n - M_{n, n}(\omega)}. \quad (8)$$

Neglecting multi-phonon processes, for the real and the imaginary part of the mass operator M (i.e. $M_{n,n}(\omega) = \Delta_n(\omega) - i\gamma_n(\omega)$) we obtain the following equations (similar as for the bulk case²⁸)

$$\begin{aligned} \Delta_n(\omega) = & \frac{1}{N} \sum_{\mathbf{k},s,n_1} |F_s(n, n_1, \mathbf{k})|^2 \left[\frac{(1 + N_{\mathbf{k},s})(\hbar\omega - E_{n_1} - \Delta_{n_1}(\omega - \omega_s(\mathbf{k})) - \hbar\omega_s(\mathbf{k}))}{[\hbar\omega - E_{n_1} - \Delta_{n_1}(\omega - \omega_s(\mathbf{k})) - \hbar\omega_s(\mathbf{k})]^2 + \gamma_{n_1}^2(\omega - \omega_s(\mathbf{k}))} \right. \\ & \left. + \frac{N_{\mathbf{k},s}(\hbar\omega - E_{n_1} - \Delta_{n_1}(\omega + \omega_s(\mathbf{k})) + \hbar\omega_s(\mathbf{k}))}{[\hbar\omega - E_{n_1} - \Delta_{n_1}(\omega + \omega_s(\mathbf{k})) + \hbar\omega_s(\mathbf{k})]^2 + \gamma_{n_1}^2(\omega + \omega_s(\mathbf{k}))} \right] \end{aligned} \quad (9)$$

and

$$\begin{aligned} \gamma_n(\omega) = & \frac{1}{N} \sum_{\mathbf{k},s,n_1} |F_s(n, n_1, \mathbf{k})|^2 \left[\frac{(1 + N_{\mathbf{k},s})\gamma_{n_1}(\omega - \omega_s(\mathbf{k}))}{[\hbar\omega - E_{n_1} - \Delta_{n_1}(\omega - \omega_s(\mathbf{k})) - \hbar\omega_s(\mathbf{k})]^2 + \gamma_{n_1}^2(\omega - \omega_s(\mathbf{k}))} \right. \\ & \left. + \frac{N_{\mathbf{k},s}\gamma_{n_1}(\omega + \omega_s(\mathbf{k}))}{[\hbar\omega - E_{n_1} - \Delta_{n_1}(\omega + \omega_s(\mathbf{k})) + \hbar\omega_s(\mathbf{k})]^2 + \gamma_{n_1}^2(\omega + \omega_s(\mathbf{k}))} \right], \end{aligned} \quad (10)$$

Note that for $T = 0$ the above system of equations simplifies as then $N_{\mathbf{k},s} = 0$.

In order to solve the system of Eqs. (9)–(10) let us make some material estimations. For GaAs we assume:²⁹ $m_e^* = 0.067m_0$, $m_h^* = 0.38m_0$ (isotropic—for the sake of simplicity) and $\epsilon_0 = 12.9$, $\epsilon_\infty = 10.9$, $\sigma_e = 6.7$ eV, $\sigma_h = -2.7$ eV, $\hbar\Omega = 36.4$ meV, $\rho = 5.36$ g/cm³, $C_a = 4.8 \cdot 10^5$ cm/s. For the model InAs/GaAs self-assembled QD we assume $\hbar\omega_0^e = 20$ meV, $\hbar\omega_0^h = 3.5$ meV, $l_e = \sqrt{\frac{\hbar}{m_e^*\omega_0^e}} = l_h = \sqrt{\frac{\hbar}{m_h^*\omega_0^h}} = 7.5$ nm—i.e. the same lateral dimension for noninteracting e and h; for vertical confinement $l_z^{e(h)} \simeq 2$ nm (suitably to the appropriately chosen $\omega_z^{e(h)}$).

An approximate wave function which describes the ground state of the exciton (including the Coulomb interaction) with a high accuracy is of the form

$$\Phi_0(\mathbf{r}_e, \mathbf{r}_h) = \frac{1}{(\pi)^{3/2}} \frac{1}{L_e L_h L_z} e^{-\frac{r_e^2}{2L_e^2} - \frac{r_h^2}{2L_h^2} - \frac{z_e^2 + z_h^2}{L_z^2}}, \quad (11)$$

where (for the parameters used here) $L_e = 6.6$ nm and $L_h = 5.1$ nm, $L_z = l_z$. The difference for e and h is due to the fact that the Coulomb interaction energy is comparable to the inter-level distance for a heavier hole while the lowest excited electron states are much higher in the energy.

For

$$|F_o(0, 0, \mathbf{k})|^2 \simeq \frac{\pi e^2 \hbar \Omega k^2}{18 v \tilde{\epsilon}} (L_e^2 - L_h^2)^2 e^{-\alpha k^2} = g_o \frac{k^2}{k_m^2} e^{-\alpha k^2}$$

and

$$|F_a(0, 0, \mathbf{k})|^2 \simeq \frac{\hbar k}{2MC_a} (\sigma_e - \sigma_h)^2 e^{-\alpha k^2} = g_a \frac{k}{k_m} e^{-\alpha k^2},$$

where $k_m = (6\pi^2/v)^{1/3}$ is the Debye wave number ($\simeq 1.2 \cdot 10^8$ cm⁻¹), $\alpha = l^2/2$, and l is the size of the QD averaged over all directions (i.e. the averaged ground state size), the same for the electron and the hole (it is well smaller than the lateral dimension $l_{e(h)}$, but greater than the vertical one l_z). The exponential factor $e^{-\alpha k^2}$ reflects the bottle-neck effect for QDs (for InAs/GaAs QDs one can estimate $g_o \approx 3 \cdot 10^{-2}$ eV², $g_a \approx 4 \cdot 10^{-4}$ eV²).

For the interaction with LO phonons the Fröhlich constant $\alpha_e = \frac{e^2}{\hbar \tilde{\epsilon}} \sqrt{\frac{m_e^*}{2\hbar\Omega}}$ is important. In GaAs-bulk $\alpha_e \simeq 0.068$, whereas for electrons confined in InAs/GaAs QD it has been reported to be ca. 100% larger¹⁹). This strong enhancement is explained by nonadiabatic corrections³⁰.

In Eq. (9) the major term is the first one, i.e. the energy red-shift is mainly due to the interaction of the exciton with optical phonons (polaron effect). Taking $\gamma_n(\omega) = 0$ on the rhs of Eq. (9) as the zeroth approximation, one arrives with the equation for the energy shift

$$\begin{aligned} \Delta_n(\omega) = & \frac{1}{N} \sum_{\mathbf{k},n_1} |F_o(n, n_1, \mathbf{k})|^2 \left[\frac{1 + N_{\mathbf{k},o}}{\hbar\omega - E_{n_1} - \Delta_{n_1}(\omega - \Omega) - \hbar\Omega} + \frac{N_{\mathbf{k},o}}{\hbar\omega - E_{n_1} - \Delta_{n_1}(\omega - \Omega) + \hbar\Omega} \right] \\ & + \frac{1}{N} \sum_{\mathbf{k},n_1} |F_a(n, n_1, \mathbf{k})|^2 \left[\frac{1 + N_{\mathbf{k},a}}{\hbar\omega - E_{n_1} - \Delta_{n_1}(\omega - C_a k) - \hbar C_a k} + \frac{N_{\mathbf{k},a}}{\hbar\omega - E_{n_1} - \Delta_{n_1}(\omega - C_a k) + \hbar C_a k} \right] \end{aligned} \quad (12)$$

In this equation the first term determines the energy shift due to dressing with LO phonons, whereas the second one corresponds to LA phonons. The latter term is small and could be safely neglected, but it contributes to the derivative $\frac{d\Delta}{d\omega}|_{\omega=E+\Delta}$ as the derivative of the first term ($\sim \frac{F^2}{(\hbar\Omega)^2}$) is small due to the gap in the dispersion of LO phonons and it is strongly enhanced for gapless LA phonons term, which is important for estimation of a residuum of the Green function in a pole, cf. Eq. (15). Note also that in the first term in the above equation we neglected the weak LO phonon dispersion since it results in a negligible correction to Δ . Note that from the above equation yields a similar one found via Davydov diagonalization of the Fröhlich Hamiltonian for the exciton²⁷ (by taking in Eq. (12) $\hbar\omega = E_n + \Delta_n$, neglecting the LA phonon term and assuming Δ_n independent of ω).

The numerical solution of Eq. (12) for $n = 0$ provides the ground state energy shift $\Delta_0 \sim -5$ meV for a QD with the parameteres as listed above.

Assuming $\gamma = 0$ in the rhs of Eq. (10), the imaginary part of the mass operator is given by the equation

$$\begin{aligned} \gamma_n(\omega) = & \frac{\pi}{N} \sum_{\mathbf{k}, n_1} \{ |F_o(n, n_1, \mathbf{k})|^2 [(1 + N_{\mathbf{k},o})\delta(\hbar\omega - E_{n_1} - \Delta_{n_1} - \hbar\Omega_{\mathbf{k}}) + N_{\mathbf{k},o}\delta(\hbar\omega - E_{n_1} - \Delta_{n_1} + \hbar\Omega_{\mathbf{k}})] \\ & + |F_a(n, n_1, \mathbf{k})|^2 [(1 + N_{\mathbf{k},a})\delta(\hbar\omega - E_{n_1} - \Delta_{n_1} - \hbar C_a k) + N_{\mathbf{k},a}\delta(\hbar\omega - E_{n_1} - \Delta_{n_1} + \hbar C_a k)] \}. \end{aligned} \quad (13)$$

The first term in Eq. (13) describes the polarization energy transfer to the LO phonon sea, whereas the second one corresponds to the deformation energy transfer from a gradually dressing exciton to the LA phonon sea. γ can be estimated for the ground state ($n = 0$) neglecting higher exciton levels, by performing the integral over \mathbf{k} and employing the δ functions

$$\begin{aligned} \gamma_0(\omega) \simeq & Ax^3 e^{-\frac{\alpha x^2}{\hbar^2 C_a^2}} [\Theta(x)(1 + N(x)) - \Theta(-x)N(-x)] \\ & + B \left[\Theta(\hbar\Omega - x)(\hbar\Omega - x)^{3/2} e^{-\frac{\alpha(\hbar\Omega - x)}{\hbar\beta}} \Theta(-0.9\hbar\Omega + x)(1 + N(x)) \right. \\ & \left. + \Theta(\hbar\Omega + x)(\hbar\Omega + x)^{3/2} e^{-\frac{\alpha(\hbar\Omega + x)}{\hbar\beta}} \Theta(-0.9\hbar\Omega - x)N(-x) \right], \end{aligned} \quad (14)$$

where $x = \hbar\omega - \tilde{E}_0$, $\tilde{E}_0 = E_0 - \Delta_0$ is the energy of the exciton-polaron, $N(x) = (e^{\frac{x}{k_B T}} - 1)^{-1}$, $A = \frac{(\sigma_e - \sigma_h)^2}{4\pi\rho\hbar^3 C_a^5}$, $B = \frac{e^2 \hbar\Omega (L_e^2 - L_h^2)^2}{36\epsilon^2 (\hbar\beta)^{5/2}}$. For GaAs we assumed the dispersion of LO phonons in the form³¹ $\Omega_{\mathbf{k}} = \Omega - \beta k^2$ and for $k = k_m$, $\Omega_{\mathbf{k}_m} = 0.9\Omega$. The first term in Eq. (14) corresponds to the LA channel and the second one to the LO channel of energy dissipation. For GaAs $A \simeq 0.053$ meV⁻², $B \simeq 1.47 \cdot 10^7$ meV^{-1/2}, $\frac{\alpha}{\hbar^2 C_a^2} \simeq (l[\text{nm}]/4)^2 0.8$ meV⁻² and $\frac{\alpha}{\hbar\beta} \simeq (l[\text{nm}]/4)^2 8.79$ meV⁻¹.

As γ_0 is zero at $x = 0$ (cf. Eq. (14)) this point is a well defined pole of the causal Green function. It corresponds to a quasiparticle—the dressed exciton (i.e. a generalized exciton-polaron being an exciton dressed both with LO and LA phonon clouds). The time evolution of the dressing process is described by the correlation function which can be found from the imaginary part of the causal Green function. This function is of the form

$$G_c(0, 0, \omega) = \frac{1}{\hbar\omega - E_0 - \Delta(\omega) + i\gamma(\omega) + i\epsilon} = \frac{a^{-1}}{x + i\gamma'(x) + i\epsilon}, \quad (15)$$

where

$$a = 1 - \frac{d\Delta(\omega)}{\hbar d\omega} \Big|_{\omega=\tilde{E}'_0} = 1 + \frac{1}{N} \sum_{\mathbf{k}, \mathbf{s}} \left| \frac{F_s(0, 0, \mathbf{k})}{\hbar\omega_s(\mathbf{k})} \right|^2 [1 + 2N_s(\mathbf{k})], \quad (16)$$

$\gamma'(x) = \gamma(x)/a$ (also x is renormalized, $x = \hbar\omega - \tilde{E}'_0$, $\tilde{E}'_0 = \tilde{E}_0/a$), and $\epsilon = 0^+$. The imaginary part of the Green function (15) has the form

$$\text{Im } G_c(0, 0, \omega) = -a^{-1} \pi \delta(x) - \frac{a^{-1} \gamma'(x)/x^2}{1 + (\gamma'(x)/x)^2}. \quad (17)$$

In order to find the needed correlation function dependence upon time, the inverse Fourier transform of the spectral function must be performed. Employing Eq. (4) one has

$$I(t) = -2\hbar \frac{1}{2\pi} \int_{-\infty}^{\infty} d\omega \text{Im } G_c(0, 0, \omega) e^{-i\omega t}, \quad (18)$$

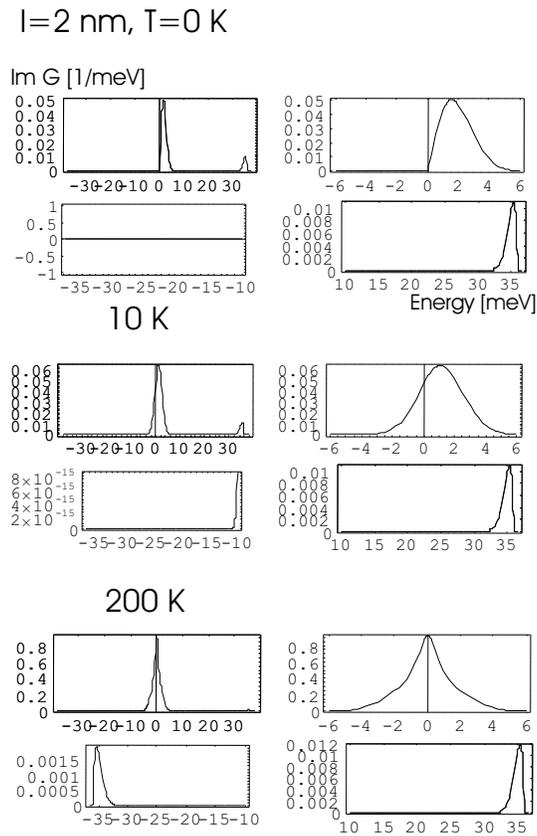


FIG. 1: Spectral intensity vs energy (Fourier representation of the correlation function for several temperature values and the averaged dot size $l = 2 \text{ nm}$); the satellite peaks correspond to the LO phonon channel, whereas the central peak with a side-band to the LA phonon channel; absorption processes are important only at higher temperatures (left peaks).

where the indices $n_1 = n_2 = 0$ of I are suppressed.

The first term in Eq. (17) yields

$$I^{(1)}(t) = a^{-1} e^{-i \frac{E_0}{\hbar} t}.$$

Note that in the latter term of Eq. (17) for temperature $T < 100 \text{ K}$ one can safely neglect the second term in the denominator, consistently with the accuracy assumed within the perturbative treatment. This permits the change of the order of integration over ω and \mathbf{k} —the integration over ω can be performed first, employing the delta-function form (13). Note that in the derivation of Eq. (14) the integration over \mathbf{k} was performed first, and the whole denominator contributed. Integrating first over frequencies we arrive with a simplified but convenient representation in the form

$$I^{(2)}(t) = a^2 - a^{-1} \frac{1}{N} \sum_{\mathbf{k}, s} \left| \frac{F_s(0, 0, \mathbf{k})}{\hbar \omega_s(\mathbf{k})} \right|^2 \left\{ [1 + N_s(\mathbf{k})] e^{-i[\tilde{E}_0/\hbar + \omega_s(\mathbf{k})]t} + N_s(\mathbf{k}) e^{-i[\tilde{E}_0/\hbar - \omega_s(\mathbf{k})]t} \right\}.$$

It may be noted, comparing with Eq. (16), that at $t = 0$ one has $I^{(1)}(t = 0) = a^{-1}$ and $I^{(2)}(t = 0) = a^{-1}(a - 1)$, thus providing a correct normalization of the correlation function. Moreover this attitude results in formulas allowing for explicit estimations of characteristic dressing times for both LA and LO channels, expressing them by means of the LO and LA phonon dispersions.

Both the spectral density and its inverse Fourier transform (the correlation function) calculated numerically are plotted in Figs. 1–3 for various temperatures and dot sizes, and compared with the experimental data. The coincidence of the time behavior of the fidelity measure (the correlation function) calculated above with that observed experimentally for a 0.2 ps pulse exciton in a small QD³² is very good (Fig. 2). In Fig. 3 (insets) the dressing time for LA and LO channels vs dot size is plotted. The LA dressing channel is the most effective one and for typical QDs it gives a picosecond time scale of dressing. The LO channel is slower and accompanied by oscillations related to the

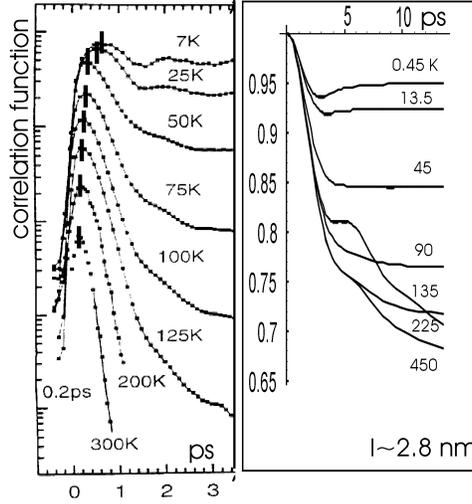


FIG. 2: Modulus of the correlation function $|\langle a(t)a^\dagger(0) \rangle|$ (the Fourier transform of the spectral density—a measure of the fidelity of the *ground* excitonic state) vs time (right); the same from experiment for a 0.2 ps pulse exciton, after Ref. 32 (left); vertical dashes indicate the origins of curves to allow a better comparison with the right picture.

gap in the LO phonon dispersion. The inclusion of the LO channel does not significantly modify the simultaneous LO and LA dephasing in comparison to that due to the LA channel solely (cf. Fig. 3). Thus the most effective and the quickest—the LA dressing channel provides the limit for the adiabatic creation of a generalized exciton-polaron in InAs/GaAs QDs.

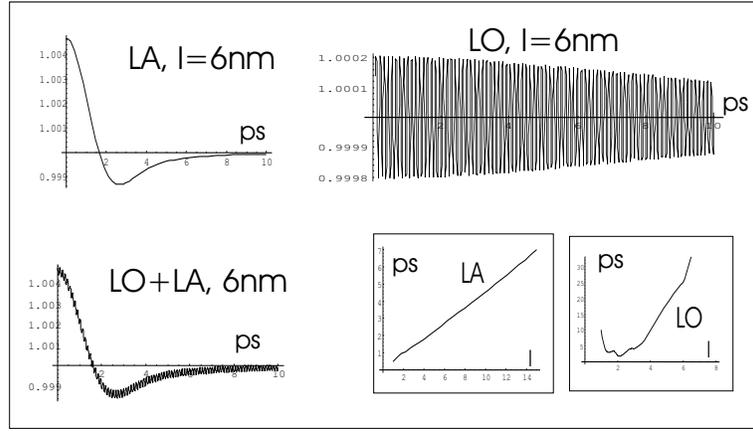


FIG. 3: The typical shape of the modulus of the correlation function for LA phonons only (upper-left), for LO phonons only (upper-right), for LO and LA phonons jointly (lower-left); the oscillations correspond to the gap in the LO phonons dispersion; dressing time vs averaged QD size for the LA channel and the LO channel (lower-right).

III. CONCLUSIONS

For a nanometer confinement scale for carriers in semiconductors like in the case of QDs we deal with a specific and completely different type of phonon-induced phenomena compared to the bulk case. The main features of this behavior can be listed as follows:

- The nanometer confinement scale results in the energy scale close to the resonance with optical phonons—thus coupling with LO phonons is always in the strong regime¹⁹ resulting in a significant modification of the electron/exciton spectrum, i.e. in strong polaronic effects—the typical energy shift for an electron-polaron is of the order of 10% of the confinement energy, whereas for an exciton-polaron it is of the order of 5%.

- The nanometer scale of confinement leads to a significant enhancement of the effective Fröhlich constant due to nonadiabatic effects. This additionally rises the electron-LO phonon interaction³⁰.
- The interaction with LA phonons in nanostructures is diminished since the energy conservation and the fact that nanostructure dimensions are incommensurate with phonons wavelengths result in a pronounced so-called bottle-neck effect³³.
- Relaxation rates in QDs (diagonal decoherence) for polarons (induced by the LO-TA and LO-LA anharmonicity) are at least one order of magnitude longer than in the bulk case, mostly due to coherent effects, combined bottle-neck effect and Fröhlich constant enhancement²⁰.

This list can be supplemented by the following statement, according to the above discussion.

- Dephasing in QDs (the off-diagonal decoherence) for a generalized exciton–polaron is of a ps time-scale due to the process of dressing of the exciton with LA phonons via the transfer of the local excess of the deformation energy to the LA phonon sea.
- Dephasing due to LO phonons is slower—at 100 ps scale, due to the weak LO phonon dispersion near to the Γ point; the transfer of the excess polarization energy out the QD in due of the polaron formation is however accelerated by the LO-TA anharmonic coupling, which is very efficient in GaAs, leading again to a few ps scale.

All these effects refer not only to QDs but rather to all nanometer-scale structures. Therefore phonon-induced phenomena will probably play much more important role in the whole imminent nanotechnology than in the 3D and 2D semiconductor structures. An example of such a new phenomenon can be related to the spin of the electron in a QD. When the electron is stored in a QD for a sufficiently long time, then it is completely dressed with all types of phonons, i.e. it is a generalized polaron. The next electron rapidly excited to this dot is however a bare electron, unless the excitation is performed adiabatically. The new bare electron in the dot does not satisfy the exact Pauli blockade since it is a different (undressed) particle, and we can refer this property to a temporal, partial limitation of the instant Pauli blockade. This limits spin-charge conversion schemes (using Pauli blocking) to times longer than ps.

Acknowledgments

Supported by the Polish Ministry of Scientific Research and Information Technology under the grant No 2 P03B 085 25 and No PBZ-MIN-008/P03/03

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- ¹ D. Bouwmeester, A. Ekert and A. Zeilinger, *The Physics of Quantum Information*, Springer Verlag 2000.
- ² E. Biolatti, Phys. Rev. Lett. **85**, 5647 (2000).
- ³ X. Li *et al.*, Science **301**, 809 (2003).
- ⁴ F. Troiani, E. Molinari and U. Hohenester, Phys. Rev. Lett. **90**, 206802 (2003).
- ⁵ S. De Rinaldis *et al.*, Phys. Rev. B **65**, 081309 (2002).
- ⁶ T. Calarco *et al.*, Phys. Rev. A **68**, 12310 (2003).
- ⁷ D. Loss and D. P. DiVincenzo, Phys. Rev. A **57**, 120 (1998).
- ⁸ G. Burkard, D. Loss and D.P. DiVincenzo, Phys. Rev. B **59**, 2070 (1999).
- ⁹ A. Imamoglu *et al.*, Phys. Rev. Lett. **83**, 4204 (1999).
- ¹⁰ T.H. Stievater *et al.*, Phys. Rev. Lett. **87**, 133603 (2001).
- ¹¹ H. Kamada *et al.*, Phys. Rev. Lett. **87**, 246401 (2001).
- ¹² H. Htoon *et al.*, Phys. Rev. Lett. **88**, 087401 (2002).
- ¹³ A. Zrenner *et al.*, Nature **418**, 612 (2002).
- ¹⁴ P. Borri *et al.*, Phys. Rev. B **66**, 081306 (2002).
- ¹⁵ M. Bayer *et al.*, Science **291**, 451 (2001).
- ¹⁶ *A Quantum Information Science and Technology Roadmap*, <http://qist.lanl.gov>
- ¹⁷ L. Jacak, P. Hawrylak and A. Wojs, *Quantum Dots*, Springer Verlag 1998.
- ¹⁸ J.M. Kikkawa and D.D. Awschalom, Phys. Rev. Lett. **80**, 4313 (1998).
- ¹⁹ S. Hameau *et al.*, Phys. Rev. Lett. **83**, 4152 (1999).
- ²⁰ L. Jacak *et al.*, Phys. Rev. B **65**, 113305 (2002).
- ²¹ O. Verzelen, R. Ferreira and G. Bastard, Phys. Rev. Lett. **88**, 146803 (2002).
- ²² L. Jacak *et al.*, Eur. Phys. J. D **22**, 319 (2003).
- ²³ H. Castella and R. Zimmermann, Phys. Rev. B **59**, R7801 (1999).

- ²⁴ A.A. Abrikosov, L.P. Gorkov and M.E. Dzyaloshinski, *Methods of Quantum Field Theory in Statistical Physics*, Moscow 1962 (in Russian).
- ²⁵ W.L. Bonch-Bruевич and S.W. Tiablikov, *Green Function Methods in Statistical Mechanics*, Moscow 1961 (in Russian).
- ²⁶ A. Suna, Phys. Rev. **135**, A111 (1964).
- ²⁷ A.S. Davydov, *Solid State Theory*, Moscow 1976 (in Russian).
- ²⁸ S.A. Moskalenko *et al.*, Fiz. Tverdogo Tela **10**, 356 (1968).
- ²⁹ S. Adachi, J. Appl. Phys. **58**, 1 (1985).
- ³⁰ L. Jacak, J. Krasnyj and W. Jacak, Phys. Lett. A **304**, 168 (2002).
- ³¹ D. Strauch and B. Dorner, J. Phys. Cond. Matter **2**, 1457 (1990).
- ³² P. Borri *et al.*, Phys. Rev. Lett. **87**, 157401 (2001).
- ³³ U. Bockelmann and G. Bastard, Phys. Rev. B **42**, 8947 (1990).