Dephasing of charge and spin in semiconductor quantum dots

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Phonon induced unavoidable decoherence of orbital degrees of freedom (charge) in quantum dots is studied and the relevant time scales are estimated for state-of-the-art self-assembled nanostructures. An significant enhancement of the effective Fröhlich constant due to localization is indicated. Temporal partial inefficiency of spin Pauli blocking in quantum dots, caused by lattice inertia, is predicted. For quantum dots placed in a diluted magnetic semiconductor medium a magnon-induced dephasing of spin localized in quantum dot is also estimated. The assessed decoherence rates for both orbital and spin degrees of freedom in semiconductor nanostructures turn out to not satisfy DiVincenzo conditions required for quantum error correction scheme implementations.

Keywords quantum dots; decoherence

1. Introduction

Recently the growing attention is paid to quantum infgrmation processing (QIP) [1] scenarios within solid state technology employing charge (orbital) [2,3] or spin [4,5] degrees of freedom of quantum dots (QDs) [6]. According to commonly accepted DiVincenzo conditions [1], QIP schemes require high degree of coherence of quantum states. Therefore, the most important problem emerging within the field of QD-based quantum computing is how to overcome decoherence in optically driven QD gates (orbital degrees of freedom) or how to accelerate spin single-qubit operations in magnetic-field-driven QD gates (spin Rabi oscillations are slow in semiconductors due to small value of a giromagnetic factor). As QDs are embedded in a surrounding crystal structure, the dominating role in decoherence (relaxation and dephasing) of orbital degrees of freedom is played by phonons.

Charges in QDs can be excited even on femtosecond time scale [7]. The fast (nonadiabatic) excitation of charges leaves the lattice in its ground state, leading to creation of a bare electron-hole pair. The eigenstates of an interacting carrier-phonon system correspond however to a composite quasi-particle of a polaronic type: an electron-hole pair accompanied by a phonon cloud (lattice polarization and deformation) [8,9]. This phonon cloud corresponds to the energy-minimizing state of the lattice. It is accompanied by a red-shift with respect to the original bare exciton energy. The excess energy is transferred from the QD region to the rest of the crystal, to the phonon subsystem, while locally in the QD a dressed polaron is formed which is a coherent composition of a bare exciton and a local phonon cloud. This combined carrier-phonon dynamics results in dephasing of the exciton quantum state, which may be observed as a decay of coherent polarization in optical experiments [10].

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

The relatively slow dressing of electrons with phonon clouds is also a source of the temporal inefficiency of Pauli blocking, important for possible spin-charge conversion schemes [4]. It is caused by a fact that a quickly (nonadiabatically) excited electron differs as a particle from an electron stored in a QD

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and thus already dressed with phonons, which limits fidelity of spin exclusion Pauli principle for too quick excitation.

Because of the inconvenient for QIP time scale of charge decoherence (dephasing) in QDs the growing interest is paid recently to spin degrees of freedom in QDs placed in a diluted magnetic semiconductor (DMS) medium (promising for the coherent spin control due to a giant giromagnetic factor). Carrier-induced ferromagnetism is observed in particular in *p* doped semiconductors *III-V* or *II-VI*. In magnetically ordered phase of DMS spin fluctuation however occur. These spin waves can play an analogous role for QD exciton spin as phonons for QD charge, and they can cause the similar decoherence effect—i.e. the dressing of QD-local spin degrees of freedom with collective spin waves in DMS. The temporal characteristics of dressing QD spin with DMS spin waves we compare with experimental data, in order to verify the model.

2. Dephasing of charge (exciton) in QD due to dressing with phonons

In order to investigate the time evolution of the non-stationary state corresponding to a rapidly (nonadiabatically) created QD exciton (in practice on sub-ps time scale [10]) we consider the Hamiltonian describing a single exciton interacting with phonons,

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

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

$$F_o(n_1,n_2,q) = -\frac{e}{q}\sqrt{\frac{2\pi\hbar\Omega}{v\tilde{\varepsilon}}} \int \Phi_{n_1}^*(R_e,R_h)\left(e^{iq\cdot R_e} - e^{iq\cdot R_h}\right)\Phi_{n_2}(R_e,R_h)\, d^3R_e\, d^3R_h \tag{2}$$

and

$$F_a(n_1,n_2,q) = -\sqrt{\frac{\hbar q}{2MC_a}} \int \Phi_{n_1}^*(R_e,R_h)\left(\sigma_e e^{iq\cdot R_e} - \sigma_h e^{iq\cdot R_h}\right)\Phi_{n_2}(R_e,R_h)\, d^3R_e\, d^3R_h. \tag{3}$$

Here $c_{q,s}^{(\dagger)}$ is the bosonic annihilation (creation) operator for LO or LA phonon with quasi-momentum q and with the frequency $\omega_o(q) \equiv \Omega_q \simeq \Omega - \beta q^2$ for LO phonons (Ω denotes the gap of LO phonons at the Γ point) and $\omega_a(q) = C_a q$, C_a—the sound velocity for LA phonons, M —the mass of ions in an elementary cell, $\sigma_{e,h}$—the deformation potential constant for electrons and holes, respectively, v—the volume of an elementary cell, N —the number of cells in the crystal, $\tilde{\varepsilon} = (1/\varepsilon_\infty - 1/\varepsilon_0)^{-1}$—the effective dielectric constant R_e, R_h are coordinates of the electron and hole, respectively, $\Phi_n(R_e,R_h)$ is the exciton wave function and $a_n^{(\dagger)}$ —annihilation (creation) operator of the exciton in the QD.

We will consider the exciton single-particle correlation function $\left\langle a_{n_1}(t) a_{n_2}^\dagger(0)\right\rangle$. For $n_1 = n_2$ it corresponds to the overlap of the excitonic state at time t with this state at the initial moment $t = 0$ (in particular of the *ground* state for $n_1 = n_2 = 0$). The modulus of this correlation function gives a measure of fidelity of the time dependent excitonic state. The Fourier transform of the correlation function,

$$I_{n_1 n_2}(\omega) = \int_{-\infty}^{\infty} \left\langle a_{n_1}(t) a_{n_2}^\dagger(0)\right\rangle e^{i\omega t} dt$$

is usually called the spectral density. It can be expressed by the imaginary part of the Green function,

$$ImG_r(n_1,n_2,\omega) = -I(n_1,n_2,\omega)/(2\hbar), \tag{5}$$

where

$$G_r(n_1,n_2,t) = -\frac{i}{\hbar}\Theta(t)\left\langle [a_{n_1}(t), a^{\dagger}_{n_2}(0)]_- \right\rangle = \frac{1}{2\pi}\int_{-\infty}^{\infty} G_r(n_1,n_2,\omega)e^{-i\omega t}d\omega \quad (6)$$

is the retarded Green function. The results of the analysis made (in [11]) according these lines can be presented in the Figs 1. and 2.

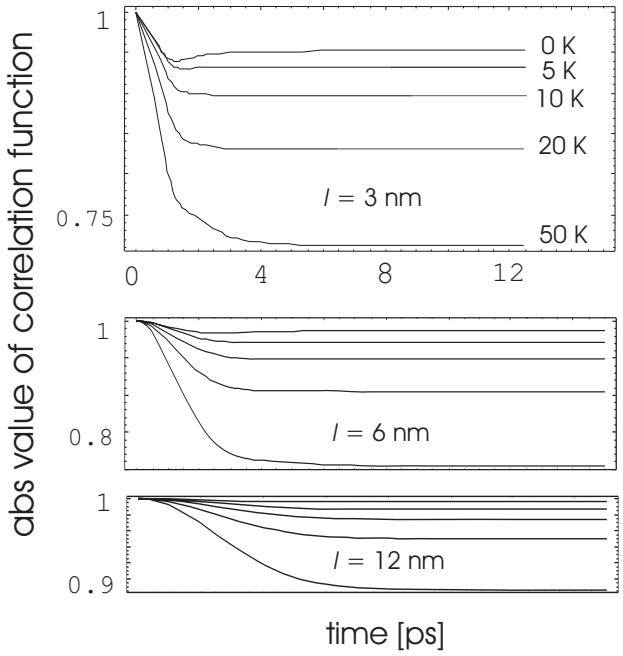

Fig. 1. Modulus of the correlation function $\left|\left\langle a(t)a^{\dagger}(0)\right\rangle\right|$ (the fidelity measure of the *ground* excitonic state) vs time for increasing temperatures, with LA phonons included. The three plots correspond to small, medium and large QDs and contain curves corresponding to the same set of temperatures as given in the upper plot. For the small QD, the experimentally observed fidelity loss for 0.2 ps pulse exciton in a QD [10] is well reproduced.

The coincidence of the time behavior of the fidelity measure (the correlation function) with that observed experimentally for 0.2 ps pulse excitation in a small QD [10] is good (Fig. 1). The LA channel of dressing is the most effective one and for typical QDs it gives a picosecond scale of dressing. The LO channel is slower and accompanied by oscillations (of ~ 100 fs scale) related to the gap in the LO phonon dispersion. The dephasing caused by LO phonons is significantly smaller than LA phonon dephasing, oppositely to the corresponding energy shifts [11]. Inclusion of the LO channel does not modify significantly the simultaneous LO and LA dephasing in comparison to LA channel solely (cf. Fig 2). The dressing time for LA and LO channels vs. dot size show linear and quadratic dependence for LA and LO channels, respectively. This property well corresponds to the qualitative picture of dressing with phonons, when the dressing time can be estimated as the dot dimension $\sim l$ divided by group velocity of phonons: C_a, for LA phonons, and $2\beta k \sim 2\beta/l$, for LO phonons. For LA channel it gives a linear dependence $\sim l/C_a$ while for LO channel a quadratic one $\sim l^2/(2\beta)$.

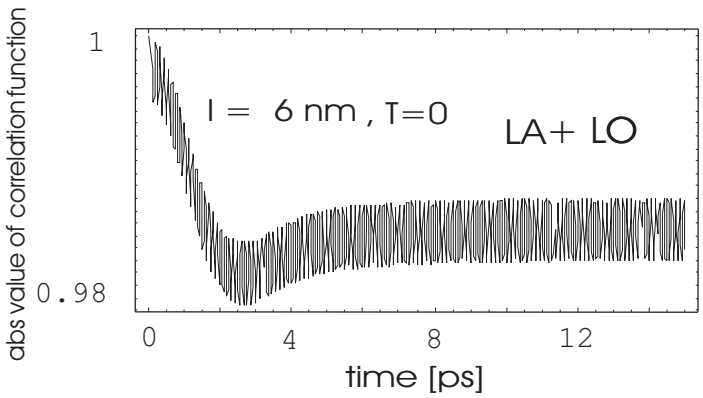

Fig. 2. The typical shape of the modulus of the correlation function for LO and LA phonons simultaneously. The oscillations correspond to the gap of LO phonon.

2.1. Renormalization of the Fröhlich constant for a QD

The Fröhlich constant [12], which describes interaction of LO phonons with electrons, is given by $\alpha_e = \frac{e^2}{\tilde{\varepsilon}}\sqrt{\frac{m^*}{2\hbar^3\Omega}}$. For GaAs bulk, $\varepsilon_0 = 12.9$, $\varepsilon_\infty = 10.9$, $m^* = 0.067 m_e$ and $\hbar\Omega = 36$ meV, then $\alpha_e = 0.07$. For a nanometer scale confinement of electrons, as in QDs, the recent experimental data (polaronic anti-crossing of spectra in magnetic field measured by far-infra-red spectroscopy) [8] indicated that $\alpha_e \sim 0.15$ (for QD of $\sim 10-15$ nm radius); for smaller QDs, the enhancement of α_e constant is even greater if one extracts it from the measured [13,14] Huang-Rhys coefficient [15] (this coefficient expresses a ratio of LO-satellite PL replica to main PL peak).

To account for the increase of α_e let us remind that the electron–LO phonon interaction in a polar material is caused by the action of a local polarization of the crystal on the electron, but only the *inertial* part of this local polarization is important. The *non-inertial* part, accompanying the moving electron, is included into the crystal field and thus already accounted for via definition of appropriate electron states. Therefore, the *inertial* part of the local polarization acting on the free-lattice electron is usually written as: $P(r) = P_0(r) - P_\infty(r)$, where $P_0 = \frac{\varepsilon_0 - 1}{4\pi\varepsilon_0} D, \quad P_\infty = \frac{\varepsilon_\infty - 1}{4\pi\varepsilon_\infty} D$, correspond to the static and the high-frequency polarizations, respectively ('high-frequency' means frequency much larger than the phonon frequencies but smaller than the frequencies of electrons in atoms; D is the electrical induction). $P(r) = D/(4\pi\tilde{\varepsilon})$, which results further, in a standard manner, in the formula for the Fröhlich constant.

For a confined electron, as in a QD, the *inertial* part of the polarization is, however, greater in comparison to a free-moving lattice electron. The physical reason for this is connected with the enhancement of a *quasi-classical* velocity of the electron placed in an additional confining potential. The lower limit of this velocity in a QD with the diameter d, equals to $v_d \simeq \hbar/(m^* d)$. This velocity is greater than the conduction band electron velocity (especially close to the Γ point, where it was zero), thus a smaller part of the local polarization can accompany the more quickly moving electron in a QD in comparison to a band electron. Therefore the *inertial* part of polarization for this electron increases. When confinement parameter d attains the atomic scale, given by a (a is the diameter of an elementary cell), then the lower limit for the quasi-classical velocity attains atomic scale $v_d = v_a \simeq \hbar/(m^* a)$. In the latter case

the *inertial* local polarization equals to P_0. Thus, if we assume that the *inertial* part of the polarization increases proportionally to the value of the quasi-classical electron velocity, from $P_0 - P_\infty$ for the free-lattice electron, up to P_0 for the electron confined on the atomic scale, we can write for this polarization $P'(r) = P_0(r) - \eta P_\infty(r),$ with some factor η ($0 \le \eta \le 1$), depending on the localization scale. It is clear that $\eta = 1$ when $d \to \infty$ and $\eta = 0$ when d attains dimensions of atoms, i.e. when $d \simeq a$. Therefore, within the linear approximation with respect to the small parameter a/d, we get $\eta = 1 - a/d$ and, for a confined electron, $P'(r) = D/(4\pi\tilde{\varepsilon}')$, with the effective dielectric constant $\tilde{\varepsilon}'$, $\frac{1}{\tilde{\varepsilon}'} = \frac{1-a/d}{\varepsilon_\infty} - \frac{1}{\varepsilon_0} + \frac{a}{d}$. This formula leads to the renormalized Fröhlich constant with $\tilde{\varepsilon}$ substituted by $\tilde{\varepsilon}'$. For an InAs/GaAs QD with a radius of order of 10 nm (i.e. $d \approx 20$ nm), the renormalized Fröhlich constant ≈ 0.15 in agreement with FIR spectroscopy measurement for such a dot [8] . The enhancement of electron-LO phonon interaction for QDs manifests itself also via a significant increase of the Huang–Rhys factor [15] for a satellite LO phonon-assisted photoluminescence feature in QDs (InAs/GaAs) [13,14]. The Huang–Rhys parameter scales as α_e (some further corrections result from the different Fröhlich constant for electrons and holes due to distinct effective mass). For dots of diameter of order of $5 - 9$ nm [14], the corresponding $\alpha_e \sim 0.4 - 0.3$, and for dots with diameter $\sim$ $15 - 19$ nm [13], $\alpha_e \sim 0.25 - 0.18$. In the former case it gives the factor 6 – 5 and in the latter 4 – 3 for the Huang-Rhys parameter, which agrees with experimental data.

2.2. Fidelity restrictions for spin Pauli blocking in QD due to phonon induced dephasing

Phonon induced dephasing of orbital degrees of freedom in QDs leads also to spin-related decoherence. Dressing of charge excitations with phonons causes temporal inefficiency of Pauli exclusion principle in the charge-spin conversion scheme needed for QIP in hybrid spin-charge solutions [4], where an electron spin in a QD would be used for storage of information while qubit operations would be performed on charge degrees of freedom. The charge-spin conversion in this proposal is done by employing spin Pauli blocking: when the state in QD is occupied by an electron, only an electron with the opposite spin can be excited to this state. However, a quickly (non-adiabatically) excited electron is a bare one, while another electron stored in the QD is already dressed with phonons and thus they are different particles. This difference causes a temporal inefficiency, of order of 10% in magnitude, of spin blocking unless the electron is excited slower, in an adiabatic manner, in order to allow for some inertia of the phonon system. Thus the time of dressing of the charge with phonons (several ps in typical QDs) determines the lower limit for a rate of adiabatic spin-charge switching.

3. Dephasing of spin of carrier localized in QD due to dressing with magnons in diluted magnetic semiconductor medium

Recently, much attention has been focused on QDs placed in diluted magnetic semiconductors (DMS) [16-20] of the type *III-V* (e.g. Ga(Mn)As) or *II-VI* (e.g. Zn(Mn)Se). In DMS, part of cations is randomly substituted by ions of a transition metal. The spin localized on admixtures can interact with spins of carriers (holes in that case) which leads to a ferromagnetic transition controlled by hole concentration [17]. Simultaneously, expectations arose that giant enhancement of effective giromagnetic factor in ferromagnetic phase could accelerate QIP on exciton spins in QDs in a DMS medium (e.g. in self-assembled quantum dots Ga(Mn)As/InAs or Zn(Mn)Se/CdSe [18-20]).

By the analogy to the decoherence of a non-adiabatically created QD exciton due to dressing of charges with phonons and formation of a polaron in QD, one can expect the similar dressing of the exci-

ton spin with a cloud of magnons (in magnetically ordered phase of DMS) and formation of an excitonic-magneto-polaron (EMP). The Hamiltonian for an exciton which interacts with the spin wave subsystem in surrounding DMS is analogous to that one for an exciton with phonons. The exciton–spin wave interaction has a similar three particle form corresponding to spin flip of e-h components of exciton and simultaneous absorption or emission of a magnon to DMS host medium. The resulting decoherence due to the dressing of the exciton spin with magnons corresponds to the formation of EMP localized in the QD. The time of formation of this magneto-polaron is governed by the dispersion of magnons. It is given by the ratio of the dot dimension and magnon group velocity, similarly as for the formation of ordinary excitonic–polaron [8,9,11]. The rapidly (i.e. nonadiabatically) excited QD bare exciton is not a stationary state of the total system including surrounding DMS medium. The formation of hybridized state of the exciton spin with the cloud of spin waves corresponds to transfer of an excess energy (of spin exchange interaction) outside the QD region. Though energy of the corresponding nonstationary state is not determined, the mean energy is shared in time between QD subsystem and the rest of the crystal (the localized in QD eventually dressed particle, EMP, has lower energy that the initial bare exciton). This process cannot be accounted for within a scheme of Fermi-golden-rule-transitions as it is not a transition between stationary states, but rather the evolution of a nonstationary state. In order to estimate a time of formation of the local dressed particle (EMP in QD) we apply a phenomenological picture, motivated within Green function approach (the kinetics of this dressing is displayed by the out-of-pole spectral intensity properties given by the imaginary part of the retarded Green function) [11]. In this picture the time scale of dephasing is of order of the ratio of the dot diameter and the group velocity of the collective mode (spin wave in DMS). It is the velocity of the spin wave packed, which transfers the excess of interaction energy to surrounding medium, outside the QD. Dressing of the exciton spin with magnons is similar to dressing of charge with LO phonons, since both LO phonons and magnons have a quadratic dispersion [11] (cf. Fig. 3 (right)):

$$\begin{cases} e_1(\vec{q}) = e_0 - b\,q^2, \\ e_2(\vec{q}) = b\,q^2. \end{cases}$$

Thus, the time of dressing with magnons scales as d^2 (d is the QD dimension), and it is relatively long for typical dot dimensions. Using the formulae for spin wave energies one can obtain the following assessment for the time of formation of the EMP:

$$\tau \simeq \frac{d}{v_g} = d\left(\frac{\partial e}{\partial q}\right)^{-1} \simeq \frac{\hbar d^2}{2b}$$

(for QDs important are only modes with $q \approx \frac{1}{d}$, due to a bottle-neck effect). This dephasing time of the exciton spin depends on magnetic admixture and band-hole concentration in the DMS via *b* coefficient [11]. Some examples are shown in the Fig. 3. For QD Zn(Mn)Se/CdSe with dimension ~ 10 nm and $x = 0.25$, $x_p = 0.025$ for DMS, the spin dephasing time is of order of 150 ps, which coincides with the experimentally observed timing of formation of EMP in $Zn_{0.75}Mn_{0.25}Se$/CdSe (for this sample, the time of exciton annihilation is sufficiently longer ~ 600 ps [18-20], which allows for complete formation of EMP).

One can thus state that even though the DMS medium of QDs could allow for significant acceleration of QD spin control in ferromagnetic DMS phase (due to a giant Pauli term), promising for spin qubit definition for quantum information processing, however the spin wave induced decoherence also considerably grows. Decoherence (dephasing) of the spin degrees of freedom in QD due to the dressing of spin of carriers trapped in a QD with spin waves from hosting DMS material is similarly inconvenient for quantum information processing as phonon induced dephasing of charges in QDs [11].

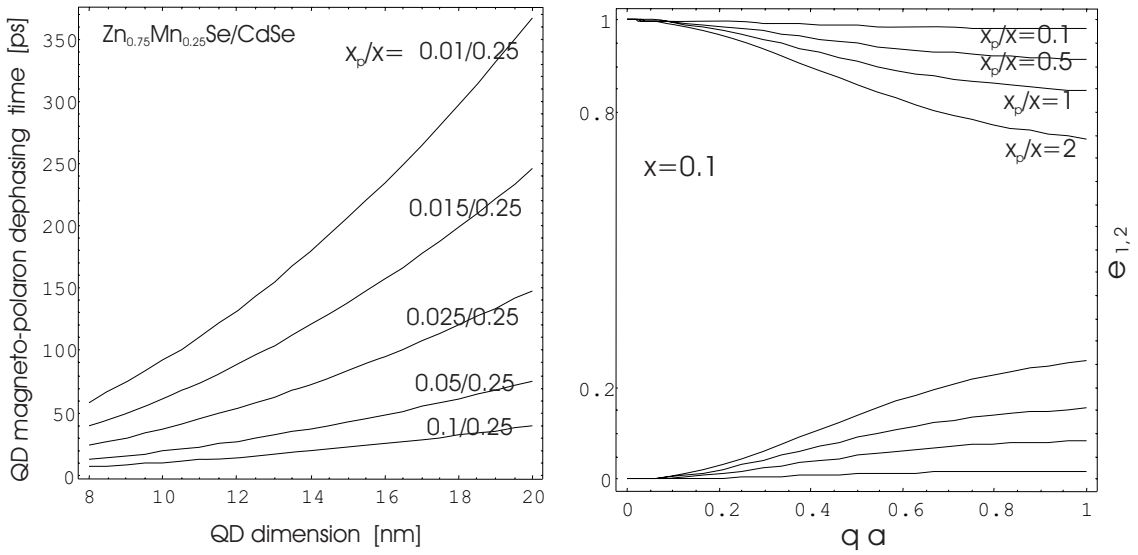

Fig.3. EMP formation time $\tau = \frac{d}{v_g}$, i.e. the spin dephasing time of an exciton localized in QD embedded in DMS, versus dot dimension d for various hole concentration rates in DMS/QD structure (x_p - hole concentration in DMS, x - Mn concentration in DMS) (left); spin waves dispersion in DMS for several hole/dopant concentrations [11] (a - cell constant) (right).

4. Conclusions

The phonon induced decoherence of orbital degrees of freedom in QDs is studied in order to verify the feasibility of quantum information processing within QD technology. The source of an unavoidable partial leakage of 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 (sound velocity). For a polar medium (e.g. self-assembled InAs/GaAs dots) polarization effects (LO phonons) contribute to dephasing but this process is characterized by a longer time scale ($\sim$ 100 ps), proportional to the square of the QD diameter. Anharmonicity effects (e.g. LO-TA in GaAs) enhance the efficiency of polaron formation to a few ps time scale [21]. Dressing time scale, inconveniently located between the possible sub-ps operation time and the ns exciton recombination time, probably precludes feasibility of implementing QIP error correction schemes for all-optically controlled gates in QD technology. The relatively slow dressing of charge with phonon clouds is also a source of the temporal inefficiency of Pauli blocking (in spin-charge conversion schemes), as a quickly excited electron differs from an electron stored in a QD already dressed with phonons. In so-called magnetic QDs, i.e. dots placed in a diluted magnetic semiconductor medium (promising for 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 order of 150-200 ps, due to weak quadratic magnon dispersion similarly as for LO phonons). Thus dephasing of QD spin due to magnons in DMS is similarly inconvenient for QIP as dephasing of QD charge due to phonons. The depahasing rates for QD spin and charge seem to strongly limit possibilities for scaling of QIP gates in QD solid state technology.

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