Reducing of magnon-induced spin pure dephasing in quantum dots at low temperatures

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We demonstrate disappearance at $T=0$ (and strong reduction at low temperatures) of the magnon-induced spin pure dephasing of an exciton located in a quantum dot embedded in a magnetic medium (e.g., in diluted magnetic semiconductor)—in contrast to the charge dephasing by phonons, which remains strong even at $T=0$.

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I. INTRODUCTION

The experimental realizations of charge Rabi oscillations in quantum dots (QDs) and optically driven QD gates[1–3] encountered strong fidelity limitations caused by the lattice inertia. The most destructive are bulk longitudinal acoustical (LA) phonons (due to their wide dispersion), and hybridization of QD charge with the LA phonons results in a picosecond scale of dephasing,[4–6] pronounced also at low temperatures,[4] and inconveniently located in the middle between the quickest femtosecond time scale of control and nanosecond scale of exciton relaxation. The charge dephasing in QDs may be observed as a decay of coherent polarization in optical subpicosecond experiments.[5,6] Since the unfavorable time scale of phonon-induced dephasing of charges in QDs, the growing interest is paid to spin degrees of freedom in QDs (Refs. 7 and 8) that are not directly affected by phonons. Decoherence of QD spin in nonmagnetic media is caused by interaction with nuclear spins from surroundings and by spin-orbit coupling (the latter is important in multielectron QDs).[9] Both interactions are weak, which results in relatively small and slow spin-decoherence rates. The drawback of spin is, however, a very slow spin control available in nonmagnetic semiconductors due to a small value of a gyromagnetic factor in typical materials (e.g., in GaAs the Zeeman splitting is only ~ 0.03 meV/T). In order to accelerate the control over spin, the magnetic materials can be used. Especially interesting are nanostructures embedded in diluted magnetic semiconductors[10] (DMS), promising for acceleration of the spin control due to a giant increase in the gyromagnetic factor in a magnetically ordered phase of a DMS. The convenient carrier-induced ferromagnetism[11] is observed in DMS, in particular in III(Mn)V or in p-doped II(Mn)VI.[10,11]

In a magnetically ordered phase of a DMS, spin fluctuations however occur[12,13] and these magnons (spin waves) can play an analogous destructive role for QD spin as phonons do for QD charge. The magnon-induced QD spin decoherence is stronger than decoherence caused by nuclear spins or spin-orbit interaction since the energy scale of spin-exchange interaction is large[10] (~ 1 eV). Thus, in spite of possible acceleration of spin control in a DMS medium, spin decoherence induced by magnetic surroundings also grows inconveniently. In the present Brief Report we show that QD spin dephasing caused by magnons can be however frozen at low temperatures, which again supports the advantage of spin in a magnetic medium for coherent control. We prove that QD spin pure dephasing caused by magnons disappears at $T=0$, which allows for minimizing of this dephasing up to an arbitrarily low threshold by lowering the temperature, unlike the phonon-induced QD charge dephasing being strong even at $T=0$. We describe dressing of local QD spin with magnons accompanying the formation of an exciton-magnetic polaron (EMP) localized in the QD and explain the difference between magnon- and phonon-assisted decoherence phenomena. The advantage of spin over charges for coherent control at low temperatures is caused by the spin-conservation rule and thus is of a general character not addressed to a selected material only.

The dephasing of QD instant excitation (electron or exciton) corresponds to a time evolution of the nonstationary state—the bare excitation in the initial moment and gradually dressed next with inertial collective excitations of the surrounding crystal. This process, beyond the scheme of Fermi-golden-rule transitions, corresponds to the evolution of an entangled nonstationary state which inseparably links local and global degrees of freedom of the QD and the host material. It can be illustrated via appropriate correlation functions playing the role of a fidelity measure.

II. CORRELATION FUNCTION

The time evolution of the QD exciton state can be described by the exciton single-particle correlation function $\langle a_{n_1}(t)a^{+}_{n_2}(0)\rangle$ [$a^{(+)}_n$ annihilation (creation) operator of a QD exciton in the state n]. 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 Fourier transform of the correlation function, $I_{n_1,n_2}(\omega)=\int_{-\infty}^{\infty}\langle a_{n_1}(t)a^{+}_{n_2}(0)\rangle e^{i\omega t}dt=-2\hbar\,\mathrm{Im}\,G^{r}_{n_1,n_2}(\omega)$, is the spectral intensity related to the retarded Green's function $[G^{r}_{n,n}(\omega)]^{-1}=\hbar\omega-E_n-\Delta_n(\omega)+i\gamma_n(\omega)$ and its imaginary part $\mathrm{Im}\,G^{r}=-a^{-1}\pi\delta(x)-\frac{a^{-1}\gamma'(x)/x^2}{1+[\gamma'(x)/x]^2}$ (a is a residuum in the pole, $x=\hbar\omega-E-\Delta$, and $\gamma'=\frac{\gamma}{a}$).

For short-time scale the imaginary part of the retarded Green's function is proportional to the imaginary part of the mass operator (γ can be omitted in the denominator of $\mathrm{Im}\,G^r$ for large ω). For weak exciton-phonon coupling, the lowest contribution to the imaginary part of the exciton mass operator is given by a double-vertex graph with three-particle exciton-phonon interaction [cf. Fig. 1(a)]. For the ground

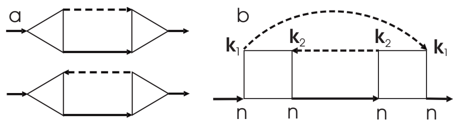

FIG. 1. The lowest-order contribution to the QD exciton mass operator (its imaginary part) due to interaction (a) with phonons [emission (upper), absorption (lower)] and (b) with magnons in DMS, [dashed line: (a) phonon and (b) magnon; solid line: exciton; and the exciton quantum numbers (j,n,s_z) suppressed to n].

state it is given by the equation (to main order) $\gamma \sim \int dk|F(k)|^2\delta[\omega-E_0-\omega(k)]$, with an interaction vertex $F(k)\sim\langle\Psi_0|e^{ikr}|\Psi_0\rangle$, where Ψ_0 is the QD carrier (exciton) wave function of the ground state (here, for simplicity, single particle in one dimension, $\hbar=1$). Thus, the correlation function, $I(t)\sim e^{-iEt}\int dr|\Psi_0(r)|^2\int dkF^*(k)e^{i[kr-\omega(k)t]}$, has the form of a time dependent overlap of QD-particle distribution $|\Psi_0(r)|^2$, with the collective excitation wave packet $\int dkF^*(k)e^{i[r-\partial\omega(k)/\partial kt]k}$ [with k center $\sim 1/d$ (d is the QD diameter) due to the QD bottleneck effect[14,15] entered here via $F(k)$]. This packet escapes from the QD-space region with the group velocity $v_g=\frac{\partial\omega(k)}{\partial k}$ (at $k\sim 1/d$). The dephasing time corresponding to the time scale of decreasing $I(t)$ is thus of the order of $\tau\simeq\frac{d}{v_g}$. The linear dispersion of LA phonons results in constant group velocity and then in linear scaling of τ versus d (upper part of Fig. 2), while for the longitudinal optical (LO) phonons their quadratic dispersion (close to the Γ point) gives $\tau\sim d^2$ (lower part of Fig. 2). In order to apply a similar approach to dressing of a QD spin with magnetic fluctuations, the dispersion of spin waves in magnetic medium is needed.

III. SPIN WAVES IN DMS

For the description of a DMS, a model of indirect (mediated by band holes) exchange of magnetic dopants is commonly adopted[10,11,16] with the Hamiltonian $\hat{H}=\hat{H}_{pd}+\hat{H}_p$, where $\hat{H}_{pd}=-2\Sigma_{j=1}^{N_p}\Sigma_{\mathbf{n}}A_p(\mathbf{R}_j-\mathbf{R}_{\mathbf{n}})\hat{\mathbf{s}}_j\cdot\hat{\mathbf{s}}_{\mathbf{n}}$ describes the spin subsystem of DMS (here $\hat{\mathbf{s}}_j,\mathbf{R}_j$ and $\hat{\mathbf{s}}_{\mathbf{n}},\mathbf{R}_{\mathbf{n}}$ are spin operators and positions of band hole and impurity atom, respectively; $\mathbf{n}$ summation goes over lattice points occupied by magnetic dopants; and N_p is the number of band holes). $A_p(\mathbf{R}_j-\mathbf{R}_{\mathbf{n}})$ is p-d exchange integral [$A_p<0$ (antiferromagnetic) and typically $|A_p|\sim 1$ eV]. $\hat{H}_p$ is the fermionic band-hole Hamiltonian—a subject for modeling of the system, e.g., for III(Mn)V or p-doped II(Mn)VI structures.[12,16]

Changing to the Holstein-Primakoff (HP) bosonic representation for dopant and hole spins, $\hat{S}_{+\mathbf{n}}\simeq\sqrt{2S}\hat{B}_{\mathbf{n}}$, $\hat{S}_{-\mathbf{n}}\simeq\sqrt{2S}\hat{B}_{\mathbf{n}}^+$, and $\hat{S}_{z\mathbf{n}}=S-\hat{B}_{\mathbf{n}}^+\hat{B}_{\mathbf{n}}$, where $\hat{B}_{\mathbf{n}}^{(+)}$—HP operators for dopants—and assuming that hole spin alignment is opposite (similarly as in ferrite),[16] $\hat{s}_{+j}\simeq\hat{b}_j$, $\hat{s}_{-j}\simeq\hat{b}_j^+$, and $\hat{s}_{zj}=-\frac{1}{2}+\hat{b}_j^+\hat{b}_j$, where $\hat{b}_j^{(+)}$—HP operators for holes (at low temperatures only main terms in the HP representation are left). After integrating out fermionic degrees of freedom and averaging over random distributions of dopants, which restores translational symmetry in a continual limit (as was done in Ref. 12), one can diagonalize $\hat{H}_{pd}$ (in the Fourier picture) via the transformation $\hat{B}(\mathbf{k})=v_{\mathbf{k}}\hat{\alpha}_1(\mathbf{k})+u_{\mathbf{k}}\hat{\alpha}_2(\mathbf{k})$ and $\hat{b}(\mathbf{k})=u_{\mathbf{k}}\hat{\alpha}_1(\mathbf{k})-v_{\mathbf{k}}\hat{\alpha}_2(\mathbf{k})$. It leads to two branches of spin waves (as in ferrite): $\varepsilon_1(\mathbf{k})\hat{\alpha}_1^+(\mathbf{k})\hat{\alpha}_1(\mathbf{k})+\varepsilon_2(\mathbf{k})\hat{\alpha}_2^+(\mathbf{k})\hat{\alpha}_2(\mathbf{k})$, $\varepsilon_1(\mathbf{k})=Dk^2$, and $\varepsilon_2(\mathbf{k})=D_0-Dk^2$, with $D_0=-\tilde{A}_p(0)(x_p+2Sx)$ and $D=-\tilde{A}_p(0)\frac{2Sxx_p}{x_p+2Sx}l_{ex}^2$ (l_{ex} is the range of the exchange, x,x_p are the concentration of magnetic dopants and of holes in DMS, respectively, $\tilde{A}_p(0)$ is the zeroth Fourier component of A_p, and $S=5/2$ for Mn).[12]

IV. EXCHANGE INTERACTION WITH QD EXCITON

The exchange spin interaction of QD exciton with magnetic dopants in DMS (band holes do not contribute) has the form

$$\hat{H}_{sd}(\mathbf{R}_e,\mathbf{R}_h)=-2\beta_0\sum_n A_e(\mathbf{R}_e 2-\mathbf{R}_n)\hat{\mathbf{s}}_e\cdot\hat{\mathbf{S}}_{\mathbf{n}} -2\beta_0\sum_{\mathbf{n}} A_h(\mathbf{R}_h-\mathbf{R}_n)\hat{\mathbf{s}}_h\cdot\hat{\mathbf{S}}_{\mathbf{n}}, \qquad (1)$$

where $\hat{\mathbf{s}}_{e(h)}$ is the spin operator of the electron (hole) of the QD exciton and $\hat{\mathbf{S}}_{\mathbf{n}}$ is the spin operator of the magnetic dopant in $\mathbf{R}_{\mathbf{n}}$ position. The factor $A_{e(h)}[\mathbf{R}_{e(h)}-\mathbf{R}_{\mathbf{n}}]$ describes the exchange spin interaction between the electron (hole) of the QD exciton and the dopant (with a phenomenological coefficient β_0 accounting for an additional decrease in the exchange due to dot-structure separation).

By $a_{jns_z}^{(+)}$ we denote the bosonic annihilation (creation) operator of the QD exciton in the state (jns_z) ($j=1,2$ for the opposite and the same spin alignment in an e-h pair, respectively, and s_z is the spin orientation of the electron in e-h pair). These states are split in spin structure in the ordered phase of DMS and $(1,0,\frac{1}{2})$ is the ground state. The Hamiltonian (1) can be rewritten in the basis of these excitonic states. Taking the HP representation for dopant spins and averaging over random distributions of dopants and then changing to the magnons (via the diagonalization transfor-

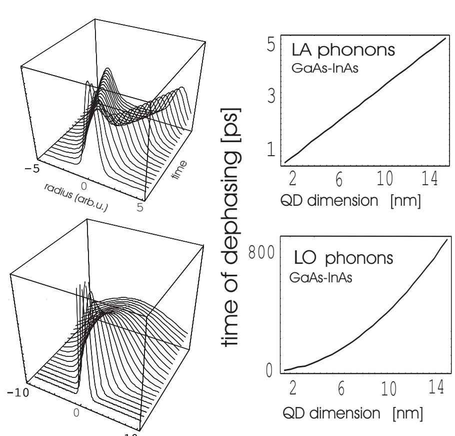

FIG. 2. Time evolution of phonon wave packet transferring excess deformation (upper) and polarization (lower) energy out of QD region during local polaron formation and corresponding dephasing time versus dot dimension.

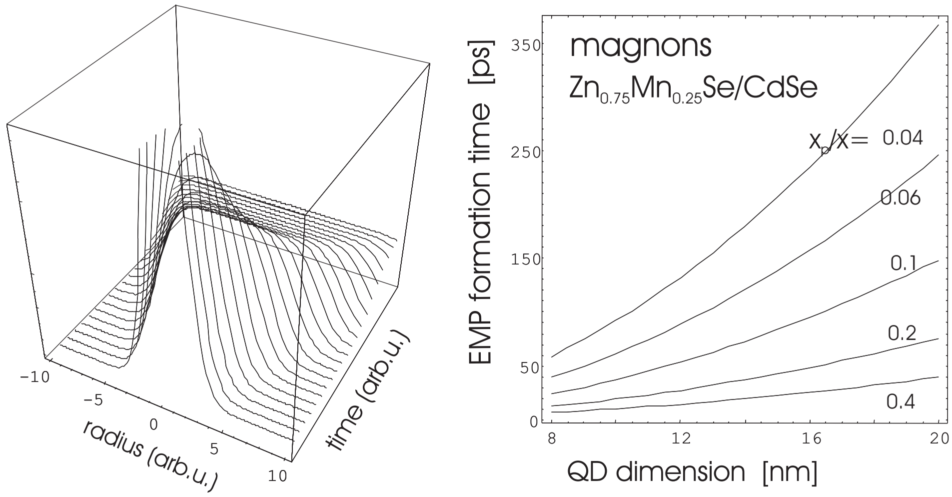

FIG. 3. Time evolution of magnon wave packet transferring excess exchange energy out of QD region during local EMP formation and corresponding dephasing time for various hole concentration rates (in $Zn_{0.75}Mn_{0.25}Se$-CdSe).

mation), we arrive with $\hat{H}_{sd}=\hat{H}^1_{sd}+\hat{H}^2_{sd}$, where in the first term two magnon operators are involved,

$$\hat{H}^1_{sd}=-2Sx\beta_0\sum_{\mathbf{k}_1}\sum_{\mathbf{k}_2}[v_{\mathbf{k}_2}\hat{\alpha}^+_1(\mathbf{k}_2)+u_{\mathbf{k}_2}\hat{\alpha}^+_2(\mathbf{k}_2)]$$
$$\times[v_{\mathbf{k}_2+\mathbf{k}_1}\hat{\alpha}_1(\mathbf{k}_2+\mathbf{k}_1)+u_{\mathbf{k}_2+\mathbf{k}_1}\hat{\alpha}_2(\mathbf{k}_2+\mathbf{k}_1)]$$
$$\times\sum_{n,n',s_z}s_z\{[F^e_{nn'}(\mathbf{k}_1)-F^h_{nn'}(\mathbf{k}_1)]\hat{a}^+_{1ns_z}\hat{a}_{1n's_z}+[F^e_{nn'}(\mathbf{k}_1)$$
$$+F^h_{nn'}(\mathbf{k}_1)]\hat{a}^+_{2ns_z}\hat{a}_{2n's_z}+[F^e_{nn'}(\mathbf{k}_1)\hat{a}^+_{1ns_z}\hat{a}_{2n's_z}+\mathrm{H.c.}]$$
$$+[F^h_{nn'}(\mathbf{k}_1)\hat{a}^+_{1ns_z}\hat{a}_{2n'-s_z}+\mathrm{H.c.}]\},\qquad(2)$$

while in the second term only single,

$$\hat{H}^2_{sd}=-\sqrt{2Sx}\beta_0\sum_{\mathbf{k}}\sum_{n,n'}\{[v_{\mathbf{k}}\hat{\alpha}_1(\mathbf{k})+u_{\mathbf{k}}\hat{\alpha}_2(\mathbf{k})][F^e_{nn'}(\mathbf{k})$$
$$\times(\hat{a}^+_{1n1/2}\hat{a}_{1n'-1/2}+\hat{a}^+_{2n1/2}\hat{a}_{2n'-1/2}+\hat{a}^+_{1n1/2}\hat{a}_{2n'-1/2}$$
$$+\hat{a}^+_{2n1/2}\hat{a}_{1n'-1/2})+F^h_{nn'}(\mathbf{k})(\hat{a}^+_{1n-1/2}\hat{a}_{1n'1/2}+\hat{a}^+_{2n1/2}\hat{a}_{2n'-1/2}$$
$$+\hat{a}^+_{1n1/2}\hat{a}_{2n'-1/2}+\hat{a}^+_{2n1/2}\hat{a}_{1n'-1/2})]+\mathrm{H.c.}\}.\qquad(3)$$

$F^{e(h)}_{n,n'}(\mathbf{k})=\tilde{A}_{e(h)}(\mathbf{k})\int d^3R_e\int d^3R_h\Psi^*_n(\mathbf{R}_e,\mathbf{R}_h)e^{i\mathbf{k}\cdot\mathbf{R}_{e(h)}}\Psi_{n'}(\mathbf{R}_e,\mathbf{R}_h)$, $\tilde{A}_{e(h)}$ is the Fourier picture of $A_{e(h)}$, and Ψ_n is the orbital part the nth state QD exciton wave function. One can select now the term $\hat{H}^{1,0}_{sd}$, which contributes to the pure dephasing—it is only in $\hat{H}^1_{sd}$ ($\hat{H}^2_{sd}$ does not contribute to the pure dephasing since it does not conserve the exciton state),

$$\hat{H}^{1,0}_{sd}=-2Sx\beta_0\sum_{k_1,k_2}[v_{\mathbf{k}_2}\hat{\alpha}^+_1(\mathbf{k}_2)+u_{\mathbf{k}_2}\hat{\alpha}^+_2(\mathbf{k}_2)]$$
$$\times[v_{\mathbf{k}_1}\hat{\alpha}_1(\mathbf{k}_1)+u_{\mathbf{k}_1}\hat{\alpha}_2(\mathbf{k}_1)]\sum_{n,n',s_z}s_z[F^e_{nn'}(\mathbf{k}_1-\mathbf{k}_2)$$
$$-F^h_{nn'}(\mathbf{k}_1-\mathbf{k}_2)]\hat{a}^+_{1ns_z}\hat{a}_{1n's_z}.\qquad(4)$$

$\hat{H}^{1,0}_{sd}$ corresponds (for $n=n'=0$, $s_z=1/2$) to the interaction without any change of the (ground state) exciton spin (neither of the electron nor of the hole in e-h pair)— thus, the spin of created (annihilated) magnon must be taken off by an annihilated (created) additional magnon (spin is conserved—it is the difference in comparison to phonons). The rest of $\hat{H}_{sd}$ describes e-h spin-flip processes (including those with only single magnon involved in $\hat{H}^2_{sd}$) not contributing to the pure dephasing as not conserving the exciton state. With the term $\hat{H}^{1,0}_{sd}$ one can associate the vertex, which gives an imaginary part of the mass operator, cf. Fig. 1(b), leading to the pure dephasing of the exciton spin.

V. PURE DEPHASING OF SPIN

The imaginary part of the mass operator given by the graph in Fig. 1(b) has the form $\gamma_n(\omega,T)=\pi\Sigma^2_{i,j=1}\Sigma_{\mathbf{k}_1,\mathbf{k}_2}|V(n,\mathbf{k}_1,i;n,\mathbf{k}_2,j)|^2[n_i(\mathbf{k}_1)+1]n_j(\mathbf{k}_2)\delta[\hbar\omega-E_n-\varepsilon_i(\mathbf{k}_1)+\varepsilon_j(\mathbf{k}_2)]$ [here n comprises all quantum numbers (j,n,s_z); $n=0$ for $(1,0,\frac{1}{2})$] and

$$V(n,\mathbf{k}_1,i;n,\mathbf{k}_2,j)|_{n=0}=2Sx\beta_0\begin{pmatrix}v_{\mathbf{k}_2}v_{\mathbf{k}_1}, & v_{\mathbf{k}_2}u_{\mathbf{k}_1}\\ u_{\mathbf{k}_2}v_{\mathbf{k}_1}, & u_{\mathbf{k}_2}u_{\mathbf{k}_1}\end{pmatrix}$$
$$\times[F^e_{00}(\mathbf{k}_1-\mathbf{k}_2)-F^h_{00}(\mathbf{k}_1-\mathbf{k}_2)];$$

the matrix is addressed to the magnon branches $[ij]$, $n_i(\mathbf{k})$ is the Bose-Einstein distribution for the ith magnon branch, and $u_{\mathbf{k}}$, $v_{\mathbf{k}}$ are the coefficients of the magnon diagonalization transformation (for small k, $u^2_k=\frac{x_p}{x_p+2Sx}-Bk^2$, $v^2_k=\frac{2Sx}{x_p+2Sx}+Bk^2$, and $B=\frac{x_p}{x}\frac{2S-x_p/x}{(2S+x_p/x)^3}2Sl^2_{ex}$).[12]

The above formulae allow for direct calculation of the correlation function $I(t)$ and estimation of the spin-dephasing timing (Fig. 3) [for Zn(Mn)Se-CdSe structure: $\tilde{A}_h(0)\simeq\tilde{A}_p(0)=-1.3$ eV, $\tilde{A}_e(0)=0.26$ eV, and $\beta_0=0.1$].[17]

The dressing of the localized spin with magnons results in a relatively long-time scale of dephasing (due to the weak quadratic magnon dispersion near Γ point, similarly for LO phonons) of the order of 150 ps for $d\sim10$ nm (Fig. 3), which coincides with the experimentally observed timing of formation of EMP in $Zn_{0.75}Mn_{0.25}Se$-CdSe.[17,18] The overall time scale for QD spin kinetics is shifted by three orders of magnitude to longer periods in comparison to QD charge, but again with the dephasing time fallen in the middle between control and relaxation.

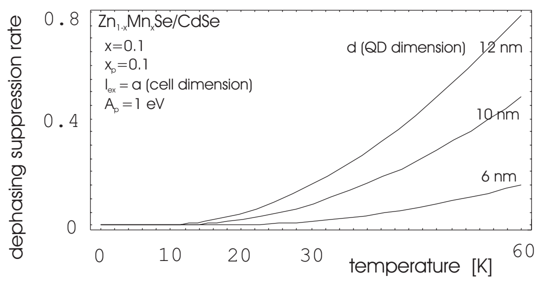

FIG. 4. The freezing of spin pure dephasing for several QD dimensions (in $Zn_{1-x}Mn_xSe$-CdSe).

Nevertheless, due to spin conservation in the vertex [cf. the form of spin-exchange (1)], two magnons must simultaneously take part in the interaction responsible for the dressing of the exciton state with magnon cloud (pure dephasing)—cf. Eq. (4). It causes, however, the temperature dependent factors $[n_i(k_1)+1]n_j(k_2) \xrightarrow{T\to 0} 0$ in the equation for $\gamma(\omega,T)$. These factors preclude pure dephasing of exciton spin at $T=0$ and reduce its amplitude at low temperatures (unlike the charge pure dephasing by phonons, which remains strong even at $T=0$ owing to the emission term with the factor $1+n(k) \xrightarrow{T\to 0} 1$). Note that for magnons at low temperatures, $n_1 \gg n_2$ due to the magnon gap and only $n_1(1+n_i)$ $(i=1,2)$ contributes. The freezing of spin dephasing caused by these factors is depicted in Fig. 4 (for typical QD/DMS parameters the freezing region can reach $T \sim 10$ K but it is sensitive to the dot dimension).

VI. CONCLUSIONS

We proved that the pure dephasing of the exciton spin in QD embedded in DMS disappears at $T=0$ and can be arbitrarily suppressed in amplitude by lowering the temperature, in contrast to the charge dephasing by phonons being always strong even at $T=0$. This property has a more general character (applies to any local spin dephasing induced by collective spin excitations in magnetic medium) since it originates from the conservation of spin at vertex; thus, the creation of magnon must be accompanied with the annihilation of another one (or conversely) unless the state of local spin had been changed (not pure dephasing). It results in the different low-temperature behavior in comparison to phonon dephasing of QD charge.

The dressing of the localized spin with magnons leads to the relatively long dephasing time scale, of the order of several hundreds of picoseconds, due to the weak quadratic magnon dispersion similarly for LO phonons. The overall time scale for QD spin kinetics is shifted by three orders of magnitude to longer periods in comparison to QD charge, but again (similarly as for QD charge) with the dephasing time unfavorably located right in the middle between control and relaxation. Thus, the reduction in amplitude of QD spin dephasing at low temperatures is of primary importance and supports an idea of some advantages of spin over charge for coherent control in QDs.

ACKNOWLEDGMENT

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