Temperature Dependence of Exciton Spin Relaxation Rates in Semiconductor Quantum Dots

A. Reznitsky[1], A. Klochikhin[1,2], S. Permogorov[1], L. Tenishev[1], K. Mironenko[1], E. Tsitsishvili[3], R. v. Baltz[3], H. Kalt[4] and C. Klingshirn[4]

[1] *A.F.Ioffe Physical-Technical Institute, 194021, St.Petersburg, Russia*
[2] *Nuclear Physics Institute,188350, St.Petersburg, Russia*
[3] *Institute of Theory of Condensed Matter, University of Karlsruhe (TH), 76128 Karlsruhe, Germany*
[4] *Institute of Applied Physics, University of Karlsruhe (TH), 76128 Karlsruhe, Germany}*

Abstract. We have studied temperature dependence of the signals of optical orientation of excitons at resonant excitation of the ensemble of planar self-assembled CdSe/ZnSe quantum dots and show that spin memory in this system survives up to 100K. To describe the experimental results we apply the model that considers explicitly the effect of multi-phonon processes on the broadening of bright-exciton-phonon sublevels and demonstrate that calculated temperature dependences of spin relaxation rates are in good agreement with obtained experimental data.

Keywords: self-assembled quantum dots, excitons, optical orientation, optical alignment, temperature dependence.
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Nowadays a considerable attention is paid to comprehensive study of relaxation of an electron spin localized in a quantum dot (QD). Much less is known about the temperature dependence of the spin relaxation rates. Basically, the spin relaxation between discrete energy levels in QD's is hampered by the phonon bottleneck due to the energy conservation law. We demonstrate that multi-phonon processes transforming discrete levels into continuum bands relax the limitations connected with phonon bottleneck and allow to fit experimental findings.

The samples under study were grown by molecular-beam epitaxy of ultra narrow quantum wells (QWs) formed by insertion of CdSe with nominal thickness of 2.1 monolayers into ZnSe matrix [1] and revealed well developed planar quantum dots [2]. We have studied temperature dependences of the optical orientation of exciton spins and optical alignment of exciton dipole momenta at resonant excitation of the emitting states of excitons localized in self-assembled CdSe QDs.

Photoluminescense (PL) of localized excitons was excited by polarized light from Ar^+ laser (λ_{ex}=514.5nm). The laser beam was directed at small angle to the growth axis z, along which recombination emission was detected in back-scattering geometry. The linear polarization of the exciting light was achieved using a linear polarizer. Signal of PL passed through rotating half-wave plate followed by linear analyzer with fixed orientation and then was sent to spectrometer (for circular polarization measurements quarter-wave plates were placed after the linear polarizer in the exciting beam and before rotating half-wave plate for detection). For detection we used two-channel photon-counting system, which allowed parallel recording of the PL spectra in both polarizations I^{α}_{α} and I^{α}_{β} and calculation spectra of polarization degree $P^{\alpha}=(I^{\alpha}_{\alpha}-I^{\alpha}_{\beta})/(I^{\alpha}_{\alpha}+I^{\alpha}_{\beta})$. Here, symbols α and β stand for two components of polarization of exciting and/or detecting beams (x or y and σ_+ or σ_- for linear and circular polarizations, respectively), while superscript and subscript symbols indicate the polarization of excitation and detection, respectively. In order to exclude the polarization of PL which stems from the lateral anisotropy of the quantum dot the linear polarization degree has been detected for two orientations of polarizer x and y and spectra of optical alignment of excitons P_{lin} were obtained as $(P^x-P^y)/2$. Similar procedure with circular polarized excitations allowed to diminish the role of non-ideality of the used quarter-wave plates.

In Fig.1 the PL spectrum of the localized excitons and the spectra of polarization degrees P_{lin} and P_{circ} at resonant polarized excitation are shown. It was shown earlier [3, 4] that PL spectra of the samples under

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study are formed by the overlapping sub-bands corresponding to the recombination of the ground and metastable exciton states. As a consequence, at resonant excitation by polarized light both optical orientation and optical alignment signals can be detected, the relative intensity of which reflects the relative number of charged and neutral excitons.

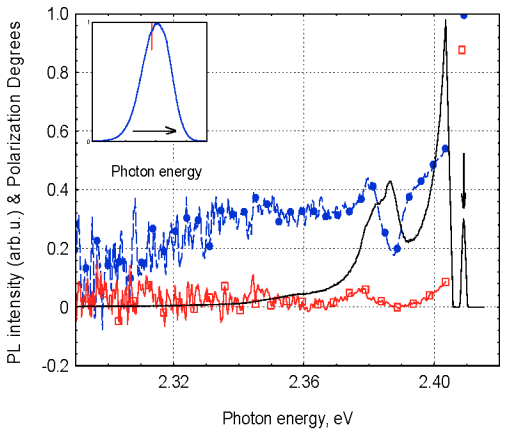

FIGURE 1. PL spectrum at resonant excitation at T=6K (black solid line) and spectra of optical orientation P_{circ} and optical alignment P_{lin} degrees of excitons (blue dashed line with closed symbols and red solid line with open symbols, respectively). The black vertical arrow indicates the spectral position of the excitation energy E=2.409 eV. Insert shows PL spectrum at above band gap excitation. Red vertical line in the insert indicates the photon energy used for resonant excitation.

In the two level approximation the temperature dependence of polarization degree of PL at polarized excitation can be obtained by solving the master equation for the populations of two spin states E_1 and E_2. In the simplest case of strictly polarized excitation the solution of this equation can be presented as

$$P(T) = \frac{1+\tau[w_{2,1}(T) - w_{1,2}(T)]}{1+\tau[w_{2,1}(T) + w_{1,2}(T)]}. \qquad (1)$$

Here, τ is the PL decay time, $w_{1,2}(T)$ and $w_{2,1}(T)$ are transition rates between two spin sublevels, which are connected, as usual, by the ratio $w_{1,2}(T)/w_{2,1}(T)=\exp(E_{12}/kT)$. For degenerate states $E_1=E_2$ the rates are equal $w_{1,2}(T)=w_{2,1}(T)$ and tend to zero at $T\to 0$. These relations and Eq.(1) allow to connect the product $\tau\, w_{1,2}(T)$ and $P(T)$.

In Fig.2 the temperature dependence of the products $\tau\, w_{1,2}(T)$ calculated from the experimental values of polarization degrees P_{lin} and P_{circ} are shown.

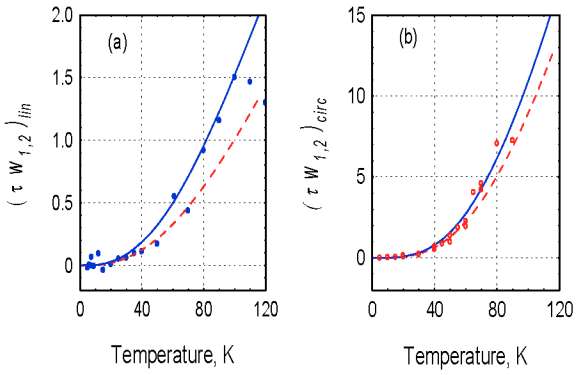

FIGURE 2. Temperature dependences of the momentum relaxation rate (*a*) and the spin relaxation rate (*b*). Symbols are the experimental data obtained from polarization degrees P_{lin} and P_{circ} measured at photon energy of 2.405 eV in spectra shown in Fig.1. For calculated dependences (dashed and solid curves) the product $\tau\, w_{1,2}$ at T=5K was taken to be equal to (2.5 or 5) 10^{-4} (panel *a*) and (10 or 12) 10^{-4} (panel *b*), respectively.

We have performed the fitting of experimental data and have shown that the temperature dependence of transition rates $w_{1,2}(T)$ and $w_{2,1}(T)$ can be obtained by taking into account the broadening of electronic levels due to multi-phonon processes. These processes are not able by themselves to provide the spin-flip. However, they modify the spin relaxation indirectly leading to the transformation of the discrete exciton spin sub-levels into continuum bands.

The best fit of the experimental data presented in Fig.2 was achieved under assumption that the levels 1 and 2 are splitted for the states which demonstrate optical alignment signal (panel *a*) and are degenerated for those which give optical orientation signal (panel *b*). The estimated values of the parameters $\tau\, w_{1,2}$ which are of order 10^{-4} – 10^{-3} at T=5K, mean that at low temperature spins of the emission states do not relax during the radiative lifetime. This conclusion is in reasonable agreement with presently available experimental findings (see, for example [5]).

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