

Nanosecond Spin Coherence Time of Nonradiative Excitons in GaAs/AlGaAs Quantum Wells

A. V. Trifonov,^{1*} E. S. Khramtsov,¹ K. V. Kavokin,¹ I. V. Ignatiev,¹ A. V. Kavokin,^{2,1}
Y. P. Efimov,³ S. A. Eliseev,³ P. Yu. Shapochkin,³ and M. Bayer^{4,5}

¹*Spin Optics Laboratory, St. Petersburg State University, St. Petersburg 198504, Russia*

²*Westlake University, 18 Shilongshan Road, Hangzhou 310024, Zhejiang Province, China and Institute of Natural Sciences, Westlake Institute for Advanced Study, 18 Shilongshan Road, Hangzhou 310024, Zhejiang Province, China*

³*Resource Center “Nanophotonics,” St. Petersburg State University, St. Petersburg 198504, Russia*

⁴*Experimentelle Physik 2, Technische Universität Dortmund, D-44221 Dortmund, Germany*

⁵*A. F. Ioffe Physical-Technical Institute, Russian Academy of Sciences, St. Petersburg 194021, Russia*



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We report on the experimental evidence for a nanosecond timescale spin memory based on nonradiative excitons with large in-plane wave vector. The effect manifests itself in magnetic-field-induced oscillations of the energy of the optically active (radiative) excitons. The oscillations detected by a spectrally resolved pump-probe technique applied to a GaAs/AlGaAs quantum well structure in a transverse magnetic field persist over a timescale, which is orders of magnitude longer than the characteristic decoherence time in the system. The effect is attributed to the spin-dependent electron-electron exchange interaction of the optically active and inactive excitons. The spin relaxation time of the electrons belonging to nonradiative excitons appears to be much longer than the hole spin relaxation time.

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Excitons are crystal quasiparticles that can be generated by light and that may eventually recombine emitting light [1,2]. As such, they are promising for storing the optically encoded information and keeping memory of the intensity, phase, and polarization of light. Applications of excitons for optical storage are limited by their short radiative lifetime (typically, on the order of 10–100 ps) and even shorter coherence time (on the order of a few picoseconds). Nonradiative, also referred to as dark or optically inactive, excitons that are decoupled from light due to the specific selection rules for optical transitions are widely discussed as the most promising exciton memory agents [3–7]. They possess lifetimes on a nanosecond or longer scale and affect many processes in optically excited quantum wells (QWs) [8–13], quantum dots [14,15], microcavities [16–19], and 2D materials [20,21]. On the other hand, a rapid thermalization of the reservoir of nonradiative excitons usually leads to the loss of coherence on a few-picosecond scale.

The capacity of a reservoir of nonradiative excitons to serve as an optical polarization or spin storage is yet to be fully revealed. Here we study the spin memory effects in the excitonic system by means of time-resolved magneto-optical spectroscopy. In our experiment, the reservoir consists of excitons with large in-plane wave vectors strongly exceeding the wave vector of light [see Fig. 1(a)] so that the k -vector selection rules do not allow these excitons to absorb or emit light. Nevertheless, dark excitons can be optically addressed via their interaction with the optically active (bright) excitons [13,22].

We have developed an experimental approach allowing for direct access to the spin polarization of reservoir excitons. We observe a robust exciton spin polarization lasting several nanoseconds. It manifests itself in magnetic-field-induced oscillations of the optically active exciton energy due to their exchange interaction with the reservoir of spin-polarized nonradiative excitons.

A high-quality heterostructure with a 14-nm GaAs/Al_{0.03}Ga_{0.97}As quantum well was experimentally studied. The structure was grown by molecular beam epitaxy on an n -doped GaAs substrate. Because of the small content of Al in the barrier layers, their height is relatively small, about 25 meV for electrons and 12 meV for holes. Figure 1(b) shows a reflectance spectrum of the sample in the spectral range of the exciton resonances. The main features observed in the spectrum can be ascribed to optical transitions to the quantum-confined heavy-hole (Xhh) and light-hole (Xlh) exciton states in the QW. The very small spectral widths of the exciton resonances confirm the ultrahigh quality of the structure. These resonances can be precisely modeled by a phenomenological theory described in Refs. [13,23,24].

Within this model, the amplitude reflection coefficient of light from a QW exciton resonance can be written in the form

$$r_X = \frac{i\Gamma_0}{\tilde{\omega}_0 - \omega - i(\Gamma_{\text{NR}} + \Gamma_0)}. \quad (1)$$

Here, the parameter Γ_0 describes the radiative decay rate of the exciton state, Γ_{NR} is the rate of nonradiative relaxation

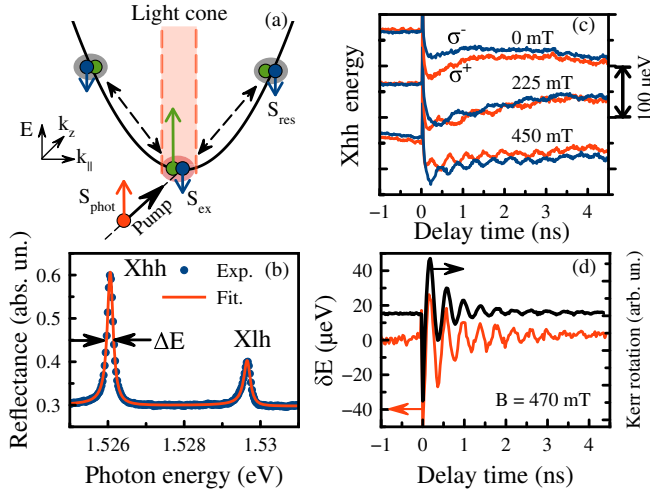


FIG. 1. (a) A scheme illustrating optically active and inactive excitons and the exchange interaction of spins of the radiative excitons within the light cone with the spin-polarized excitons in the nonradiative reservoir. (b) Reflectivity spectrum of the 14-nm GaAs/AlGaAs QW (blue dots). The red dashed line shows the fit of the exciton resonances by Eqs. (1) and (2). $\Delta E \approx 180 \mu\text{eV}$. (c) Time evolution of the Xhh exciton energy E_{Xhh} in the transverse magnetic field in the σ^+ (red curves) and σ^- (blue curves) polarizations measured for σ^+ -polarized excitation. The magnetic field magnitudes are indicated near each pair of curves. The curves are shifted vertically for clarity. (d) Comparison of the dynamics of the Kerr rotation signal (black curve) and of the exciton energy splitting (red curve). The sample temperature is $T = 6 \text{ K}$.

from this state, and $\tilde{\omega}_0$ is the frequency of the exciton transition. These three quantities are considered to be fitting parameters of the model. The reflectivity spectrum of the structure is then given by

$$R = \left| \frac{r_s + r_X e^{i\phi}}{1 + r_s r_X e^{i\phi}} \right|^2, \quad (2)$$

where r_s is the amplitude reflection coefficient of the sample surface, and ϕ is the phase shift of the light traveling from the sample surface to the QW and back.

The good agreement of the experimental and modeled spectra shown in Fig. 1(b) indicates that no significant inhomogeneous (Gaussian-like) broadening is present in this structure. This allows us to obtain reliable values of all the fitting parameters. For the Xhh resonance shown in Fig. 1(b), the fitting parameters are $\hbar\Gamma_0 = 30 \pm 1 \mu\text{eV}$, $\hbar\Gamma_{\text{NR}} = 45 \pm 2 \mu\text{eV}$, and $E_{\text{Xhh}} = \hbar\tilde{\omega}_0 = 1526.061 \pm 0.002 \text{ meV}$. One can see that the energy of the exciton states E_{Xhh} can be obtained with a high accuracy of about $2 \mu\text{eV}$. This opens the way to highly sensitive experiments for the study of interaction of photocreated excitons with other quasi-particles in the structure.

We have developed a spectrally resolved pump-probe experimental technique with the circularly polarized 2-ps

pump-pulse exciting the structure at some spectral point while the spectrally broad 100-fs probe pulse is used to detect the reflection spectrum at each delay between the pump and probe pulses. The linearly polarized probe beam reflected from the sample is split into two circularly polarized components. Spectra of both components are simultaneously measured by an imaging spectrometer equipped by a CCD detector (see details in the Supplemental Material [25]). In this way, two reflectance spectra in both circular polarizations are detected. The analysis of the spectra measured at different delays using Eqs. (1) and (2) allows us to obtain the dynamics of the essential excitonic parameters.

Figure 1(c) shows the dependence of the energy of the Xhh exciton resonance on the delay between the pump and probe pulses. A small magnetic field is applied to the structure perpendicular to the growth axis (Voigt geometry). We see that the exciton energy undergoes an instantaneous jump and rapid decay at small delays followed by a smooth change when no magnetic field is applied. There is a noticeable difference between the curves measured in the σ^+ and σ^- polarizations. This difference is inverted when the excitation helicity is changed. It disappears under the linearly polarized pumping. When the magnetic field is applied, the tail of the exciton energy dynamics becomes oscillating. The oscillations are opposite in sign for the σ^+ and σ^- polarizations of detection.

To clarify the origin of these oscillations, we have compared the oscillations in energy splitting $\delta E = E_{\text{Xhh}}^{\sigma^+} - E_{\text{Xhh}}^{\sigma^-}$ with the oscillating Kerr rotation signal measured in the same experimental conditions; see Fig. 1(d). One can see that both signals look very similar. However, the oscillating entities are different in these two measurements. In particular, the Kerr signal is proportional to the rotation angle of the light polarization plane oscillating due to the spin precession about the magnetic field [29–31]. Therefore, we may conclude that the energy splitting δE may also be a result of the exchange interaction of the radiative excitons with some reservoir of polarized spins precessing in the external magnetic field.

To reveal the physical nature of the spin reservoir that is responsible for the oscillations, we have measured the oscillating energy splitting δE exciting into the heavy-hole and light-hole resonances as shown in Fig. 2. One can see that the phase of the δE oscillations is opposite in the two experiments. It is well known [23] that the optically active heavy-hole and light-hole excitons created by light with the same helicity of polarization involve electrons with opposite spins. Therefore, the opposite phase is a signature of the difference in the selection rules of the Xhh and Xlh exciton transitions.

The dependence of the oscillation frequency on the applied external magnetic field is shown in the inset of Fig. 2. It is clearly seen that the frequency dependence on the magnetic field is linear. From the slope of this line we

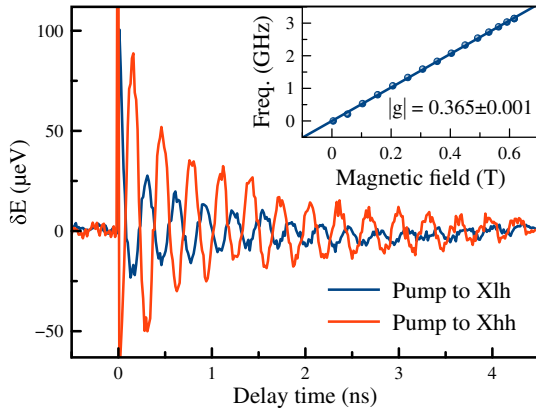


FIG. 2. The time delay dependence of the energy difference $\delta E = E_{\text{Xhh}}^{\sigma^+} - E_{\text{Xhh}}^{\sigma^-}$ of the Xhh exciton energies measured at the σ^+ and σ^- circular polarizations under the σ^+ excitation into the Xhh (red curve) and Xlh (blue curve) exciton resonances. Excitation density $P = 50 \text{ W/cm}^2$; magnetic field strength $B = 0.6 \text{ T}$; sample temperature $T = 5.6 \text{ K}$. Inset shows the oscillation frequency vs magnetic field strength extracted from the experiment (points) and the linear fit by function $\Omega = (|g|\mu_B B)/\hbar$.

can determine the g factor $|g| = 0.365 \pm 0.001$, which nearly coincides with the known value of the electron g factor in QWs [32] of such widths.

The magnitude of the g factor and the inversion in the phase of the oscillations upon excitation of the Xlh and Xhh exciton resonances are two key experimental findings that point to the mechanism of the oscillations. We may conclude that the oscillating energy splitting of the exciton levels is due to the exchange interaction of excitons with the long-lived electrons whose spins precess about the external magnetic field. These can be free resident electrons, photocreated free electrons, or electrons in the excitons of a long-lived nonradiative reservoir.

To identify the origin of these electrons, we have performed a theoretical estimate of the exchange interaction between the bright excitons and the long-lived electrons as well as of the electron density n_e required to obtain the observed energy shifts. The spin Hamiltonian of the exchange interaction reads [33,34]

$$\hat{H}_S = \Delta_0 \hat{i}_z \hat{s}_z + J_{ee} n_e (\hat{s} \cdot \langle \vec{S} \rangle). \quad (3)$$

Here, \hat{s}_z and \hat{i}_z are the projections of the electron and hole spins belonging to the bright exciton on the growth axis z , Δ_0 is electron-hole exchange interaction energy, $\langle \vec{S} \rangle$ is the average spin in the reservoir, and J_{ee} is the exchange interaction constant. The explicit expression for J_{ee} and its numerical calculation are given in the Supplemental Material [25]. We have also estimated the constant Δ_0 for the structure under study. The obtained value [25] $\Delta_0 < 20 \mu\text{eV}$ is small compared to the observed exciton energy splitting caused by the interaction of the exciton

spin with the reservoir of electron spins. We should also note that the exchange interaction of a hole in the bright exciton with the reservoir electrons is much weaker and can be neglected [33].

The diagonalization of the Hamiltonian (3) gives rise to four eigenstates. When the average spin $\langle S \rangle$ is directed along the growth axis z , the bright and dark exciton states are not mixed and optical transitions are allowed only to the bright exciton states. When the reservoir spin is rotated perpendicular to the z axis by the applied magnetic field, the bright and dark exciton states are mixed and all four exciton transitions are allowed. All the splittings, however, are much smaller than the exciton line broadening $\hbar(\Gamma_0 + \Gamma_{\text{NR}})$. Therefore, the effect of exchange interaction is observed as a shift of a single exciton resonance when the reservoir spin is rotated. The difference in the energy positions of the single resonance seen in the σ^+ and σ^- polarizations is described by [see Eq. (8) in the Supplemental Material [25]]

$$\delta E = J_{ee} n_e \langle S_z \rangle, \quad (4)$$

where $\langle S_z \rangle$ is the z projection of the reservoir spin.

Let us first consider the exchange interaction of bright excitons with a reservoir of free electrons. The corresponding interaction constant is [25] $J_{ee}^{xe} = 18 \pm 2 \mu\text{eV} \times \mu\text{m}^2$. If the reservoir electrons are totally polarized, that is, $\langle S_z \rangle = 1/2$, we obtain from Eq. (4) the minimum areal electron density, $n_e = 1.1 \times 10^9 \text{ cm}^{-2}$ (for $\delta E = 100 \mu\text{eV}$; see Fig. 2). If the reservoir is composed by resident electrons, their average polarization $\langle S_z \rangle \ll 1/2$ and the required areal density $n_e \gg 10^9 \text{ cm}^{-2}$. The electrons with such an areal density should give rise to the trion (negatively charged exciton) peaks in the optical spectra [35,36]. We, however, did not observe such features in both the photoluminescence and reflectance spectra. We should note that the trion peaks are observed in intentionally n -doped structures [36], but our structure is undoped. But our structure is undoped. Therefore, we may assume that the resident electrons cannot be responsible for the observed effect.

The free electrons in the reservoir could be, in principle, created by optical pumping. However, we have used resonant pumping into the lowest exciton state, which makes this scenario unlikely. Indeed, the electrons are coupled with holes in the photocreated excitons. The Coulomb energy of the coupling $R_X \approx 7 \text{ meV}$ in the QW under study [37], which is 1 order of magnitude larger than the thermal energy, $kT \approx 0.5 \text{ meV}$. Therefore, the photocreation of free electrons is supposed to be a highly inefficient process in our case.

Eventually we come to the conclusion that the electrons that belong to the reservoir excitons are responsible for the observed energy shifts. We should note here that there are excitons with total angular momentum $J = 2$ which are

also nonradiative even when their k vector is within the light cone. These excitons, however, cannot contribute to the observed phenomenon because the spin rotation of the electron comprising the exciton converts it into the bright one. After the conversion, such excitons immediately disappear via the electron-hole recombination. Therefore, only the excitons with large in-plane k vector comprise the nonradiative reservoir responsible for the discussed effect. The interaction of the bright excitons with those in the nonradiative reservoir via the electron-electron exchange is characterized by the constant [25] $J_{ee}^{xx} = 11.4 \pm 0.8 \mu\text{eV} \times \mu\text{m}^2$. Therefore, the required areal density of the reservoir excitons with totally polarized electron spins, $n_X \approx 1.8 \times 10^9 \text{ cm}^{-2}$. Such a density can be easily created in our experiments. Indeed, the number of absorbed photons per excitation pulse n_{phot} is calculated using the well-known expressions for the absorption coefficient [23], $\eta = (2\Gamma_0\Gamma_{\text{NR}})/[(\tilde{\omega}_0 - \omega)^2 + (\Gamma_0 + \Gamma_{\text{NR}})^2]$. Taking into account the spectral overlap of the pump laser pulses and the Xhh resonance, we obtain for the experimental conditions of Fig. 2: $n_{\text{phot}} \approx 2 \times 10^{11}$ photons per pulse per cm^2 . Only a small fraction of bright excitons created by the absorbed photons can be scattered from the light cone into the nonradiative reservoir $f \sim \tau_X/\tau_{\text{AC}} \sim 0.1$, where $\tau_X \approx 10$ ps is the characteristic time of the exciton radiative recombination, and $\tau_{\text{AC}} \sim 100$ ps is the exciton-phonon scattering time [13]. Finally the areal density of excitons in the nonradiative reservoir $n_X \sim 2 \times 10^{10} \text{ cm}^{-2}$ is still 10 times larger than the required exciton density. Taking into account the possible loss of the electron spin polarization during the exciton scattering, we obtain an energy shift comparable with the experimentally observed one.

To further support this conclusion, we have measured the dynamics of the exciton energy splitting $\delta E(t)$ at different temperatures and compared it with the dynamics of the nonradiative broadening of the Xhh resonance $\hbar\delta\Gamma_{\text{NR}}(t) = \hbar\Gamma_{\text{NR}}(t) - \hbar\Gamma_{\text{NR}}(t_m)$, where t_m is a small negative delay time. A representative set of these data is shown in Fig. 3. A close similarity is observed in the dynamics of both the energy oscillations and the broadening. A phenomenological fit of the dynamics of the exciton energy splitting by $\delta E(t) = A_{\delta E} \exp(-t/\tau_{\delta E}) \cos(\omega t + \varphi)$ and of the broadening by $\hbar\delta\Gamma_{\text{NR}}(t) = A_{\text{NR}} \exp(-t/\tau_{\text{NR}})$ allows one to obtain the characteristic decay times $\tau_{\delta E}$ and τ_{NR} of these processes. Their dependence on the sample temperature is shown in the inset of Fig. 3(b).

The obtained data clearly demonstrate that $\delta E(t)$ decays in time nearly with the same rate as the nonradiative broadening $\hbar\delta\Gamma_{\text{NR}}(t)$. Having in mind that the decay of the broadening is governed by the depopulation of the exciton reservoir [13], we conclude that the decay of the oscillations is also mainly related to the exciton depopulation. The observed small difference in the decay times could be, in principle, explained by electron spin relaxation

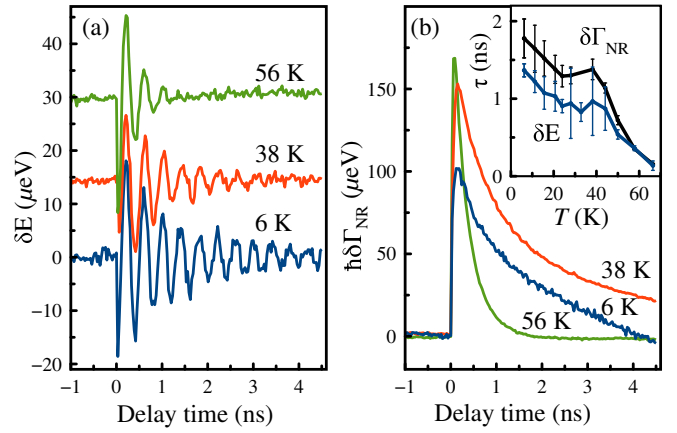


FIG. 3. The pump-probe time delay dependence of the exciton energy splitting δE (a) and of the nonradiative broadening $\hbar\delta\Gamma_{\text{NR}}(t)$ (b) at different temperatures. The curves in (a) are shifted for clarity; in (b) only the single pump-pulse-induced broadening is shown. Inset in (b) shows the temperature dependence of the characteristic decay times of the oscillations and of the nonradiative broadening. The excitation density is $P = 15 \text{ W/cm}^2$.

or dephasing. The effective time of these processes in high-quality QWs is very large [38,39].

The analysis above supports our conclusion that the observed behavior of the oscillating signal is caused by polarized electrons belonging to excitons in the nonradiative reservoir. The spins of these electrons are coupled with those of holes via the exchange interaction. The magnitude of this interaction $\Delta_0 \sim 10\text{--}20 \mu\text{eV}$ is larger than the Zeeman splittings observed in our experiments, $\delta E_Z = g\mu_B B \approx 11 \mu\text{eV}$ at $B = 0.5 \text{ T}$. In the presence of the exchange interaction, the magnetic field dependence of the oscillation frequency should be nonlinear [40], which would contradict the experimental observation; see the inset in Fig. 2.

We assume that the exchange interaction is effectively switched off in the nonradiative excitons [31]. During their relatively long lifetime, the hole spin can be lost due to the spin-orbit interaction because the thermal energy $kT \gg \Delta_0$. In fact, due to the interaction with the phonon bath, the hole spin orientation can be changed many times during one period of the electron spin precession in the external magnetic field. A characteristic time of the hole spin relaxation is of the order of several tens of ps [31,40]. Even when the hole spin polarization is lost, there are fluctuations of electron-hole exchange interaction which might destroy the electron spin polarization [41]. However, in our case the exchange interaction is much weaker ($\Delta_0 < 20 \mu\text{eV}$) than the exciton-collision-induced broadening ($\Gamma_{\text{NR}} > 100 \mu\text{eV}$). During the period of electron spin precession in the fluctuating field, the exciton is scattered many times by other excitons so that the fluctuating field is effectively averaged. This process is similar to the well-known motional narrowing [42].

In conclusion, we have directly observed the exciton energy shift caused by the exchange interaction of the photocreated excitons with those in the nonradiative reservoir in high-quality QWs. The shift oscillates in time when the transverse magnetic field is applied to the structure. The oscillations decay on a nanosecond time-scale, which is orders of magnitude longer than the exciton radiative lifetime. We attribute the oscillations to the spin precession of electrons belonging to the nonradiative excitons with large in-plane k vectors. Our experiment has clearly shown that the electron-hole exchange interaction in the nonradiative excitons is suppressed due to depolarization of holes during the large lifetime of these excitons. We have theoretically modeled the exchange interaction of electrons in the photocreated excitons with those in the nonradiative excitons and obtained relevant interaction constants. Our finding opens up the way to create an optically controllable spin memory system based on the long-lived reservoir of nonradiative excitons.

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*Corresponding author.

a.trifonov@spbu.ru

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