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On the Suppression of Electron-Hole Exchange Interaction in a Reservoir of Nonradiative Excitons

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Abstract—Mechanisms of the suppression of the electron-hole exchange interaction in nonradiative excitons with a large in-plane wave vector in high-quality heterostructures with quantum wells are analyzed theoretically. It is shown that the dominant suppression mechanism is exciton-exciton scattering accompanied by the mutual spin flips of like carriers (either two electrons or two holes), comprising the excitons. As a result, the electron spin polarization in nonradiative excitons may be retained for a long time. The analysis of experimental data shows that this relaxation time can exceed one nanosecond. This long-term and optically controllable spin memory in an exciton reservoir may be of interest for future information technologies.

Keywords: exciton, exchange interaction, quantum well

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1. INTRODUCTION

It was demonstrated in [1] that, in high-quality heterostructures with quantum wells based on direct-gap semiconductors like GaAs, the accumulation of a reservoir of nonradiative excitons with a large in-plane wave vector exceeding the wave vector of light in the quantum-well material is possible under optical excitation. The exciton density in this reservoir may exceed by several orders of magnitude that of radiative excitons with wave vectors within the light cone. The large difference in the densities of radiative and nonradiative excitons is related to a difference in their lifetimes. The radiative lifetime of excitons with small wave vector is very short, ~10 ps. At the same time, nonradiative excitons in high-quality heterostructures may live tens of nanoseconds [1].

Although nonradiative excitons do not interact with light, their optical probing is possible via the effects they cause on radiative excitons. In particular, the scattering of radiative and nonradiative excitons causes a broadening and an energy shift of the exciton resonances in reflectance spectra. Moreover, our recent study has shown [2] that the shift of exciton states is sensitive to the excitation polarization. Under the excitation of a sample by circularly polarized radiation, the shift of the exciton resonances is different in the co- and cross-circular polarizations of detection. This observation indicates the conservation of spin polarization in a reservoir of nonradiative excitons.

The dynamics of the spin polarization of an exciton reservoir was studied in [2] by the pump-probe method with both spectral and polarization resolution in a magnetic field applied across the heterostructure-growth axis (Voigt geometry). A high-quality heterostructure with a 14-nm GaAs quantum well grown by molecular-beam epitaxy was studied. An example of the reflectance spectrum of this heterostructure in the region of optical transitions to the quantum-confined states of heavy-hole (Xhh) and light-hole (Xlh) excitons is shown in Fig. 1. The observed exciton resonances are well described in the framework of the standard reflectance model [3–6]. This allows one to determine with high accuracy the spectral position and the broadening of the exciton resonances. Under excitation by pump pulses, the exciton resonances are broadened and slightly shifted, which allows one to study the dynamics of the shift and the broadening by detecting the reflectance spectra of the probe pulses delayed in time relative to the pump pulses. Examples of such dependences for the Xhh resonance are shown in Fig. 2. As seen from the figure, a sharp red shift of the exciton resonance and its considerable broadening are observed when the pump pulse arrives. Then, a relatively slow decrease in the shift and in the excess broadening is observed for several nanoseconds. This behavior is explained by the creation of nonradiative excitons by pump pulses followed by gradual depopulation of the reservoir of these excitons [2].

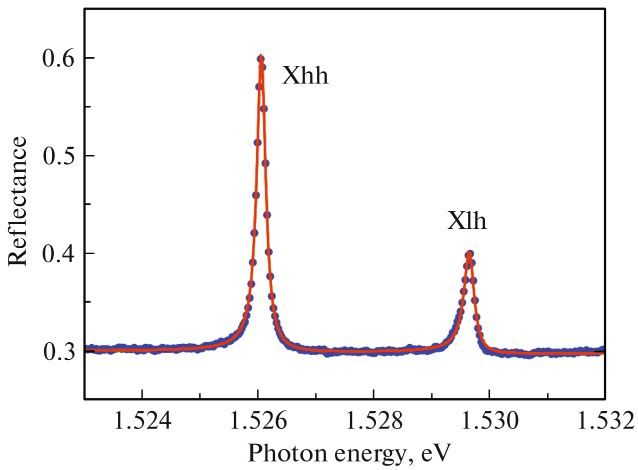

Fig. 1. Reflectance spectrum of a sample with a 14-nm quantum well (points) and its fit in the framework of the standard model (solid line). Resonances of the reflectance corresponding to optical transitions with the creation of heavy-hole (Xhh) and light-hole (Xlh) excitons are clearly seen. The sample temperature $T = 6$ K.

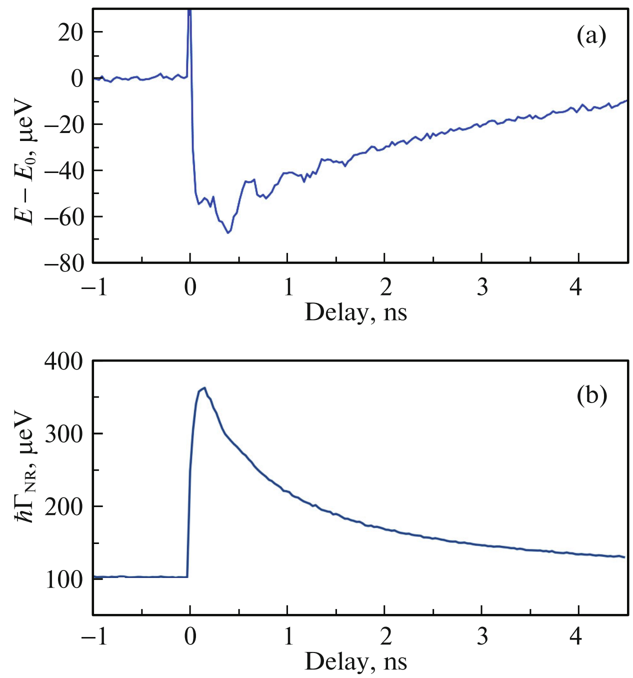

Fig. 2. Energy shift of a heavy-hole exciton (a) and broadening of the Xhh resonance (b) as functions of the delay between the pump and probe pulses. Magnetic field $B_x =$ 0.5 T. The oscillations of the exciton-energy position are clearly seen (a). They are absent in the dynamics of the broadening (b).

When a transverse magnetic field is applied and the heterostructure is excited by circularly polarized pump pulses, a component of the exciton resonance shift which oscillates in time is observed in the reflectance spectrum (see Fig. 2a). The phase of the oscillating shift is opposite in the co- and cross-polarizations relative to the polarization of the pump beam. An example of the difference between these shifts is shown in Fig. 3. It is experimentally found that the oscillation frequency linearly depends on the magnetic field applied, $\hbar\omega = g\mu_B B_x$. The value of the factor g agrees well with the free electron g factor for a quantum well of this width, known from publication [7]. It would seem that this fact indicates the presence of a reservoir of spin-polarized free electrons in the heterostructure under study. The exchange interaction of radiative excitons with polarized electrons should result in an energy shift of the exciton peak. Moreover, the sign of the shift should depend on the mutual orientation of the exciton and electron spins. The electron-spin precession in an external magnetic field with the Larmor frequency should, therefore, lead to an oscillating exciton shift.

However, there are no free electrons in the quantum well under study, as was verified in [2] by additional experiments. There are electrons coupled with holes in nonradiative excitons. The spins of these electrons experience an exchange interaction with the spins of holes, which should lead, in general, to a nonlinear frequency dependence of the oscillations [8]. It is claimed in [2], however, that the fast relaxation of hole spins in an ensemble of nonradiative excitons promotes electron-spin conservation. Due to relaxation, the electron spins do not experience the exchange interaction with holes at the time scale in

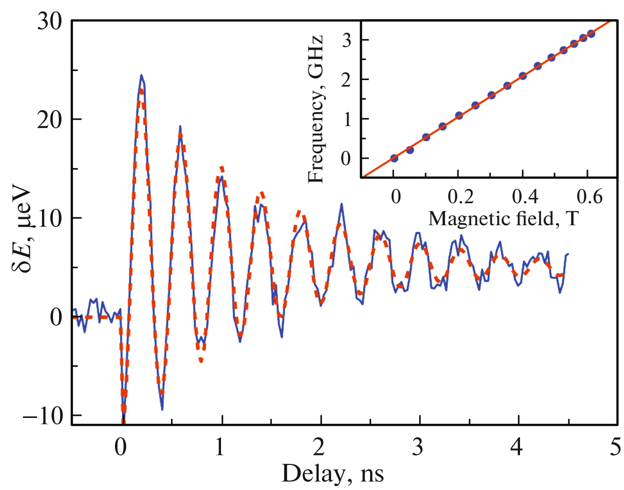

Fig. 3. Dynamics of the exciton-shift difference measured upon excitation by circularly polarized pump pulses to the heavy-hole exciton transition and signal detection in co- and cross-circular polarizations (noisy curve). Magnetic field $B_x = 0.5$ T. Dashed curve shows the fit by function: $f(\tau) = \exp\left(-\frac{\tau}{T_2}\right)\cos(\omega\tau) + \text{const}$, with the parameters: $T_2 = 1.36$ ns, $\omega/(2\pi) = 2.5$ GHz. The inset shows the oscillation frequency as a function of magnetic field B_x (points). Solid line is the approximation by linear function, $\delta E = g\mu_B B$, where μ_B is the Bohr magneton and $g =$ 0.365.

tens and hundreds of picoseconds. They freely undergo precession in an external magnetic field, which is an order of magnitude smaller than the effective field of the exchange interaction. In the current paper, we present detailed substantiation of this statement.

2. MODEL

Let us consider the exchange interaction of an electron and a hole in an exciton. The magnitude of this interaction is characterized by the energy splitting δ_0 between exciton states with the total spin ± 1 (which are bright excitons if their wave vector is inside the light cone) and the exciton states with spin ± 2 (the dark excitons at any wave vector). Estimates show [2] that $\delta_0 \approx 20$ μeV, which is much smaller than the thermal energy, $k_B T \approx 430$ μeV, at the sample temperature $T = 5$ K. Hence, hole polarization in nonradiative excitons can relax rather quickly [9]. However, even a nonpolarized hole can noticeably affect the electron-spin dynamics in an exciton because the exchange interaction of an electron with a hole exceeds its interaction with an external magnetic field of a strength relevant to this study. Nonpolarized holes create a fluctuating effective magnetic field, which can cause dephasing of the electron-spin precession in the ensemble. Let us consider this process in more detail.

The spin of a nonpolarized hole can be $J = \pm 3/2$ with equal probabilities. Although the average value of the hole spin $\langle \vec{\mathbf{J}} \rangle = 0$, averaging of the spin squared gives rise to a non-zero value, $\langle J_z^2 \rangle = 9/4$. Therefore the electron is affected by the fluctuating effective field of the hole, which is parallel to the z axis and has root-mean-square (rms) amplitude B_h^{eff} defined by the relation $\delta_0 = g_e \mu_B B_h^{\text{eff}}$, where g_e is the electron g factor and μ_B is the Bohr magneton. The correlation time τ_h, during which the electron spin "feels" the unchanged field B_h^{eff}, is governed by two processes. First, the hole spin is efficiently coupled with lattice vibrations via its orbital component and, therefore, changes its orientation rather quickly, $\tau_{h1} \sim 100$ ps [9]. Second, exciton-exciton collisions in the reservoir cause mutual spin flips of like carriers (either two electrons or two holes) in the excitons. As a result, effective averaging of the exchange field of holes occurs. This is a well-known effect frequently called "motional narrowing", which is observed in various spin processes [10, 11]. The characteristic time of this process, τ_{h2}, can be estimated in our case from the broadening of the exciton resonance after pulsed excitation (see Fig. 2b): $\hbar\Gamma_{NR} \approx$ 0.1 meV. Correspondingly, $\tau_{h2} = \dfrac{1}{(2\Gamma_{NR})} \approx 3$ ps. These estimates show that motional narrowing is the dominant process for suppression of the effective field of hole-spin fluctuations: $\tau_h = (1/\tau_{h1} + 1/\tau_{h2})^{-1} \approx \tau_{h2} \sim 3$ ps.

The effective field of hole-spin fluctuations is directed along the z axis therefore it does not cause relaxation of the electron-spin z component. Relaxation of the S_x and S_y components in the mode of motional narrowing is described by the well-known expression [11]: $1/\tau_x = 1/\tau_y = \omega_h^2 \tau_h$, where $\omega_h = \delta_0/\hbar = 2\pi/T_{\text{exch}}$. This expression can be explained as follows. During the correlation time τ_h, the electron spin rotates around the z axis by the angle

$$\varphi = 2\pi \frac{\tau_h}{T_{\text{exch}}} \ll 1, \tag{1}$$

where $T_{\text{exch}} = h/\delta_0 \approx 200$ ps. The component S_z of the electron spin does not change during such precession. The component S_y, which appears as a result of regular electron-spin precession in an external magnetic field B_x, decreases in the time τ_h by

$$\delta S_y = S_y \cos\varphi - S_y \approx -S_y \frac{\varphi^2}{2}. \tag{2}$$

We assume that the directions of electron-spin rotation in the field of hole-spin fluctuations are not correlated on a time scale exceeding τ_h. Correspondingly, the relaxation rate of this spin component is

$$\dot{S}_y = \frac{\delta S_y}{\tau_h} = -\frac{1}{2} S_y \tau_h \left(\frac{2\pi}{T_{\text{exch}}}\right)^2 = -\frac{S_y}{\tau_y}, \tag{3}$$

where $\tau_y = \dfrac{2}{\tau_h}\left(\dfrac{T_{\text{exch}}}{2\pi}\right)^2 \approx 700$ ps is the characteristic relaxation time. A similar expression is also valid for the x component of the electron spin.

Dynamic equations for regular electron-spin precession in an external magnetic field taking into account relaxation of the S_y component are easily derived:

$$\dot{S}_z = -\omega S_y,$$

$$\dot{S}_y = \omega S_z - \frac{S_y}{\tau_y}. \tag{4}$$

Here the precession frequency is determined by the equation $\hbar\omega = g_e \mu_B B_x$. The removal of S_y from these equations gives rise to a second-order differential equation for S_z:

$$\ddot{S}_z + \frac{1}{\tau_y}\dot{S}_z + \omega^2 S_z = 0. \tag{5}$$

A solution of Eq. (5) is easily obtained by the substitution of a trial solution in the form: $S_z(t) = S_z(0)\exp(-\lambda t)$. This substitution leads to a quadratic equation for parameter λ,

$$\lambda^2 - \frac{\lambda}{\tau_y} + \omega^2 = 0 \tag{6}$$

with the roots:

$$\lambda_{\pm} = \frac{1}{2}[\tau_y^{-1} \pm \sqrt{\tau_y^{-2} - 4\omega^2}]. \tag{7}$$

To provide oscillating $S_z(t)$, as observed experimentally, the roots $\lambda_{\pm}$ should contain an imaginary part. From this requirement we obtain the condition:

$$\omega > \frac{1}{2\tau_y} = \frac{1}{4}\tau_h\left(\frac{2\pi}{T_{\rm exch}}\right)^2. \tag{8}$$

The condition (8) imposes a restriction on the frequency of Larmor precession in an external magnetic field, for the oscillation behavior of the spin polarization to be observed:

$$f_L = \frac{\omega}{2\pi} > \frac{1}{2\pi \cdot 2\tau_y} \approx 0.1\ \text{GHz}. \tag{9}$$

This is a relatively weak restriction compared to the frequencies observed experimentally (see Fig. 3). The corresponding restriction for an external magnetic field is:

$$B_x = \frac{\hbar\omega}{g_e\mu_{\rm B}} > 0.02\,\text{T}. \tag{10}$$

We note that the minimal value of the external magnetic field is much smaller than the effective field of electron-hole exchange interaction in the exciton:

$$B_h^{\rm eff} = \frac{\delta_0}{g_e\mu_{\rm B}} \approx 1\ \text{T}. \tag{11}$$

If the hole spin were retained for a long time, the electron spin would precess in the total field, $\mathbf{B}_{\rm tot} = \mathbf{B}_x + \mathbf{B}_h^{\rm eff}$, with the frequency [8, 12]:

$$\Omega = \sqrt{\omega^2 + (\delta_0/\hbar)^2} \approx (\delta_0/\hbar)\left[1 + \frac{1}{2}\left(\frac{\omega}{\delta_0/\hbar}\right)^2\right]. \tag{12}$$

Hence, the precession frequency would nonlinearly depend on the external magnetic field and it would be limited from below by the frequency of precession in the effective exchange field, $\Omega_{\min} = (\delta_0/\hbar) = 2\pi/T_{\rm exch} \approx 2\pi \cdot 5$ GHz. The frequencies observed in the experiment are below this limit (see Fig. 3), indicating efficient depolarization of the hole spin and efficient averaging of its fluctuations.

We note that, for the case $\omega \gg 1/(2\tau_y)$, the solution (7) gives rise to the following temporal dependence for the electron-spin polarization: $S_z(t) = S_z(0)\exp[-t/(2\tau_y)]\cos(\omega t)$. This dependence describes well the observed oscillations, see Fig. 3. The decay time of these oscillations, $2\tau_y \approx 1.4$ ns, is very close to the value obtained from the fit of the curve in Fig. 3, $T_2 = 1.36$ ns.

3. CONCLUSIONS

Our analysis shows that the main process efficiently suppressing electron-hole exchange interaction in nonradiative excitons is the scattering of these excitons followed by the mutual spin flips of like carriers (either two electrons or two holes). The characteristic time scale at which such scattering occurs in the experiments under discussion is units of picoseconds. During this time, the electron spin, undergoing precession in the effective field of a hole, does not noticeably change its orientation. Scattering results in the efficient averaging of hole-spin fluctuations, which is similar to the motional narrowing effect discussed in publications [10, 11]. As a result, the electron spins in nonradiative excitons undergo precession in an external magnetic field almost as free electron spins. Some traces of the exchange interaction are seen in the relaxation of spin polarization, which, as the analysis shows, is caused by this interaction rather than by interaction with phonons.

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CONFLICT OF INTEREST

The authors declare no conflict of interest.

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