Exciton spin decay modified by strong electron-hole exchange interaction

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(Dated: March 23, 2022)

We study exciton spin decay in the regime of strong electron-hole exchange interaction. In this regime the electron spin precession is restricted within a sector formed by the external magnetic field and the effective exchange fields triggered by random spin flips of the hole. Using Hanle effect measurements, we demonstrate that this mechanism dominates our experiments in CdTe/(Cd,Mg)Te quantum wells. The calculations provide a consistent description of the experimental results, which is supported by independent measurements of the parameters entering the model.

PACS numbers: 72.25.Rb, 72.25.Fe, 78.67.-n

Spin is a fundamental invariant of an electron. In semiconductors, the electron and hole spins can be optically oriented by a circularly polarized light \cite{1}. Recently much interest has been attracted to this field caused by possible applications (spintronics) \cite{2}. In particular, the spin trapped in a quantum dot has been proposed to serve as a qubit in the quantum computation schemes \cite{3} using a single CdTe/(Cd,Mg)Te quantum well. The calculations provide a consistent description of the experimental results, which is supported by independent measurements of the parameters entering the model.

The most widespread method for studying the dynamics of the non-equilibrium spin populations in semiconductors is optical orientation \cite{1}. Circularly polarized light excites electrons with a predominant spin polarization along the direction of the incident light beam. When an external magnetic field $B$ is applied in Voigt geometry, i.e., in the plane of the sample, the optically oriented electron spins start precessing with Larmor frequency $\Omega = g_e \mu_B B$ around the field direction (Fig. 1). Therefore, a detailed understanding of the spin decay processes are of crucial importance for the functionality of spin-based devices.

The half width at half maximum (HWHM) of the Hanle curve at $B = B_{1/2}$ corresponds to the condition $\Omega T_{se} = 1$, and hence allows determination of the electron spin lifetime $T_{se}$. For optically created electrons it is governed by the electron spin relaxation time $\tau_{se}$ and the lifetime time $\tau$ of the electron-hole pair (exciton): $T_{se}^{-1} = \tau_{se}^{-1} + \tau^{-1}$.

This classical picture will be modified drastically by isotropic electron-hole (eh) exchange interaction. It manifests itself as a splitting $\Delta_0$ between the radiative ($\pm 1$) and nonradiative ($\pm 2$) doublets of the neutral exciton \cite{5}. The electron spin now precesses in a total field formed by the external magnetic field $B$ and the exchange field of the hole $B_{ex} = \Delta_0 / |g_e| \mu_B$. Moreover, due to the hole spin relaxation, the exchange field $B_{ex}$ changes with time introducing additional difficulties for the theoretical description. The influence of the eh-exchange interaction \cite{6,7} and the importance of the hole spin relaxation rate on the electron spin decay \cite{8,9} have been recognized in previous theoretical works. However, the Hanle effect in the regime of the strong electron-hole exchange coupling, when the frequencies associated with the exchange interaction become greater than relaxation rates of electrons and holes, has not been investigated so far. The role of the exchange coupling is especially high in systems with reduced dimensionality like quantum dots (QDs) or quantum wells (QWs), where the exciton is confined in a small volume.

In this Letter we consider exciton spin decay in the regime of strong eh-exchange interaction. We demonstrate that in this limit the classical picture is not valid any more. While the Hanle curve still originates from depolarization of the electron spin in an external magnetic field, it occurs in a rather different manner and $B_{1/2}$ now depends on the eh-exchange splitting $\Delta_0$, the hole spin relaxation time $\tau_{ih}$, and the exciton life time $\tau$. We provide strong experimental evidence by comparing the Hanle curves of a single CdTe/(Cd,Mg)Te QW detected at the neutral exciton ($X^0$) and positively charged trion ($T^+$), where in the latter case the eh-exchange interaction is suppressed. We find that the characteristic depolarization field of the exciton Hanle curve $B_{1/2}^X$ is an order of magnitude larger than that of the trion Hanle curve $B_{1/2}^T$, indicating their different physics.

We now consider schematically the regime of strong eh-exchange interaction following the ideas of Ref. \cite{10} and discuss the limitations of this approach (see Fig. 1). Let as an example circularly polarized light excite exciton with the $z$-projection of the angular moment $J_z = -1$, consisting of an electron with spin up (+1/2) and a hole with spin down (–3/2). How will this system evolve in a magnetic field $B$ directed perpendicular to the quantization axis (Fig. 1)?

First, we assume that the in-plane hole g-factor is close to zero. This is valid when the heavy-hole band is split off...
Such a chain represents the evolution of the mean spin of the electron: the exchange field of the hole $B_{ex}$ and the spin dynamics the hole is capable of are random parallel to the $z$-axis (perpendicular the sample plane), and the spin evolution of the exciton. The flip of the hole moment of the hole spin flip, while the transverse component of the electron spin becomes random (Fig. 1). The above process is only a first link in the chain of decay of the new portion of the electron spin, etc. (Note that we implicitly assume a long electron spin relaxation time $\tau_{se} > \tau_{sh}$.)

As a result of the above deliberation, we can introduce a relaxation sector shown by the gray area in Fig. 1 which lies in the plane formed by $B$ and $B_{ex}$ fields. The relaxation sector restricts the directions of the electron spin and determines its decay path. This picture is thus valid for zero in-plane hole $g$-factor, and provided $1/\Omega_0 < \tau_{sh} < \tau_{se}, \tau$. As we show in the following, these conditions can indeed be realized experimentally.

The sample under study was grown by molecular beam epitaxy on a (001) GaAs substrate. The structure consists of a single (38-Å-wide) CdTe/Cd$_0.7$Mg$_0.3$Te QW and is nominally undoped. The photoluminescence (PL) was excited by a dye laser with tunable photon energy and detected with a 1-m spectrometer and a photomultiplier tube. In order to detect the polarization of the emission we used a standard scheme with a piezo-elastic modulator and a two-channel photon counter. Magnetic field was applied either perpendicular to the sample plane (Faraday geometry) or in the sample plane (Voigt geometry).

All experiments were carried out at a temperature 1.6 K. A typical PL spectrum is shown in Fig. 2(b). Generally, the spectrum consists of two bands. The high-energy peak is attributed to the neutral exciton $X^0$, and the low-energy one is ascribed to a charged exciton, or trion, $T^+$. Our analysis shows that we are dealing with the positively charged trion $T^+$, which consists of one electron and two holes. The holes form a spin singlet with zero total spin $s_h = 0$ as shown in Fig. 3(a), so the optical orientation signal from $T^+$ always reflects the spin polarization of the unpaired electron $s_e$. Moreover, for the same reason (i.e., $s_h = 0$) the exchange field is zero for a $T^+$ trion. $B_{ex} = 0$, which allows us to study the electron spin relaxation free of exchange interaction.

Figure 2(a) shows a typical spectrum of the optical orientation under quasi-resonant excitation at the trion line. While quite high in absence of external field, the optical orientation signal approaches zero already at a relatively low magnetic field of $B = 0.5$ T. The Hanle curve detected at the PL maximum is given in Fig. 2(c) by squares. It has a Lorentzian shape and is perfectly described by the classical formula derived for the free spin precession of an electron

$$P_{T^+} = 2s_e \frac{T_{se}}{\tau_r} \frac{2s_{e0}}{1 + (B/B_{T^+}^{1/2})^2},$$

with $s_e$ being its mean spin value and

$$B_{T^+}^{1/2} = \frac{\hbar}{g_e |\mu_B| T_{se}}$$

and

$$\frac{1}{T_{se}} = \frac{1}{\tau_r} + \frac{1}{\tau_{se}}.$$
excitation conditions as in panel (a). Solid line is the fit using Eq. 1 with characteristic field $B_{1/2} = 0.14$ T. (d) The Hanle effect measured for the exciton ($X^0$) at the same excitation conditions as in panel (b). Dashed line is the fit using Eqs. 10 and 11 with characteristic field $B_{1/2} = 1.1$ T. Magnetic fields are applied in Voigt geometry.

data in Fig. 2(c) is achieved with $2s_{e0}T_{ex}/\tau_s = 0.61$ and $B_{1/2}^T = 0.14$ T (solid line). Taking the electron g-factor $g_e \approx -1.3$ known from the spin-flip Raman scattering experiments [8] and the obtained value of $B_{1/2}^T$, we deduce for the electron spin lifetime $T_{ex} = 61$ ps. Assuming that the conditions of quasi-resonant excitation are close to the ideal case ($s_{e0} \approx 1/2$), one can estimate a recombination time of $\tau_s = 100$ ps. This value is in agreement with typical decay times of the trion PL obtained from direct time-resolved experiments on CdTe-based QWs [11]. In the case of nonresonant excitation [Fig. 2(b)] the HWHM of the trion Hanle curve [Fig. 2(d)] remains unchanged, again yields $B_{1/2}^T = 0.14$ T [Fig. 2(d)]. Reduction of the initial electron spin ($s_{e0} < 1/2$) is most likely caused by relaxation processes to the trion ground state.

In contrast to the positively charged trion, the neutral exciton $X^0$ displays a rather different behavior [see Fig. 2(d)]. First, while the magnetic field is increased, the polarization does not disappear but saturates at a nonzero level of ~ 0.1. Second, while there exists a field-dependent part of polarization which is well fitted by a Lorentzian curve, analogously to the case of the trion, the characteristic field of this depolarization equals $B_{1/2}^X = 1.1$ T, much larger than that of the $T^+$. (a) $T^+$ (b) $X^0$ (c) $B_{1/2}^T = 0.14$T (d) $B_{1/2}^X = 1.1$T

FIG. 2: (Color online) Spectra of PL and optical orientation for quasi-resonant excitation at the positively charged trion (a) and non-resonant excitation above the neutral exciton (b). (c) The Hanle effect measured for the trion ($T^+$) at the same excitation conditions as in panel (a). Solid line is the fit using Eq. 1 with characteristic field $B_{1/2} = 0.14$ T. (d) The Hanle effect measured for the exciton ($X^0$) at the same excitation conditions as in panel (b). Dashed line is the fit using Eqs. 10 and 11 with characteristic field $B_{1/2} = 1.1$ T. Magnetic fields are applied in Voigt geometry.

We now demonstrate that the experimental behavior is in good agreement with an analytical solution for the model presented in Fig. 3(a). Assuming $\tau_{sh} \ll \tau$ the electron depolarization essentially occurs for weak external fields $B \ll B_{ex}$. So the relaxation sector shown in gray in Fig. 3 is narrow, and the decrease of the electron spin density $S_e$ per single flip of the hole spin approximately equals $2S_e (B/B_{ex})^2$. The master equation for $S_e$ then takes a simple form

$$\dot{S}_e = s_{e0}G - S_e \frac{\tau_s}{\tau_{sh}} S_e \left( \frac{B}{B_{ex}} \right)^2; \quad (3)$$

here the first term describes the spin creation with the exciton recombination rate $G$ (with $s_{e0} \leq 1/2$ phenomenologically accounting for possible losses of the electron spin polarization in the higher-lying state where the exciton is actually created), the second term represents the losses by excitonic recombination, the third term describes the decrease of spin via the chain process of Fig. 3(a) ($2\tau_{sh}^{-1}$ has the sense of number of hole spin flips per unit of time). Setting $S_e$ to zero, one can find the steady-state value of...
$S_e$ as a function of $B$

$$S_e = s_{e0} \frac{G\tau}{1 + \frac{\tau}{\tau_{sh}}} \left( \frac{B}{B_{ex}} \right)^2.$$  
(4)

When $\tau_{sh} \ll \tau$, the correlation between the electron and hole spins is weak, so the intensities of the components of the luminescence with opposite circular polarizations ($I_+$ and $I_-$) depend only on the mean spin values $s_e = S_e/G\tau$ and $s_h = -\frac{1}{2}\tau_{sh}/\tau$:

$$I_\pm \propto \frac{1}{3} \left( \frac{1}{2} \pm s_e \right) \left( \frac{3}{2} \mp s_h \right).$$  
(5)

Using once more that $\tau_{sh}/\tau$ is small, we finally obtain the luminescence polarization:

$$P_{X0} \approx \frac{\tau_{sh}}{\tau} + \frac{2s_{e0}}{1 + \frac{\tau}{\tau_{sh}}} \left( \frac{B}{B_{ex}} \right)^2.$$  
(6)

The first (field-independent) contribution originates from the mean spin polarization of holes. Its value is controlled by the ratio of the hole spin relaxation time and the exciton lifetime. The inverse of this ratio also enters the second term describing the chain spin depolarization of electrons.

We now compare the result of the calculation with the experimental data. The ratio of the hole spin relaxation time and the exciton lifetime can be estimated from the high-field value in Fig. 2c, $\tau_{sh}/\tau \approx 0.1$ . This estimate looks reasonable, as for CdTe-based QWs the typical reported values of the hole spin relaxation time are on the order of tens of picoseconds [18]. Together with the value of the exchange field $B_{ex} \approx 4.5$ T taken from the experimental data in Fig. 3b, this gives a Lorentzian depolarization curve with a HWHM of 1.4 T, in reasonable agreement with the experimentally measured value $B_{1/2} = 1.1$ T. It turns out that in the strong exchange interaction regime, multiple flips of the hole spin result in a narrowing of the depolarization curve by a factor $\sqrt{\tau/\tau_{sh}} \approx 3.2$ as compared to the field of the exchange interaction:

$$B_{1/2} = B_{ex} \sqrt{\tau_{sh}/\tau}.$$  
(7)

Equation (4) summarizes our key result. Note that if the exciton lifetime $\tau$ is controlled by the radiative recombination time $\tau_r$, as for the trion ($\tau \approx \tau_r = 100$ ps), then $\tau_{sh} \approx 10$ ps and the condition of strong exchange interaction is reasonably satisfied, $g_0\tau_{sh} = 5$.

In summary, by studying the depolarization of the PL from the positive trion and from the exciton in a CdTe QW, we were able to investigate the exciton spin decay subject to strong exchange interaction. The spin of an electron interacting with a hole precesses in tilted magnetic fields, triggered by the random spin flips of the hole. This process results in a depolarization pattern with the characteristic field $B_{1/2} = \Delta_0/|g_e|\mu_B\tau_{sh}^{1/2}$, which was experimentally observed and verified by separate determination of $\Delta_0/|g_e|\mu_B$ and $\tau_{sh}^{1/2}/\tau$. The reported mechanism is of interest for nanostructures with sizable isotropic electron-hole exchange interaction (e.g., $\Delta_0 \approx 0.33$ meV in this work).

The study was supported by INTAS (03-51-5266) as well as by Russian Foundation for Basic Research and SFB 410.
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