Long-Term Hole Spin Memory in the Resonantly Amplified Spin Coherence of InGaAs/GaAs Quantum Well Electrons

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Pulsed optical excitation of the negatively charged trion has been used to generate electron spin coherence in an n-doped (In, Ga)As/GaAs quantum well. The coherence is monitored by resonant spin amplification detected at times exceeding the trion lifetime by 2 orders of magnitude. Still, even then signatures of the hole spin dynamics in the trion complex are imprinted in the signal leading to an unusual batlike shape of the magnetic field dispersion of spin amplification. From this shape information about the spin relaxation of both electrons and holes can be derived.

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The spins of carriers confined in quantum structures have attracted considerable attention because of the strongly changed spin relaxation mechanisms in these systems compared to bulk [1]. Several optical methods have been developed to address this problem [2–8]. One of the most sensitive techniques is polarized pump-probe spectroscopy measuring the photoinduced Faraday effect [7,8]. In undoped structures, this tool allows one to study the spin dynamics of excitons by tuning the laser in resonance with the corresponding optical transition. In doped structures, also the spin dynamics of resident electrons or holes can be studied by exciting the trion complex formed by the resident carrier and the photogenerated exciton [9,10].

Usually the resident carrier's spin dynamics spans over time scales exceeding by far the recombination of photocreated carriers. The spin coherence, in particular, of electrons, may even exceed the pulse separation of the periodic excitation. Therefore, spin polarization may accumulate from pulse to pulse [11,12]. If a magnetic field is applied perpendicular to the spin polarization created by light, the electron spin precession restricts this accumulation to cases where the precession frequency is a multiple of the laser repetition rate. This commensurability condition is applied in resonant spin amplification (RSA) [11], which probes the signal at long delays, shortly before the next pump pulse, when all contributions of photogenerated carriers have decayed so that a pure resident electron signal is detected.

In this Letter we generate spin coherence of quantum well electrons by optical excitation of the trion transition. The time evolution of the Faraday rotation (FR) signal, resulting from this type of excitation, is controlled by the interdependence of electron and hole spin dynamics. As a result traces of the hole spin can be found in the electron RSA long after the radiative trion decay.

We studied a sample containing two coupled, 8 nm wide $In_{0.09}Ga_{0.91}As$ quantum wells (QWs) separated by a 1.7 nm GaAs barrier and sandwiched between GaAs layers. This structure was selected on purpose because of pronounced carrier localization at cryogenic temperatures. It includes an n-doped GaAs buffer 100 nm below the QWs as a source of resident QW electrons. The two-dimensional electron density did not exceed 10^{10} cm^{-2}. In the photoluminescence (PL) spectrum two emission lines separated by 1.4 meV are observed [Fig. 1(a)], which we attribute to exciton (X, 1.4400 eV) and negative trion (T, 1.4386 eV) recombination [13]. Their full width at half maximum of 1 meV is caused by exciton localization in alloy and QW width fluctuations.

In the pump-probe FR experiments [14], we used a Ti:sapphire laser emitting 1.8 ps pulses with a repetition period $T_R = 13.2$ ns. The laser energy, same for pump and probe, was tuned into resonance with either the exciton or trion transition. The circular polarization of the pump beam was modulated at 50 kHz by an elasto-optical modulator. The excitation density was held low at 0.5 W/cm^2. FR of the linearly polarized probe beam was detected by a balanced photodiode scheme. Magnetic fields $\boldsymbol{B}$ were applied along the x axis perpendicular to the structure growth direction and light wave vector (Voigt geometry) along the z axis. The sample temperature was varied from 2 to 10 K. A typical FR trace is shown in Fig. 1(b).

Figures 1(c) and 1(d) give RSA signals recorded at the exciton and trion energies for $T = 2$ K. The probe pulse hit the sample at a delay of $(T_R - \delta)$ with $\delta = 10$ ps shortly before the pump pulse [11]. The shape of the exciton RSA curve in Fig. 1(c) agrees with RSA curves reported in literature [1,11]. The peak positions correspond to electron spin precession frequencies which are multiples of the laser repetition frequency. The decrease of the peak amplitude with increasing magnetic field is due to the ensemble spread of electron g factor.

The RSA spectrum for resonant trion excitation strongly differs from the exciton signal [see Fig. 1(d)]. First (and

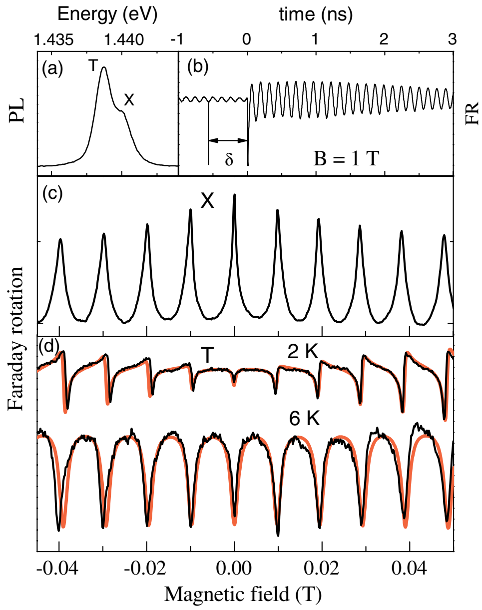

FIG. 1 (color online). (a) Photoluminescence spectrum of (In, Ga)As/GaAs QWs. (b) Time-resolved Faraday rotation signal recorded at trion resonance. (c) RSA signal for exciton resonance. (d) RSA signal for trion resonance. Black (grey) curves give experiment (theory). Calculation parameters are $|g_e| = 0.555$ and $\Delta g_e = 0.002$. For upper curve: $T_s^e = 55$ ns, $T_s^h = 2$ ns, and $\tau_r = 120$ ps. For lower curve: $T_s^e = 20$ ns, $T_s^h = 0.2$ ns, and $\tau_r = 100$ ps. In all panels $T = 2$ K, except for the lower trace in (d) with $T = 6$ K.

most important), the amplitude of the peaks increases with increasing magnetic field in contrast to a conventional RSA signal. Second, the peaks are inverted due to spectral dependence of the signal phase [15]. Third, the signal between peaks at $T = 2$ K is asymmetric.

Here we focus on the surprising magnetic field dependence of the trion RSA peak amplitude, for which we need to discuss the mechanisms of electron spin polarization: In an n-doped QW a trion is formed from a resident electron and a photocreated electron-hole pair. For circularly polarized excitation resonant with the trion, the angular momentum of the photon is transferred to an electron and hole according to the selection rules [2]. In an (In, Ga)As/GaAs quantum well absorption of circularly polarized light with angular momentum + 1 (assumed also in the following) creates an electron with spin projection $s_{e,z} = -1/2$ and a hole with angular momentum $j_{h,z} = +3/2$ along the z axis. For trion formation, the resident electron spin has to be antiparallel to the spin of the photocreated electron. Thereby the total spin of the remaining resident electron ensemble is reduced by $+1/2$ so that the ensemble becomes spin polarized [9].

At zero magnetic field, the spin polarization of the resident electrons is preserved after trion recombination only in the case of fast hole spin relaxation in the trion [16,17]: If the hole spin relaxation time, T_s^h, is long compared to the trion recombination time, τ_r, the resident electron reappears after trion recombination with the same spin orientation as before trion formation. Therefore the induced spin polarization of the resident electrons disappears. Vice versa, if the hole spin relaxation is fast such that $T_s^h \ll \tau_r$, the hole in the trion on average can recombine with electrons of both orientations. Consequently, electrons polarized by light can accumulate.

In a transverse magnetic field the spins of the resident electrons which are not bound to trions precess about the field, resulting in a rotation of the electron polarization during the trion lifetime. Therefore the induced polarization is not fully annihilated after trion recombination, even when the hole spin has not relaxed. Faster electron spin precession in stronger magnetic fields leads to an increase of the spin polarization. This is reflected in Fig. 1(d) by the increasing RSA amplitudes with increasing magnetic field at $T = 2$ K.

These considerations show the strong interdependence of electron and hole spin dynamics. For a quantitative analysis, we describe the spin states of the resident electron and singlet trion (hole) by polarization vectors $\boldsymbol{s}$ and $\boldsymbol{j}$, respectively. The coupled dynamics of these polarizations in the magnetic field can be seen from their equations of motion [14,16,18]:

$$\frac{d\boldsymbol{j}}{dt} = -\frac{\boldsymbol{j}}{T_s^h} - \frac{\boldsymbol{j}}{\tau_r} \equiv -\frac{\boldsymbol{j}}{\tau_T}, \tag{1}$$

$$\frac{d\boldsymbol{s}}{dt} = \frac{g_e \mu_B}{\hbar}(\boldsymbol{B} \times \boldsymbol{s}) - \frac{\boldsymbol{s}}{T_s^e} + \frac{(\boldsymbol{j}\boldsymbol{z})\boldsymbol{z}}{\tau_r}. \tag{2}$$

Here $\boldsymbol{z}$ is the unit vector along z axis, μ_B is the Bohr magneton, and g_e is the electron g factor. We neglect the Larmor precession of the hole spin because the in-plane hole g factor is close to zero [10,19]. In both equations phenomenological terms describing relaxation processes are included. In Eq. (1) the loss of trion polarization due to hole spin relaxation (T_s^h) and trion recombination (τ_r) is taken into account, resulting in a trion spin lifetime $\tau_T = T_s^h \tau_r/(T_s^h + \tau_r)$. Equation (2) describes the electron spin precession in the magnetic field (first term), the decrease of spin polarization due to spin relaxation (T_s^e, second term), and the change of electron spin polarization by trion recombination (third term). Note that the lifetime of the trion spin polarization is limited by the recombination time τ_r (fractions of a nanosecond), whereas the lifetime of the resident electron polarization is limited only by its spin relaxation time, T_s^e. The reason for that is that the resident electrons contributing to the FR signal are not bound to trions. The trion spin relaxation and recombination control only the generation efficiency of electron spin coherence, but do not influence its decay. The solution of Eq. (1) gives $j_z(t) = j_{z0} \exp(-t/\tau_T)$ with the initial value j_{z0}. The reference time zero coincides with the end of the pump pulse.

Besides depending on pump power, j_{z0} depends also on the resident electron spin state, which has to be antiparallel to the photocreated electron. Denoting the z component of the resident electron spin polarization right before pump pulse arrival at time $t \to T_R$ (i.e., $\delta \to 0$), by $\tilde{s}_z$, j_{z0} is given by $(2\tilde{s}_z + 1)\sin^2(\Theta/2)/4$ for σ^+ polarized excitation [14,16]. Here the pump power is expressed in terms of the pulse area $\Theta = \int 2|\langle d\rangle E(t)|dt/\hbar$, where $\langle d\rangle$ is the dipole transition matrix element, and $E(t)$ is the electric field of the laser pulse. In our experiment the pump efficiency was small, so that j_{z0} is approximately proportional to the pump power.

For solving Eq. (2) we introduce $s^+ = s_z + is_y$, for which we obtain with $s_0^+ = s_{z0} + is_{y0}$ at time zero

$$s^+(t) = \left(s_0^+ - \frac{j_{z0}}{\tau_r\Omega}\right)e^{-[i\omega_e + (1/T_s^e)]t} + \frac{j_{z0}e^{-t/\tau_T}}{\tau_r\Omega}, \quad (3)$$

where $\Omega = 1/T_s^e - 1/\tau_T + i\omega_e$, and the electron spin precession frequency $\omega_e = g_e\mu_B B/\hbar$. s_0^+ is determined by the pump intensity and the resident electron spin polarization $\tilde{s}_z$ and $\tilde{s}_y$ shortly before the pump [14,16]:

$$s_{z0} = \tilde{s}_z[1 - \sin^2(\Theta/2)/2] - \sin^2(\Theta/2)/4,$$
$$s_{y0} = \tilde{s}_y\cos(\Theta/2). \quad (4)$$

In strong magnetic fields ($\omega_e \gg 1/\tau_r$) the trion contribution to $s^+(t)$ [terms proportional to j_{z0} in Eq. (3)] is negligible due to fast dephasing in the electron spin ensemble, which removes completely the electron signal at $T_R - \delta$ delays [12]. However, in weak magnetic fields this contribution becomes important. From Eq. (3) strong oscillations appear in the FR signal after pump pulse action due to electron spin precession about the magnetic field. These oscillations decay with the electron spin relaxation time T_s^e.

An infinite sequence of pump pulses with repetition period T_R creates an electron spin polarization periodic in time [12,18]. When the first pulse arrives at time zero, $\tilde{s}_{y(z)} = s_{y(z)}(nT_R - \delta)$, $\delta \to 0$, before action of the $(n+1)$-th pulse. After an extended pulse sequence $(n \to \infty)$, $s_z[(n+1)T_R - \delta]$ should be equal to $s_z(nT_R - \delta)$, so that one obtains for $\tilde{s}_{y(z)}$

$$\tilde{s}_z = \frac{v}{2}\left[\frac{u(1+\alpha)e^{-T_R/T_s^e} - (1+\alpha)\cos(\omega_e T_R) - \beta\sin(\omega_e T_R)}{1 + u[1 - v(1+\alpha)]e^{-2T_R/T_s^e} - [1 + u - v(1+\alpha)]\cos(\omega_e T_R)e^{-T_R/T_s^e} - \beta v\sin(\omega_e T_R)}\right]e^{-T_R/T_s^e}, \quad (5)$$

$$\tilde{s}_y = \frac{v}{2}\left[\frac{\beta e^{-T_R/T_s^e} - \beta\cos(\omega_e T_R) + (1+\alpha)\sin(\omega_e T_R)}{1 + u[1 - v(1+\alpha)]e^{-2T_R/T_s^e} - [1 + u - v(1+\alpha)]\cos(\omega_e T_R)e^{-T_R/T_s^e} - \beta v\sin(\omega_e T_R)}\right]e^{-T_R/T_s^e}, \quad (6)$$

where $v \equiv \sin^2(\Theta/2)/2$, $u \equiv \cos(\Theta/2)$, $\alpha \equiv \mathrm{Re}[1/(\tau_r\Omega)]$, and $\beta \equiv \mathrm{Im}[1/(\tau_r\Omega)]$.

The RSA signal is proportional to the spin polarization summed over the electron ensemble, $S_z = \sum s_z$. Figure 2 shows S_z shortly before pulse arrival calculated with Eqs. (3)–(6) for $T_s^e = 55$ ns and different hole spin relaxation times. With increasing T_s^h the RSA shape changes strongly, both with respect to the peak amplitude and the signal shape between the peaks. The upper curve (with a

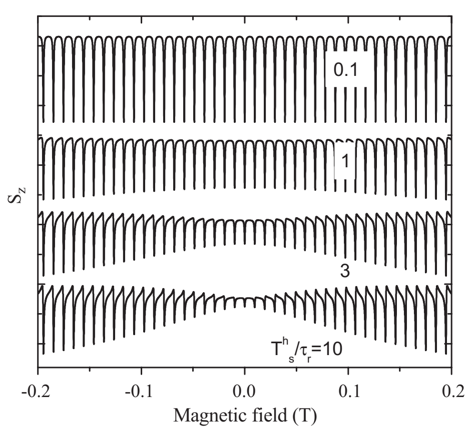

FIG. 2. Calculated RSA spectra for a homogeneous spin ensemble excited at trion resonance, based on Eqs. (3)–(6). $\delta = 10$ ps, $|g_e| = 0.555$ (determined from RSA peak distance), $\Delta g_e = 0$, $T_s^e = 55$ ns, and $\tau_r = 120$ ps for different ratios T_s^h to τ_r, given at each curve.

short $T_s^h = 12$ ps) looks like an ordinary RSA signal [11], in contrast to our experiment. If T_s^h approaches τ_r, the peak amplitudes decrease toward zero field. Only if $T_s^h \gg \tau_r$ does the RSA signal reproduce the experimental data [upper curve in Fig. 1(d)]. Therefore the imprint of the hole spin is essential for understanding the RSA spectra, even though the FR signal is solely contributed by the resident electron magnetization. This memory about the hole spin dynamics arises from the hole involvement in the generation of resident electron spin polarization. For the calculations a small pulse area $\Theta = 0.056\pi$ was used. Calculations show, however, that an increase of Θ up to 0.22π does not change the RSA signal shape.

To describe the experiment this consideration has to be extended by the ensemble inhomogeneity, which leads to a spread of the electron g factor Δg_e, resulting in reversible phase relaxation of the total spin polarization [14]. This leads to a decrease of the RSA peak amplitudes with increasing field [11]. Figure 3 compares theoretical and experimental RSA signals over an extended range of fields. The good agreement suggests that the model captures the key features of the spin dynamics. In particular, the characteristic bat shape as opposed to the conventional bell shape is well reproduced. The increase of the RSA peak amplitudes in weak fields is overpowered at stronger fields by the inhomogeneity of precession frequencies. For better comparison, Fig. 1(d) gives a close-up of the small field range. From fitting the experiment we obtained $T_s^h = 2$ ns.

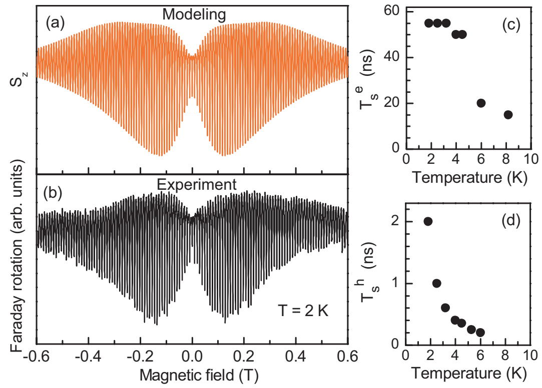

FIG. 3 (color online). (a) RSA signal calculated using Eqs. (3) and (5) with parameters as for the upper curve in Fig. 1(d). (b) Experimentally measured RSA signal for $\delta = 10$ ps. (c, d) Temperature dependencies of electron and hole spin relaxation times.

Reported hole spin relaxation times in QWs are much shorter, varying from 4 ps to 1 ns [5,9,10,19,20]. The hole spin dynamics outlasts the hole lifetime (limited by trion recombination) about 20 times, but surprisingly it still can be evaluated with high accuracy [21]. Measurements of the hole spin dynamics up to now required either resident holes in p-doped samples [10] or holes bound to excitons in undoped samples [19], for which the hole spin dynamics is shortened by exchange scattering with the electron and can be measured only during the exciton lifetime.

We used the RSA technique to measure the temperature dependencies of electron and hole spin relaxation times [Figs. 3(c) and 3(d)]. In our experiment the electron time T_s^e corresponds to the dephasing time $T_{2,e}^*$ of the electron spin ensemble precessing about B. At $T = 2$ K this time has a remarkably large value of 55 ns, which is the longest time reported so far for QWs [17,22]. The reason for that is the localization of the resident electrons, which in a ternary (In, Ga)As QW is provided by well width and alloy fluctuations. Spin relaxation of the localized electrons below 4 K is limited by their hyperfine interaction with lattice nuclei. The Dyakonov-Perel spin relaxation mechanism [1] which is efficient for free electrons is suppressed for localized ones, but becomes dominant for $T > 6$ K, when electrons became delocalized.

The hole spin dynamics shows a temperature dependence similar to the electron one, but the increase of T_s^h with decreasing temperature continues well below 5 K, where it is natural to assume that the holes are localized. The time increases from 0.2 to 2 ns in the range 2–6 K. This strong dependence is also evident from the RSA spectra in Fig. 1(d) where the decrease of the peak amplitudes toward zero field nearly vanishes by increasing temperature to 6 K, witnessing the T_s^h shortening [23]. As the hyperfine interaction is significantly weaker for holes than for electrons, the spin-orbit coupling should be responsible for the strong temperature dependence of T_s^h for the localized holes.

Calculations for strongly confined holes in quantum dots predict T_s^h times in the μs-ms range [24], as confirmed by recent experiments [25,26]. For strong confinement the phonon interaction is too weak to provide short hole spin relaxation at cryogenic temperatures. In our studies of the (In, Ga)As QW the energy separation of the localized hole state from higher lying localized states or those in the QW continuum is of order sub-meV, so that phonon scattering into the continuum may change significantly in the temperature range of relevance.

In summary, the spin dynamics of resident electrons in n-doped (In, Ga)As/GaAs QWs was studied under selective excitation of trions. The RSA signal carries not only information on the electron spin relaxation but also on its generation, from which one can obtain information on the hole spin relaxation. This allows us to measure hole spin relaxation times exceeding by orders of magnitude their lifetimes.

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