Spin Coherence of Holes in GaAs/(Al, Ga)As Quantum Wells

M. Syperek,[1,2] D. R. Yakovlev,[1,*] A. Greilich,[1] J. Misiewicz,[2] M. Bayer,[1] D. Reuter,[3] and A. D. Wieck[3]

[1]*Experimentelle Physik II, Universität Dortmund, D-44221 Dortmund, Germany*
[2]*Institute of Physics, Wrocław University of Technology, 50-370 Wrocław, Poland*
[3]*Angewandte Festkörperphysik, Ruhr-Universität Bochum, D-44780 Bochum, Germany*
(Received 5 March 2007; published 29 October 2007)

Carrier spin coherence in a p-doped GaAs/(Al, Ga)As quantum well with a diluted hole gas is studied by picosecond pump-probe Kerr rotation. For resonant optical excitation of the positively charged exciton the spin precession shows two types of oscillations: Electron spin beats decaying with the charged exciton radiative lifetime of 50 ps, and long-lived hole spin beats with dephasing times up to 650 ps, which decrease with increasing temperature, underlining the importance of hole localization. The mechanism of hole spin coherence generation is discussed.

DOI: 10.1103/PhysRevLett.99.187401 PACS numbers: 78.67.De, 78.55.Cr

Recently the investigation of the coherent spin dynamics in semiconductor quantum wells (QWs) and quantum dots (QDs) has attracted much attention, due to the possible use of the spin degree of freedom for spin-based electronics and quantum-information processing [1–3]. Until now the interest has been mostly focused on the electron spin, while information about the hole spin coherence is limited [4]. The hole as a Luttinger spinor has properties strongly different from the electron spin, such as strong spin-orbit coupling, strong directional anisotropy, etc. For localized holes most spin relaxation mechanisms are strongly suppressed, including the hyperfine interaction with the nuclear spins, which is inherent for electrons [5]. Therefore long hole spin coherence times are expected. The hole plays an important role also in coherent control of electron spins, since in many optical schemes charged electron-hole complexes are proposed as intermediate manipulation states [6].

The hole spin dynamics in GaAs-based QWs was measured by optical orientation detecting photoluminescence (PL) either time-integrated or time-resolved [4,7–10]. Experiments addressed the longitudinal relaxation time T_1 [7–9] and dephasing time T_2^* through hole spin quantum beats [4]. The reported times vary from 4 ps [7] to $\sim$1 ns [4,9], demonstrating strong dependencies on doping density and excitation energy.

A major drawback of PL techniques is, however, that spin coherence can be traced only as long as electrons and holes are present and recombine. Further, they work only for studying spin dynamics of minority carriers in a sea of majority carriers and are therefore restricted to undoped or n-type doped QWs. However, then the holes can interact with electrons, providing additional relaxation channels via exchange or shake-up processes [9,11]. It is challenging to study the hole spin coherence in p-doped QWs unperturbed by these factors. A pump-probe Kerr rotation (KR) technique using resonant excitation of the charged exciton allows realization of such measurements. Up to now it was applied only to bulk p-type GaN [12], but not yet to low-dimensional systems.

The theoretical analysis of hole spin dynamics in QWs has been focused on free holes [11,13–15], considering different relaxation mechanisms: (i) a Dyakonov-Perel–like mechanism, (ii) an acoustic phonon assisted spin-flip due to spin mixing of valence band states, (iii) an exchange induced spin-flip due to scattering on electrons, resembling the Bir-Aronov-Pikus mechanism for holes. Recently the role of hole localization and dephasing due to in-plane g factor variations were calculated [16].

In this Letter we use time-resolved pump-probe Kerr rotation [17] to investigate the spin coherence of holes in a p-doped $GaAs/Al_{0.34}Ga_{0.66}As$ single QW with a low hole density. We find spin dephasing times reaching the nanosecond range at temperature $T = 1.6$ K with a hole in-plane g factor of about 0.01. The dephasing time decreases with increasing temperature, suggesting the importance of hole localization. We discuss a mechanism that provides generation of spin coherence for the hole gas under resonant excitation of the positively charged exciton.

The QW structure was fabricated by molecular-beam epitaxy on a (100) oriented GaAs substrate. A 15 nm-wide GaAs QW was grown on top of a 380 nm-thick $Al_{0.34}Ga_{0.66}As$ barrier and overgrown by a 190 nm-thick $Al_{0.34}Ga_{0.66}As$ layer. 21 nm-thick layers with $Al_{0.34}Ga_{0.66}As$ effective composition obtained by GaAs/AlAs short-period superlattices were deposited on both QW sides, to improve the interface planarity. Two δ-doped carbon acceptor sheets were positioned symmetrically to the QW at 110 nm distance. The QW hole gas concentration and mobility in the diluted hole gas (DHG) regime are 1.51×10^{11} cm^{-2} and 1.2×10^5 cm^2/V s, respectively, as determined by Hall measurements at $T = 4.2$ K. It was possible to deplete the hole density in the QW by above barrier illumination and even invert the majority carrier type, resulting in a diluted electron gas (DEG).

A mode-locked Ti:sapphire laser with 75.6 MHz repetition rate and pulse duration of $\sim$1.5 ps was used for optical excitation. The laser beam was split into a circularly polarized pump and a linearly polarized probe. Both beams were focused on the sample to a spot diameter of

$\sim$100 μm. Magnetic fields $B \leq 10$ T were applied normal to the structure growth z axis (Voigt geometry). In the KR experiment the pump pulse propagates along z, coherently exciting carriers with spins polarized along z. The subsequent coherent spin evolution in form of precession about B is tested by the change of the linear probe pulse polarization. To detect this change (the KR angle), a homodyne technique based on phase-sensitive balanced detection was used.

PL spectra excited above and below the band gap of the $Al_{0.34}Ga_{0.66}As$ barriers are shown in Fig. 1(b) at $B = 0$ and 7 T. A single PL line corresponding to the positively charged trion T^+ (consisting of two holes and one electron) is seen in the DHG regime with below-barrier excitation. After inverting the type of majority carriers to the DEG regime by above barrier illumination the PL spectra consist of the exciton (X) and negatively charged trion (T^-) lines.

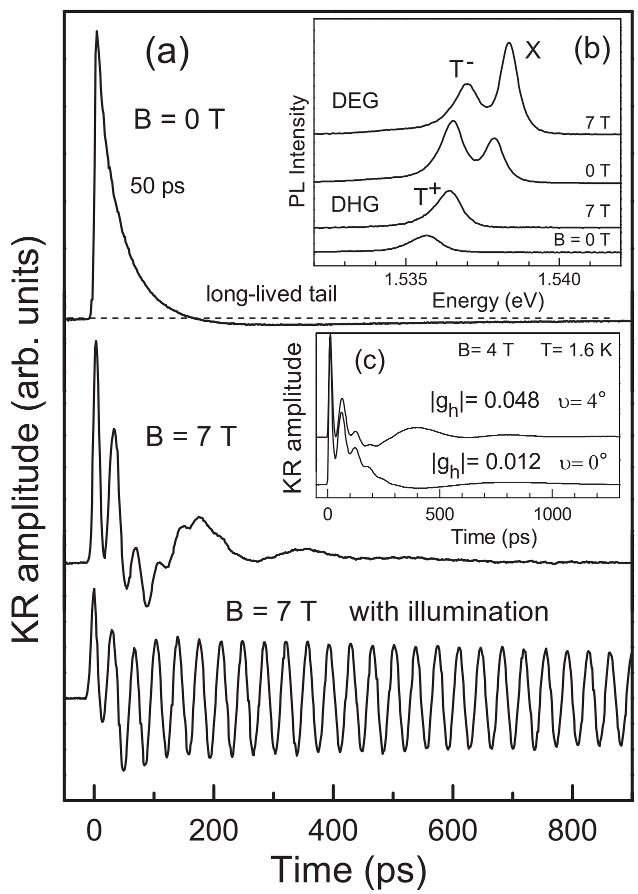

FIG. 1. (a) KR traces for a p-type 15 nm-wide GaAs/$Al_{0.34}Ga_{0.66}As$ QW versus time delay between pump and probe pulses at $B = 0$ and 7 T with field tilted by $\vartheta = 4°$ out of QW plane. Laser at 1.5365 eV is resonant with T^+. Power was 5 and 1 W/cm^2 for pump and probe, respectively. Bottom trace was recorded with additional laser illumination at 2.33 eV. $T =$ 1.6 K. (b) PL spectra for DHG (excitation at 1.579 eV) and DEG regime (above barrier excitation at 2.33 eV). (c) Comparison of KR traces for $\vartheta = 0$ and 4°.

The type of majority carriers in the QW can be identified by the KR signals at $B = 7$ T. The bottom trace in Fig. 1(a) was measured in the DEG regime and shows long-lived electron spin beats with a dephasing time of 2.5 ns, considerably longer than the radiative decay time of resonantly excited trions of $\sim$50 ps. The precession frequency corresponds to a g factor $|g_e| = 0.285 \pm 0.005$, typical for electrons in GaAs-based QWs. In the DHG regime, the fast electron precession is observed only during $\sim$200 ps after pump pulse arrival [see mid trace in Fig. 1(a)]. This signal is caused by coherent precession of the electron in the T^+ and disappears with trion recombination. The electron beats are superimposed on the hole beats with a much longer precession period. The hole beats decay with about 100 ps time constant and can be followed up to 500 ps delay. At these long times the KR signal is solely given by the hole precession.

Experimentally, it is difficult to observe the hole spin quantum beats due to the very small in-plane hole g factor. To enhance visibility we tilted the magnetic field slightly out of the QW plane by an angle $\vartheta = 4°$, due to which the hole g factor is increased by mixing the in-plane component ($g_{h,\perp}$) with the one parallel to the QW growth axis ($g_{h,\parallel}$), which typically is much larger [18]: $g_h(\vartheta) = \sqrt{g_{h,\parallel}^2 \sin^2\vartheta + g_{h,\perp}^2 \cos^2\vartheta}$. The strong change of the hole beat frequency with tilt angle is seen in Fig. 1(c). The frequency is analyzed by $\omega_h = \mu_B |g_h| B/\hbar$ with the Bohr magneton μ_B, and gives $|g_{h,\perp}| = 0.012 \pm 0.005$ for $\vartheta = 0°$ and $|g_h| = 0.048 \pm 0.005$ for $\vartheta = 4°$.

The electron and hole contributions to the KR amplitude, Θ_K, can be separated by fitting the data with a superposition of exponentially damped harmonics for electrons and holes:

$$\Theta_K = \sum_{i=e,h} A_i \exp\left(-\frac{\Delta t}{T_{2,i}^*}\right) \cos(\omega_i \Delta t). \quad (1)$$

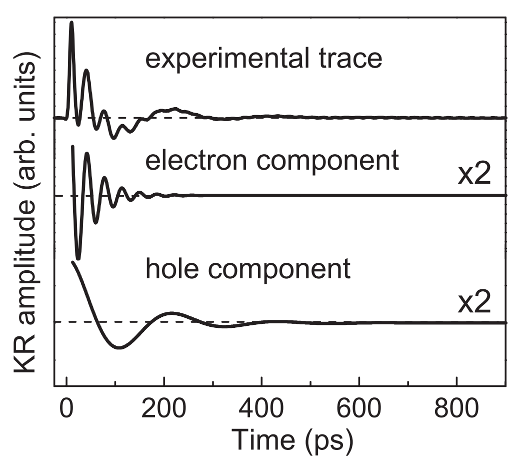

FIG. 2. Top trace: KR signal measured at $B = 7$ T for $\vartheta = 4°$. Bottom traces are obtained by separating electron and hole contributions (see text). Excitation conditions as in Fig. 1.

A_i are the signal amplitudes at pump-probe delay $\Delta t = 0$. $T_{2,i}^*$ are the spin dephasing times, which are contributed by the coherence time of individual spins, T_2, and inhomogeneous dephasing time due to spin precession frequency variations, $T_{2,\text{inh}}^*$: $1/T_2^* = 1/T_2 + 1/T_{2,\text{inh}}^*$. An example for a KR signal decomposition in the DHG regime is shown in Fig. 2.

We turn now to the hole coherence. Figure 3 shows the hole contribution to the KR signal for different B at $T = 1.6$ K. From fits by Eq. (1) we have obtained the dephasing times T_2^*, which are plotted versus B in the inset. A long-lived hole spin coherence with $T_2^* = 650$ ps is found at $B = 1$ T. With increasing B up to 10 T it shortens to 70 ps. The field dependence can be well described by a $1/B$ form showing that the dephasing arises from inhomogeneity of the hole g factor $\Delta g_h = 0.0007$, which is translated into a spread of precession frequencies: $\Delta\omega_h = \Delta g_h \mu_B B/\hbar$. Since $T_{2,\text{inh}}^* \propto 1/\Delta\omega_h$, this explains the $1/B$ dependence of the dephasing time.

In a KR experiment we do not measure the spin coherence time T_2 of an individual hole, but rather the ensemble dephasing time T_2^* [16], which is a lower boundary for the coherence time T_2. Therefore, we conclude that the T_2 for holes is at least 650 ps in our sample. Two sets of experimental data measured for pump to probe powers 1 to 5 and 5 to 1 W/cm^2 are compared in the inset of Fig. 3. The very similar results demonstrate performance of the experiment in the linear regime of both pump and probe beams for powers not exceeding 5 W/cm^2.

Insight into the origin of the long hole spin coherence can be taken from KR at varying temperatures. The data in Fig. 4 measured at $\vartheta = 5°$ show that the dephasing time T_2^* decreases from 110 to 60 ps when increasing the temperature from 1.6 to 6 K. This can be explained by hole localization in QW width fluctuations. The localization energy does not exceed 0.5 meV, which is comparable to the thermal energy at $T = 6$ K. Free holes are expected to have a short spin coherence time T_2 limited by the efficient relaxation mechanisms due to the spin-orbit interaction [11,14,15]. For localized holes these mechanisms are mostly switched off. Thermal delocalization of holes decreases the role of potential fluctuations and reduces Δg_h, which should lead to longer inhomogeneous dephasing times $T_{2,\text{inh}}^*$. On the other hand, then the fast decoherence of free holes becomes the limiting factor for the spin beat dynamics when $T_2 < T_{2,\text{inh}}^*$. The temperature dependence of T_2^* in the inset of Fig. 4 results from these two factors. The data are in line with the temperature dependence of the free hole spin relaxation times in n-type doped QWs [9].

For the tilted field geometry in Fig. 4 the spin beats are controlled by the parallel component of the hole g factor, $g_{h,\parallel}$, as the in-plane hole g factor, $g_{h,\perp}$, is close to zero for free holes [4,16,19]. With increasing temperature from 1.6 to 6 K the precession frequency decreases notably corresponding to a g factor decrease from 0.057 to 0.030. The hole g factor is controlled by mixing of heavy-hole and light-hole states, which in turn is affected by hole localization [19,20]. The temperature variation of the hole g factor is consistent with a hole delocalization.

Let us discuss now the mechanism for optical generation of hole spin coherence in a QW with a DHG, which is similar to the one suggested for singly charged QDs [21,22] and QWs with a DEG [23]. In our experiment pump and probe are resonant with T^+. Because of the considerable heavy-light hole splitting, the circularly polarized pump creates holes and electrons with well-defined

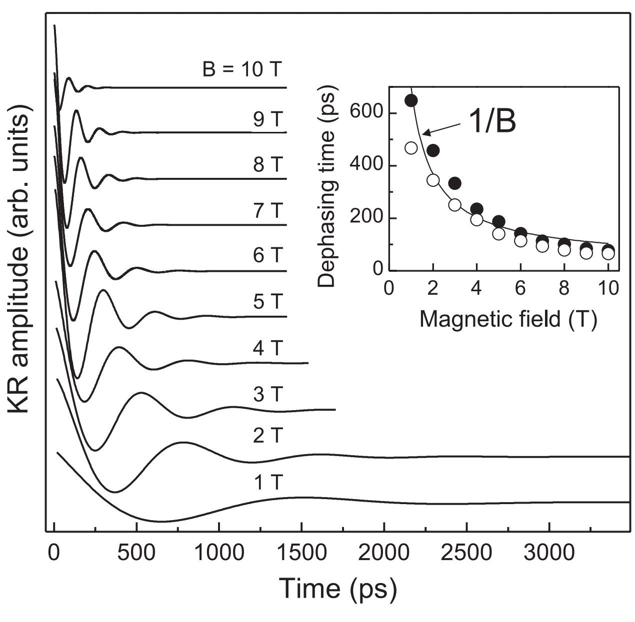

FIG. 3. Hole component extracted from fit to KR signal at different magnetic fields and $\vartheta = 4°$. Inset shows B dependence of the hole dephasing time T_2^*. Solid line is $1/B$ fit to data. Closed and open circles give data measured for pump to probe powers of 1 to 5 and 5 to 1 W/cm^2, respectively. $T = 1.6$ K.

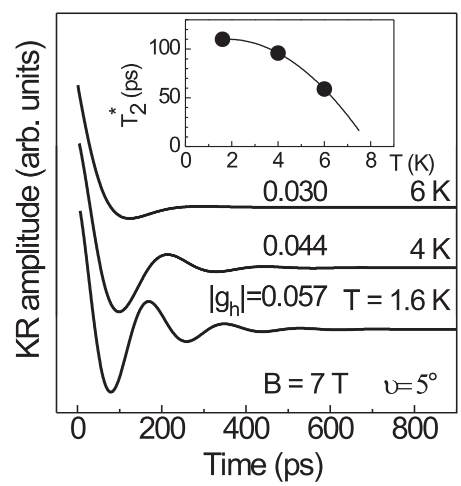

FIG. 4. Temperature dependence of hole component obtained from fit to KR signal at $B = 7$ T and $\vartheta = 5°$. Pump and probe powers are 1 and 5 W/cm^2, respectively. Inset: Hole spin dephasing time T_2^* versus temperature.

spin projections, $J_{h,z} = \pm 3/2$ and $S_{e,z} = \pm 1/2$, respectively, according to the optical selection rules [13]. Therefore, $|\Uparrow\Downarrow\downarrow\rangle$ ($|\Uparrow\Downarrow\uparrow\rangle$) trions can be generated by a σ^+ (σ^-) polarized pump. Here the thick and thin arrows give the spin states of holes and electrons, respectively.

The pump pulse duration is much shorter than the spin coherence and electron-hole recombination times. If in addition the pump duration is shorter than the charge coherence time of the trion the pulse creates a coherent superposition of a resident hole from the DHG and a hole singlet trion T^+. The spin state of a resident hole with arbitrary spin orientation before excitation can be described by $\alpha\,|\Uparrow\rangle + \beta\,|\Downarrow\rangle$, where $|\alpha|^2 + |\beta|^2 = 1$. Without magnetic field and for fields oriented normal to the z axis, the net spin polarization of the DHG is zero, so that the ensemble averaged coefficients are equal: $\bar{\alpha} = \bar{\beta}$.

For σ^+ polarized excitation, for which injection of an $|\Uparrow\downarrow\rangle$ electron-hole pair is possible, the excited superposition is given by $\alpha\,|\Uparrow\rangle + \beta\cos(\Theta/2)\,|\Downarrow\rangle + i\beta\sin(\Theta/2)\,|\Uparrow\Downarrow\downarrow\rangle$. Here $\Theta = \int \mathbf{d}\cdot\mathbf{E}(t)dt/\hbar$ is the dimensionless pulse area with the pump laser electric field $\mathbf{E}(t)$ and the dipole transition matrix element $\mathbf{d}$. Dephasing of the hole-trion superposition occurs shortly after the pulse on a time scale of a few picoseconds, converting the coherent polarization into a population of holes with spins $\Uparrow$ and $\Downarrow$ and of trions with $\Uparrow\Downarrow\downarrow$.

At $B = 0$, the carrier spins experience no Larmor precession. The electron spin relaxation time usually exceeds by 1–2 orders of magnitude the trion lifetime. Trion recombination returns the hole to the DHG with the same spin orientation as before the pump, if no electron spin scattering occurred in the meantime. This compensates the induced spin polarization and nullifies the KR signal at delays exceeding the trion lifetime. Indeed, the top trace KR signal in Fig. 1(a) shows a fast decay with a time constant of ~50 ps, which is characteristic for radiative trion recombination in GaAs QWs [24]. The weak long-lived tail of the signal is due to hole coherence provided by weak electron spin relaxation in T^+ and/or hole relaxation in the DHG during the trion lifetime.

In finite magnetic fields, the carrier spins start to precess about B. Because of the electron precession in T^+, the hole spin returned to the DHG after trion recombination will not compensate the spin polarization of the resident holes. Therefore, a long-lived hole coherence with considerable amplitude is induced. This coherence is observed in the KR signal as a low frequency spin beat signal (see Figs. 1 and 3). Note that the Larmor precession of resident holes may also contribute to generation of hole spin coherence, but the effect is proportional to the ratio of hole and electron Larmor frequencies and therefore rather small.

Let us compare the spin coherence generation for QWs with DHG and DEG resonantly excited in the T^+ and T^- states, respectively. We are interested in a long-lived spin coherence beyond the trion lifetime, i.e., in spin coherence of the resident carriers. In both cases the KR signal amplitude is controlled by the ratio of electron spin beat period to trion lifetime. Nevertheless, the two cases are quite different as for DHG the precessing electron is bound in the T^+ trion, while for DEG the background electron precesses. In the latter case the electron precession in T^- is blocked due to the singlet spin character of the trion ground state.

To conclude, a long-lived spin coherence has been found for localized holes in a GaAs/(Al, Ga)As QW with a diluted hole gas. The spin coherence time exceeds 650 ps and is masked by the spin dephasing due to g factor inhomogeneities. Localization of holes suppresses most spin relaxation mechanisms inherent for free carriers.

This work was supported by BMBF "nanoquit" and DFG (Grant No. YA65/5-1).

*Also at Ioffe Physico-Technical institute, Russian Academy of Sciences, St. Petersburg, Russia.

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