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Engineering ultralong spin coherence in two-dimensional hole systems at low temperatures

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Abstract. For the realization of scalable solid-state quantum-bit systems, spins in semiconductor quantum dots (QDs) are promising candidates. A key requirement for quantum logic operations is a sufficiently long coherence time of the spin system. Recently, hole spins in III–V-based QDs were discussed as alternatives to electron spins, since the hole spin, in contrast to the electron spin, is not affected by contact hyperfine interaction with the nuclear spins. Here, we report a breakthrough in the spin coherence times of hole ensembles, confined in the so-called natural QDs, in narrow GaAs/AlGaAs quantum wells at temperatures below 500 mK. Consistently, time-resolved Faraday rotation and resonant spin amplification techniques deliver hole-spin coherence times, which approach in the low magnetic field limit values above 70 ns. The optical initialization of the hole spin polarization, as well as the interconnected electron and hole spin dynamics in our samples, are well reproduced using a rate equation model.

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Contents

1. Introduction	2
2. Sample design and experimental methods	2
3. QW width dependence of hole SDT	4
4. Buildup of a resident hole spin polarization	8
5. RSA measurements of hole spin dynamics	10
6. Conclusions	13
Acknowledgments	14
References	14

1. Introduction

Among the most promising systems for the realization of quantum computing devices are spins in semiconductor quantum dots (QDs) [1]. Using electrostatic gating techniques, these dots may be defined within a two-dimensional electron system (2DES) by local depletion. This approach has the advantage that it allows for the fabrication of scalable arrays of quantum bits, as it is based on high-resolution lithography techniques instead of self-organized growth of QDs. While the spin dephasing of electrons in high-mobility GaAs/AlGaAs-based 2DES is extremely fast—of the order of only a few tens of picoseconds [2]–[4] in the low-excitation limit—for electrons confined in QDs, spin dephasing is strongly reduced and the main remaining spin dephasing channel is contact hyperfine interaction with the nuclei [5]. The latter may be suppressed by using elaborate spin echo techniques [6]. Several alternative material systems like silicon [7] and graphene [8] have been suggested to overcome the problem of hyperfine interaction of electrons and nuclei. Recently, hole spins confined in QDs have been considered for quantum computing, as long hole spin dephasing times (SDTs) were observed in p-doped self-organized QDs [9, 10]. Additionally, efficient electrical tuning of the effective hole g factor in low-dimensional structures was predicted [11] and experimentally demonstrated [12]. In the present work, we demonstrate that very long hole SDTs can be observed in a two-dimensional hole system (2DHS), residing in a narrow quantum well (QW) in a GaAs/AlGaAs-based heterostructure. We show that the hole SDT strongly depends on the energy splitting between the quantized heavy-hole (HH) and light-hole (LH) states within the QW, which is controlled by the QW width. Specifically, the hole SDT can reach values above 70 ns at low temperatures and in small magnetic fields, which is about two orders of magnitude longer than previously reported results on wider GaAs QWs [12, 13]. In particular, these long SDTs allow us to use resonant spin amplification (RSA) techniques for precise measurements of the hole spin dynamics. The results shown here suggest that GaAs-based 2DHS may be a viable alternative to 2DES for quantum computing applications, which rely on electrostatically confined charged carriers.

2. Sample design and experimental methods

Our samples are single-side p-modulation-doped GaAs/Al_{0.3}Ga_{0.7}As QWs containing a 2DHS with relatively low hole density (see table 1). Previous investigations on similar samples

Table 1. Sample data. Density and mobility were determined from magneto-transport measurements at 1.3 K.

Sample	QW width (nm)	Hole density p (10^{11} cm^{-2})	Hole mobility μ ($10^5 \text{ cm}^2 \text{ Vs}^{-1}$)	Electron g factor $ g_e $
A	15	0.90	5.0	0.280 ± 0.005
B	9	1.03	3.6	0.133 ± 0.01
C	7.5	1.10	5.3	0.106 ± 0.01
D	4	1.10	0.13	0.266 ± 0.003

with relatively wide wells (15 and 10 nm) [12] showed that at these low densities, the optical recombination spectra at liquid-helium temperatures are governed by recombinations of neutral and positively charged excitons, i.e. bound excitonic complexes, consisting of one electron and two holes. Most importantly, even for the wider QWs, at very low temperatures, the resident holes become localized in potential fluctuations in the plane of the QW [12]. It was shown by Syperek *et al* [13] and Kugler *et al* [12] that localization of the holes is crucial for the observation of long hole SDTs of the order of a few hundred picoseconds in those samples. Here, we report on hole SDTs in narrow QWs, which are up to two orders of magnitude longer.

The structures are grown by molecular-beam epitaxy (MBE) on [001] substrates. Some characteristic properties are listed in table 1. The table clearly shows how the hole mobility is significantly reduced for the thinnest sample D, most likely due to the increased influence of monolayer fluctuations at the AlGaAs/GaAs interfaces on the hole wavefunction within the QW. For time-resolved Faraday rotation (TRFR) measurements, the samples are first glued onto a sapphire substrate with optically transparent glue and then the semiconductor substrate is removed by grinding and selective wet etching. All samples contain a short-period GaAs/AlGaAs superlattice, which serves as an etch stop, leaving only the MBE-grown layers. The TRFR and RSA measurements are performed in an optical cryostat with ^3He insert, allowing for sample temperatures below 400 mK and magnetic fields of up to 11.5 T. A pulsed Ti-sapphire laser system generating pulses with a length of 600 fs and a spectral width of 3–4 meV is used for the optical measurements. The repetition rate of the laser system is 82 MHz. The laser pulses are split into a circularly polarized pump beam and a linearly polarized probe beam by a beam splitter. A mechanical delay line is used to create a variable time delay between the pump and the probe. Both beams are focused to a diameter of about $80 \mu\text{m}$ on the sample using an achromat, resulting in an excitation density of about 2 W cm^{-2} . The center wavelength of the laser system is tuned to near resonance with the HH excitonic absorption lines of the samples. For this, photoluminescence spectra, taken using nonresonant excitation, are used to determine the transition energies of the neutral and charged excitons [12]. As the spectral linewidth of our laser system exceeds the energy splitting between the charged and neutral exciton transitions, both are excited by the pump laser pulses. In the TRFR and RSA experiments, the circularly polarized pump beam is generating electron-hole pairs in the QW, with spins aligned parallel or antiparallel to the beam direction, i.e. the QW normal. Due to the spectral linewidth of the laser, typically, both neutral and positively charged excitons are

excited near-resonantly. In the TRFR measurements, the spin polarization created perpendicular to the sample plane by the pump beam is probed by the time-delayed probe beam via the Faraday effect: the axis of linear polarization of the probe beam is tilted by a small angle³, which is proportional to the out-of-plane component of the spin polarization. This small angle is detected using an optical bridge. A lock-in scheme is used to increase sensitivity. In the RSA measurements, the Faraday rotation angle is measured for a fixed time delay as a function of an applied in-plane magnetic field. In our investigations, we exploit the strengths of both methods in order to explore the limits of hole spin dynamics in GaAs-based hole systems: in the TRFR experiments, one measures in the time domain via a pump–probe scheme, and the SDT as well as photocarrier lifetimes can be extracted from the measurements. The disadvantage of this method, however, is the limitation of the accessible time range by the travel length of the optical delay line. In our setup, this limits the time range to about 2 ns. This hinders an accurate determination of SDTs, which are significantly longer than the measurement range. The TRFR method encounters severe problems, if the SDT is even longer than the time interval between two subsequent laser pulses in the pulse train of the mode-locked laser (in our case about 12 ns). Fortunately, with the RSA technique [14], one can overcome these problems, since here one works with a fixed time delay between pump and probe pulses. The method is based on the resonant amplification of the Faraday signal, when integer multiples of the precession period of the spins in an in-plane magnetic field coincide with the inverse laser repetition rate, i.e. the time interval between subsequent pulses. However, here the extraction of the SDT from the experimental data is more involved, since it is hidden in the linewidths of the RSA maxima. As we will show below, in particular in our case, where electron and hole recombination and spin dynamics are interconnected, this is rather complex.

3. QW width dependence of hole SDT

We start our discussion by presenting TRFR experiments on our samples as a function of an in-plane magnetic field. As discussed above, in these experiments we are detecting the charge carrier and spin dynamics of photoexcited electrons and holes within a time range < 2 ns. This will allow us to gain important information on the interconnected electron and hole photocarrier and spin dynamics. Figure 1(a) shows a series of TRFR traces measured on sample D, the sample with the thinnest QW, at 1.2 K at three different magnetic fields, applied in the sample plane. The trace at $B = 0$ shows a single exponential decay of the Faraday signal after pulsed excitation with a decay time of $\tau_R = 70$ ps. Each of the other two traces, however, exhibits the superposition of two damped oscillations with markedly different frequencies and damping constants: a fast oscillation, which is observable within about the decay time of the $B = 0$ trace, only, and a slower oscillation, which, in the $B = 2$ T trace, exceeds the measurement range by far. The sum of two damped cosine functions is fitted to the data in order to extract the precession frequencies and decay constants. As a result, we can identify the high-frequency oscillations, which decay significantly faster, as the precession of photogenerated electrons, and the low-frequency oscillations as the precession of resident holes within the sample: in figure 1(b), the dispersions of the two precession frequencies are plotted, and the electron and hole g factors

³ For the samples investigated here, the maximum TRFR signal corresponds to a tilt angle of $70 \mu\text{rad}$.

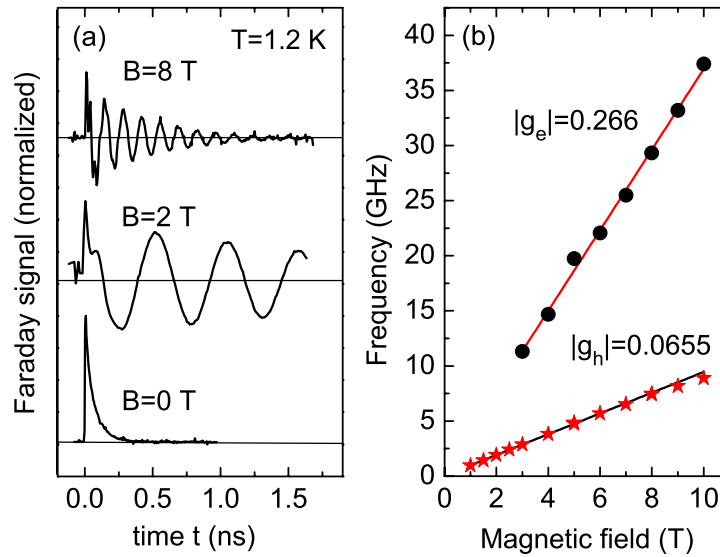


Figure 1. TRFR for different in-plane magnetic fields and dispersion of precession frequencies. (a) TRFR traces for sample D, measured at 1.2 K for different magnetic fields. (b) Dispersion of electron (black dots) and hole (red stars) spin precession determined for sample D from TRFR measurements. The solid lines are linear fits to the data.

are extracted from the data. The electron g factor $|g_e| = 0.266$ is in good agreement with values measured for QWs of similar widths [15, 16], while the hole g factor $|g_h| = 0.0655$ is close to zero, as was theoretically predicted for GaAs-based QWs grown along the [001] direction [17] and experimentally observed [12, 13, 18].

We note that the decay constant of the electron spin precession, $\tau_e = 75 \pm 5$ ps, remains almost constant throughout the investigated magnetic field range. This may be explained as follows: as our samples are p-doped, the optically oriented electron spins can only be observed during the photocarrier lifetime. The measured decay constant $\tau_e = 75 \pm 5$ ps therefore corresponds to the photocarrier lifetime in sample D, τ_R , as the electron SDT is typically longer. In stark contrast, the hole SDT decreases strongly with increasing magnetic field, as can be seen quite drastically from the $B = 2$ and 8 T traces in figure 1(a). However, at this stage the important question arises: why the trace at $B = 0$ in figure 1(a) obviously does not show a long lasting hole spin signal but a spin signal, which is governed by the photocarrier lifetime, only? We will address this question further below in the next section.

To investigate the strong magnetic field dependence of the hole SDT in more detail, we study the hole spin dynamics as a function of the applied in-plane magnetic field for samples with different QW widths. Figure 2(a) shows TRFR traces for all four samples, measured at 1.2 K with an applied in-plane magnetic field of 6 T. The traces have been scaled and vertically shifted in order to allow easy comparison of the hole spin dynamics. In all traces, again, a fast, rapidly decaying electron spin precession can be observed within the first ~ 100 ps. After photocarrier recombination, only larger-period hole spin precession is visible in the traces. Even though the applied in-plane magnetic field was fixed at 6 T, in all measurements shown in

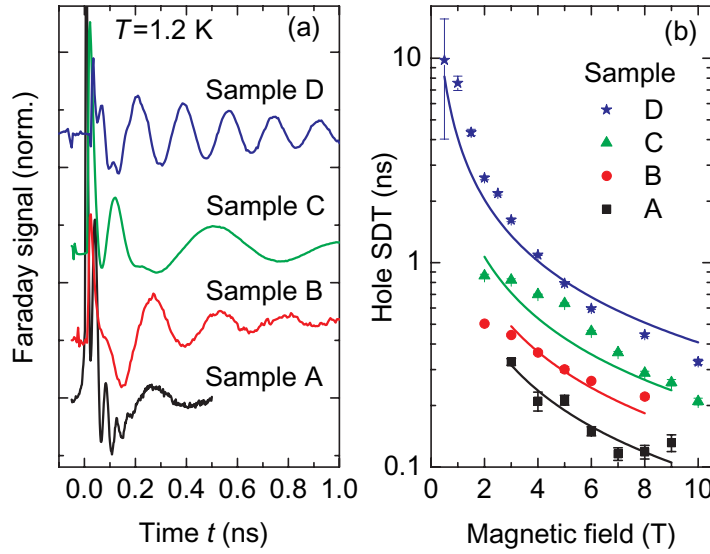


Figure 2. TRFR for different well widths and magnetic-field dependence of SDTs. (a) TRFR traces for samples A, B, C and D at 1.2 K. A 6 T in-plane magnetic field was applied during the measurements. (b) Hole SDTs for samples A, B, C and D as a function of magnetic field. The lines represent fits of a $1/B$ dependence.

figure 2(a), it can be seen clearly that both the electron and the hole spin precession frequencies are different for all four samples. For electrons, it is well known that the g factor in a QW depends on the QW width [15, 16]: due to the nonparabolicity of the conduction band in GaAs, the electron g factor changes depending on the electron energy above the conduction band edge, and in good approximation, the QW confinement energy of an electron will lead to a similar change of the g factor. Additionally, as the electron wavefunction has a nonvanishing amplitude within the QW barriers for thin GaAs QWs, the different electron g factors of pure GaAs and the barrier material are admixed. The values of the electron g factors extracted from TRFR measurements on all four samples are listed in table 1. We note that the sign of the g factor cannot be directly determined from TRFR measurements; however, by comparing our measured values to literature data [15, 16], we conclude that the electron g factor for samples A–C is negative, while it is positive for sample D.

On the other hand, the different hole g factors observed in the TRFR traces stem from the strong anisotropy of the hole g tensor, which was predicted by Winkler *et al* [17] for [001]-grown GaAs QWs. The effective hole g factor, g_h^* , measured in TRFR is given by the geometric sum of the in-plane (g_\perp) and out-of-plane (g_\parallel) components of the hole g tensor:

$$g_h^* = \sqrt{g_\perp^2 \cos^2 \alpha + g_\parallel^2 \sin^2 \alpha}. \quad (1)$$

Here, α is the tilt angle of the magnetic field with respect to the QW plane. While the in-plane component of the hole g factor is close to zero, $g_\perp \sim 0$, the out-of-plane component is $g_\parallel \sim -0.7$ for bulk GaAs [19]. Therefore, even small tilt angles α result in markedly different g_h^* .

In figure 2(b), the hole SDTs for all four samples are shown as a function of the applied magnetic field. Two effects are clearly visible here:

- (i) For all samples, the hole SDT decreases as the magnetic field is increased, following approximately a $1/B$ dependence.
- (ii) The hole SDT increases systematically as the QW width is reduced. It is smallest for sample A with the widest QW and largest for sample D with the thinnest QW.

The decrease of the hole SDT with magnetic field is a well-known phenomenon. It is believed to be caused by the inhomogeneity of the hole g factor, Δg_h^* , which leads to a dephasing of the hole spin polarization due to different precession frequencies. The dephasing rate due to this inhomogeneity is proportional to the applied magnetic field. Therefore, the hole SDT observed in the experiment, which is the SDT of an inhomogeneously broadened ensemble, T_2^* , is in first approximation given by [20]

$$T_2^* = \left(\frac{1}{T_2} + \frac{\Delta g_h^* \mu_B B}{\hbar} \right)^{-1} \quad (2)$$

if Δg_h^* is considered as the only source of inhomogeneity. Here, T_2 is the hole SDT in the absence of inhomogeneous broadening.

In order to understand the influence of the QW width on the hole SDT, we have to think about the main mechanisms for hole spin dephasing. The HH and LH states have different angular momenta, transitions between these states will therefore destroy hole spin orientation. In bulk GaAs, where HH and LH valence bands are degenerate at $k = 0$, any momentum scattering may lead to a transition between HH and LH states, which leads to hole SDTs of the order of the momentum scattering time [21]. In QWs, this degeneracy is lifted. However, for $k > 0$, the valence bands have a mixed HH/LH character [22], which may also lead to rapid hole spin dephasing due to scattering [23]. At low temperatures, resident holes in QWs may become localized at potential fluctuations within the QW, arising from QW thickness fluctuations due to monolayer steps at the interfaces, as well as from the granular distribution of the remote donors. Localization significantly reduces the hole quasi-momentum, keeping resident holes in HH states with $k \sim 0$. However, even for $k \sim 0$, there is a finite admixture of the LH states to the first HH subband [24]. The significant increase in hole SDT with decreasing QW width can therefore be attributed to an increased HH/LH splitting, which reduces the LH contribution to the HH ground state [25]. In a first approximation, if we consider infinitely high square-well potentials, this energy splitting ΔE is proportional to 1 over the well-width L squared:

$$\Delta E \sim \left(\frac{1}{m_{\text{LH}}} - \frac{1}{m_{\text{HH}}} \right) \frac{\hbar^2 \pi^2}{2L^2}. \quad (3)$$

For narrower QWs of finite potential height, however, the hole wavefunction penetrates strongly into the barrier, and the energy splitting is reduced again. For a QW width of 4 nm, the maximum HH/LH splitting was theoretically predicted and experimentally observed [26]. As figure 2(b) shows, we indeed observe the longest hole SDT in sample D with a well width of 4 nm. This suggests that the maximum HH/LH splitting in this sample is responsible for the long SDT. We will discuss the possible alternative mechanisms that might govern hole spin dephasing in the last section. Before that, we will in the following explore the limits of hole spin dephasing in our samples, starting with the optical initialization process of hole spin polarization in the next section.

4. Buildup of a resident hole spin polarization

In this section, we present experimental results and simulation data concerning the initialization process of the hole spin polarization, which was described by Syperek *et al* [13]: excitation of the sample with circularly polarized light will create spin-polarized electron–hole pairs according to the optical selection rules. If there is no significant electron spin dephasing during the photocarrier lifetime, the optically oriented electrons will recombine with holes which have matching spin orientation. Therefore, no spin polarization will remain within the sample after photocarrier recombination. This process is sketched schematically in the left panel of figure 3(c).

In an applied in-plane magnetic field, however, electrons and holes precess with different precession frequencies due to their strongly different g factors. Therefore, during their photocarrier lifetime, the electron spins will recombine with holes with arbitrary spin orientation. A part of the optically oriented holes may therefore remain within the sample after photocarrier recombination, as depicted in the right panel of figure 3(c).

To gain deeper insight into the combined dynamics of electron and hole spins and to analyze our experimental results quantitatively, we set up a model in the following. The combined dynamics of the electron and hole spins can be described via coupled differential equations for the electron (**e**) and hole (**h**) spin polarization vectors:

$$\frac{d\mathbf{e}}{dt} = -\frac{\mathbf{e}}{\tau_R} + \frac{g_e\mu_B}{\hbar}(\mathbf{B} \times \mathbf{e}), \quad (4)$$

$$\frac{d\mathbf{h}}{dt} = -\frac{\mathbf{h}}{\tau_h} + \frac{g_h\mu_B}{\hbar}(\mathbf{B} \times \mathbf{h}) + \frac{e_z\mathbf{z}}{\tau_R}. \quad (5)$$

In our model we assume the following: the electron spin polarization is reduced at a rate given by the photocarrier recombination and precesses about the in-plane magnetic field vector **B**. Electron spin dephasing has been neglected, as the electron SDT is expected to be significantly longer than the photocarrier recombination time τ_R , which is a reasonable assumption. The hole spin polarization is reduced at a rate given by the hole SDT τ_h and precesses about the in-plane magnetic field vector **B**. The last term in the second equation describes the change of hole spin polarization due to recombination of spin-polarized electrons with holes with matching spin orientation. Here, e_z is the z component of the electron spin polarization, and **z** is the unit vector along the growth direction. A similar approach was used by Yugova *et al* [27] to describe the initialization of a resident *electron* spin polarization in n-doped QWs. However, we need to include both electron and hole spin precession (the second term in equation (5)) in our differential equations to correctly model the RSA spectra that we observe, as described in the section below. In order to test the validity of this model, we compare it to experimental results. Figure 3(a) shows TRFR traces of sample C, taken at different applied in-plane fields. The sample has been carefully aligned so that the magnetic field is applied along the QW plane, i.e. $\alpha \sim 0$, resulting in a very low effective hole g factor g_h^* . For zero magnetic field, the TRFR signal decays monoexponentially with a decay constant of $\tau_R = 48$ ps, which reflects the photocarrier recombination time in sample C. For larger magnetic fields, pronounced electron spin precession can be observed during τ_R , and a significant nonzero TRFR signal remains after photocarrier recombination. This signal is due to the buildup of a resident hole spin polarization, as explained above. Its amplitude increases as the magnetic field is increased. Due to the very small hole g factor ($|g_h^*| < 0.005$), no hole spin precession is observed in the time range shown

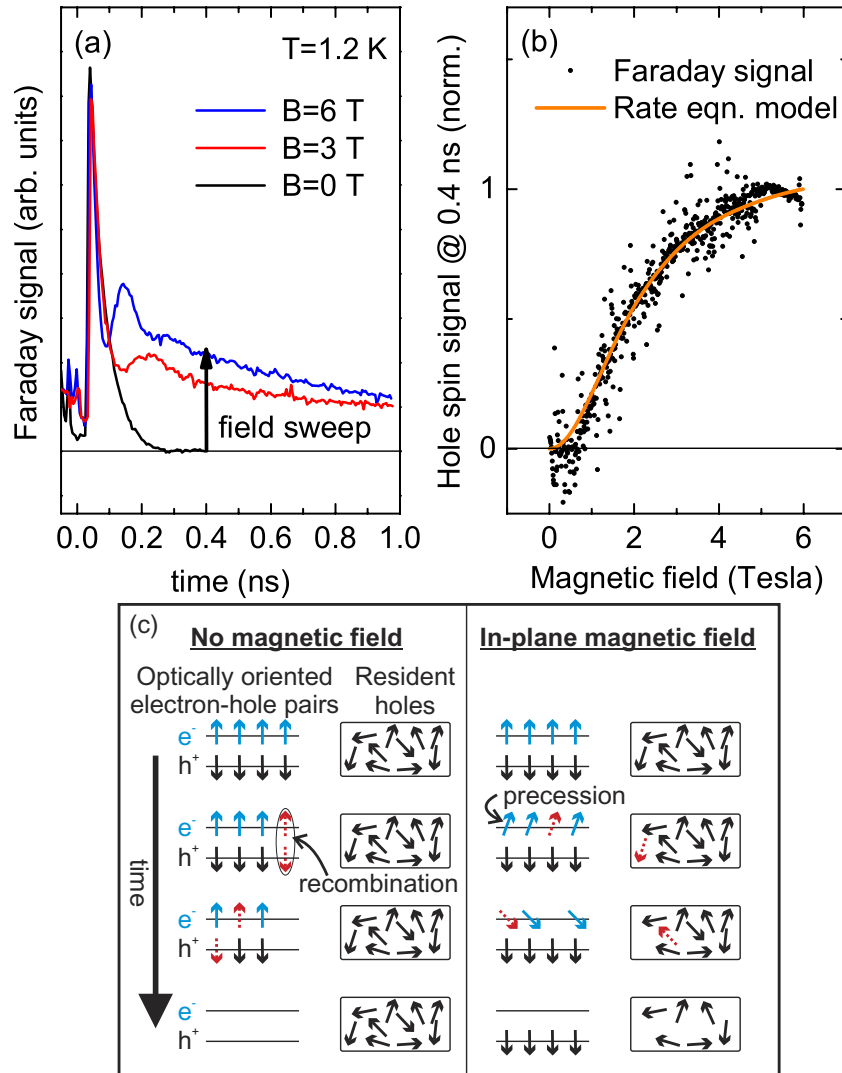


Figure 3. TRFR for different magnetic fields and buildup of hole spin polarization. (a) TRFR traces for sample C taken at 1.2 K for different magnetic fields. The arrow indicates the delay position for the magnetic field sweep shown in (b). (b) Kerr signal (black dots) as a function of magnetic field taken at a fixed time delay (400 ps) between pump and probe pulses. The orange line shows the buildup of the hole spin polarization as a function of magnetic field as calculated by the rate equations model. (c) Diagram of the combined spin and recombination dynamics with (right panel) and without (left panel) an applied magnetic field.

in figure 3(a). This allows us to study the buildup of the hole spin polarization in more detail using the following experimental technique: the TRFR signal is recorded for a fixed time delay between the pump and the probe. The time delay is chosen such that photocarrier recombination is complete, thereby leaving a TRFR signal from resident holes, only. The magnetic field is ramped up from zero in small increments. Figure 3(b) shows such a measurement for a time

delay of $\Delta t = 400$ ps. It is clearly visible how the resident hole spin polarization increases from zero to a maximum value, at which it saturates. This buildup can easily be modeled by the differential equation system described above. For this, we used the parameters extracted from the experiment: $|g_e| = 0.118$, $\tau_R = 48$ ps, $g_h^* = 0$. Hole spin dephasing was neglected to model the dataset. The resulting hole spin polarization as a function of magnetic field is shown as an orange solid line in figure 3(b). It is in excellent agreement with the experimental data. These measurements clearly demonstrate the crucial role of an applied in-plane magnetic field for transferring spin polarization from optically oriented photocarriers to resident holes.

5. RSA measurements of hole spin dynamics

To explore the hole SDT in very small applied magnetic fields and to approach the ultimate limit of hole SDT in our samples, we employ the RSA technique [14], which has been used in recent years, e.g., to study electron spin dynamics in n-doped bulk GaAs [28] and 2DES in CdTe-based QWs [29]. RSA is based on the constructive interference of spin polarizations created by subsequent laser pulses in a time-resolved Faraday or Kerr rotation measurement. The Faraday/Kerr signal is measured as a function of the in-plane magnetic field for a fixed time delay Δt , typically before the arrival of a pump pulse. If the optically oriented spins precess by an integer multiple of 2π within the time delay between two pump pulses, a maximum in the RSA signal is observed. The maxima are typically periodic in B . The SDT can be determined from the half-width of the maxima, and their spacing yields the g factor. In systems where the buildup of a resident spin polarization is governed by the interplay of electron and hole spin dynamics however, a more complex shape of the RSA signal can be expected, as will be demonstrated now.

Figure 4(a) shows a series of RSA traces, measured on sample D for different temperatures, compared to a numeric simulation, using the coupled differential equation model introduced above. The unusual ‘butterfly’ shape [27] of the RSA signals is clearly visible for the traces measured at 1.2 and 0.4 K. It is well reproduced by the simulation data. The RSA signal at 4.5 K is about 20 times weaker than the lower-temperature signals. In figure 4(b), a close-up of the first few RSA maxima measured at 0.4 K is compared to simulation data. There is no RSA maximum in the measurement and the simulation for $B = 0$, in contrast to typical RSA curves measured, e.g., in n-doped GaAs bulk. This is due to the peculiar transfer process that leads to a resident hole spin polarization. As demonstrated above, a finite in-plane magnetic field is necessary to create a resident hole spin polarization. The first maximum at finite field (numbered as $n = 1$) has a distinct shape resembling the derivative of a Lorentzian, while subsequent maxima resemble slightly asymmetrical Lorentzian curves. The amplitude of the maxima first increases with n , then decreases again for $n > 5$ (see figure 4(a)), while the full-width at half-maximum (FWHM) of the maxima increases. These features are clearly reproduced in the simulation. They can be explained as follows: for small magnetic fields, there is only a partial transfer of spin polarization to the resident holes, a process that saturates in our experiment for $B \sim 0.5$ T. For larger magnetic fields, the RSA FWHM increases due to inhomogeneous broadening, and the amplitudes of the RSA maxima decrease accordingly, until the RSA signal drops to the noise level at about 1.6 T in the low-temperature measurements. Interestingly, direct comparison of the RSA traces measured at 1.2 and 4.5 K reveals that the effective hole g factor $|g_h^*|$ decreases from 0.067 to 0.050 as the temperature is increased, in good agreement with previous observations [13].

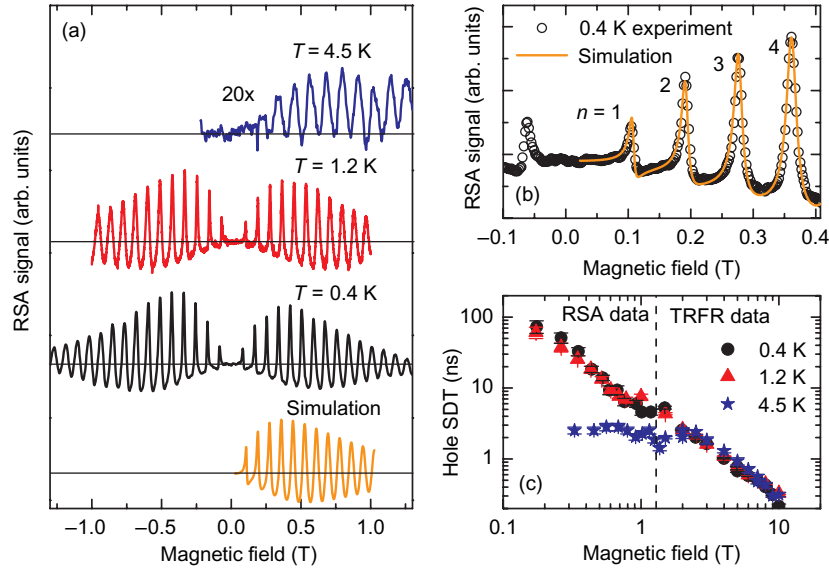


Figure 4. RSA measurements for different temperatures and magnetic field dependence of hole SDT for different temperatures. (a) RSA traces for sample D, measured at different temperatures, compared to simulation data. (b) RSA trace measured at 400 mK (open circles) compared to simulation data (orange line). (c) Hole SDT, determined from RSA data (left of the dotted line) and TRFR data (right of the dotted line), for different temperatures as a function of magnetic field.

Figure 4(c) shows the hole spin lifetime in sample D for different temperatures. The data for low magnetic fields (below 1 T) have been determined from RSA data, the data for higher magnetic fields (above 2 T) were determined from TRFR data. Interestingly, while at 4.5 K, the hole SDT saturates at about 2.5 ns, even in low magnetic fields, it follows a clear $1/B$ -like dependence down to very small magnetic fields in the data measured at lower temperatures, yielding values of $T_2^* = 74 \pm 15$ ns at 0.4 K and $T_2^* = 61 \pm 11$ ns at 1.2 K (determined from the FWHM of the second RSA maximum at $B \sim 0.2$ T)⁴. The power of the RSA technique will be evident if one compares the hole SDT measured at higher fields: in the magnetic field range where TRFR measurements yield precise results of the hole SDT, very little difference is observed in the values of the hole SDT in the temperature range from 0.4 to 4.5 K. From the magnetic field dependence of the hole SDT, we may infer that the T_2 time of the hole spin is above 80 ns at temperatures below 500 mK. By fitting equation (2) to the magnetic field dependence, we determine the g factor inhomogeneity $\Delta g_h^* = 0.003 \pm 0.0002$.

Furthermore, the RSA measurements allow for a precise determination of the effective hole g factor, which is given by $g_h^* = 2\pi f_{\text{rep}} \hbar / (\mu_B \Delta B)$. Here, f_{rep} is the laser pulse repetition frequency, and ΔB is the magnetic field spacing between two adjacent RSA maxima. Figure 5 shows RSA traces measured at 1.2 K for different angles α of the magnetic field with respect to the QW plane⁵. The geometry is sketched in the figure. The spacing of the maxima is

⁴ The hole SDT is proportional to $1/(\delta B^2)$ for all RSA maxima with $n > 1$. Here, δB is the FWHM of the RSA maximum.

⁵ The sample tilt angle is determined by measuring the angle of the pump beam that is reflected from the sample surface.

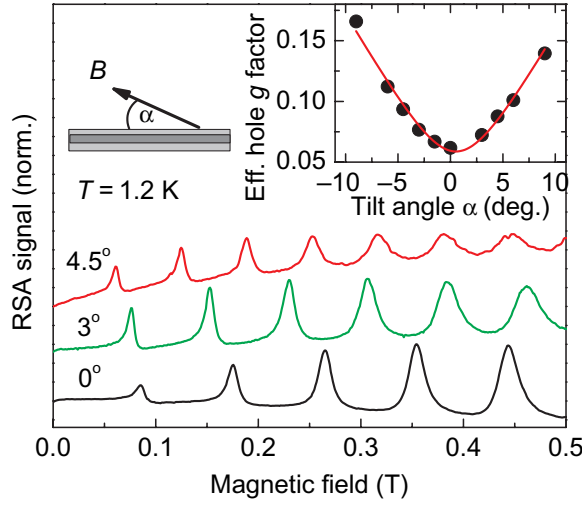


Figure 5. RSA measurements for different tilt angles of the magnetic field and effective hole g factor dependence on tilt angle. RSA traces for sample D, measured at 1.2 K, for different tilt angles of the sample with respect to the external magnetic field. The inset shows the effective hole g factor $|g_h^*|$ determined from the spacing of the RSA maxima (black dots). The solid red line represents a fit of the results using equation (1).

significantly reduced as the magnetic field acquires an out-of-plane component. The effective hole g factor $|g_h^*|$ is extracted from this spacing, and the results are shown in the inset. The increase of the effective hole g factor with the magnetic field angle is due to an admixture of the out-of-plane component of the hole g factor $g_{||}$, which is typically far larger than the in-plane hole g factor g_{\perp} , as described by equation (1). By fitting the results with this equation as indicated by the solid red line in the inset, both components of the hole g factor at 1.2 K can be determined with high accuracy: $|g_{\perp}| = 0.059 \pm 0.003$, $|g_{||}| = 0.89 \pm 0.03$.

Finally, we will discuss the possible mechanisms that might limit the hole SDT in our experiments. At low temperatures and in low magnetic fields, where the g factor inhomogeneity may be neglected, in principle, several mechanisms might be thought of to pose the ultimate limit of the hole SDT. In the following, we will discuss their dependence on QW thickness in the light of our experiments, and identify the most important mechanism.

- (i) *Thermal activation of holes from localized states.* The thermal activation rate is proportional to the Boltzmann factor $\exp((-E_{\text{Loc}})/(k_B T))$. For localization of holes at QW monolayer thickness fluctuations, the localization energy increases drastically with a reduction of the QW width [24]. For a 15 nm wide QW, it is less than 1 meV, while for a 4 nm wide QW, it is about 10 meV. In both cases, the localization energy is significantly larger than the thermal energy, even at the highest measurement temperature used in our experiments: $E_{\text{th}}(4.2 \text{ K}) = 0.36 \text{ meV}$. This indicates that thermal activation of holes out of localized states is unlikely to affect hole spin dynamics at liquid-helium temperatures and below.
- (ii) *Dipole–dipole interaction of holes with nuclei.* Even though the p-like symmetry of valence band states does not allow for a *contact* hyperfine interaction between holes and nuclei, a coupling is possible via dipolar interactions [10, 30]. The nuclei act as slowly varying

random magnetic fields, which lead to both single spin decoherence and ensemble spin dephasing. The variance of the nuclear magnetic field is inversely proportional to the number of nuclei that interact with the hole confined in the QD, which may be estimated from the dot size. We note that the typical size of QDs that arise from monolayer thickness fluctuations in high-quality GaAs QWs (TF-QDs) is of the order of $(100 \text{ nm})^2$, significantly larger than that of self-assembled InAs dots. One may estimate that a hole interacts with about 10^6 nuclei in TF-QDs or electrostatically controlled QDs [6], whereas self-assembled dots typically contain only about 5×10^4 nuclei [10]. Additionally, the random magnetic fields associated with the nuclei may be suppressed very efficiently by a small, external magnetic field applied within the sample plane [30]. It is shown above that the application of an in-plane magnetic field is necessary for the transfer of spin polarization of optically oriented carriers to resident holes. Therefore, we may neglect hole spin decoherence due to interaction with nuclei as the dominant process limiting the hole SDT in our experiment.

- (iii) *Finite admixture of LH states to HH states.* Even localized holes have a finite quasi-momentum, which is given by the hole temperature. In an analogon to the Elliott–Yafet [31, 32] mechanism, which has been studied in detail for electrons, any momentum scattering may therefore destroy hole spin orientation via hole spin flip due to the slight admixture of LH states to HH states. This admixture is of the order of 2% for thin QWs [24]. It increases significantly for wider QWs due to the decreasing HH/LH energy splitting, increasing the probability of a hole spin flip during momentum scattering [33]. The spin dephasing rate is directly proportional to the momentum scattering rate, which in turn increases with the sample temperature.

Summarizing this part: we believe that the increased HH/LH splitting is responsible for the increased hole SDT with decreasing QW width in our experiments. On the other hand, the still finite LH admixture to the HH ground state and, hence, the possibility of spin-flip scattering of Elliott–Yafet type sets the limit for hole spin dephasing.

6. Conclusions

In conclusion, we have investigated hole spin dynamics in 2DHSs embedded in QWs of different widths. Due to the increased energy splitting between HH and LH states, the hole SDT increases drastically in narrow QWs. The inhomogeneity Δg_h^* of the hole g factor leads to a typical $1/B$ -like dependence of the hole SDT. In order to transfer spin polarization from the optically oriented photocarriers to the 2DHS, a finite magnetic field is necessary. This also leads to a peculiar shape of RSA signals measured on 2DHS. From these RSA signals, the hole SDT in low magnetic fields can be extracted. It is of the order of 80 ns, one order of magnitude larger than for electrons in QDs of similar dimensions defined in 2DES by external gates [6]. This makes 2DHS an interesting material system for scalable quantum computing devices based on electrostatically confined charge carriers. Additionally, RSA measurements in tilted magnetic fields allow us to accurately determine both the in-plane and out-of plane components of the hole g factor.

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