Spin mode locking in quantum dots revisited

S. Varwig*1, A. Greilich1, D. R. Yakovlev1,2, and M. Bayer1,2

1 Experimentelle Physik 2, Technische Universität Dortmund, 44221 Dortmund, Germany
2 Ioffe Physical-Technical Institute, Russian Academy of Sciences, 194021 St. Petersburg, Russia

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*Corresponding author: e-mail steffen.varwig@tu-dortmund.de, Phone: +49 231 755 3536, Fax: +49 231 755 3674

A comprehensive overview of coherent spin manipulation in (In,Ga)As quantum dots, singly charged with resident electrons or holes, is given, from orientation to rotation of the spins. These operations are performed by excitation of the charged exciton complex with laser pulses. The specifics of the approach is performing the manipulations on dot ensembles, potentially giving robustness to the coherent spin dynamics. In particular, we focus on the spin mode-locking regime, in which the precession of the spins about an external magnetic field is synchronized with the periodic pulsed laser excitation initiating the operations. The periodic excitation protocol introduces a frequency comb through which detrimental effects of the inhomogeneities in the spin system can be avoided.

1 Introduction

Spin excitations in semiconductors have been considered as promising carriers of information that might turn out to be superior to charges for specific tasks [1]. This has contributed to the emergence of research areas such as spin electronics or quantum information [2]. A major promise of semiconductors is the possible connectivity to standard electronics for which sophisticated state-of-the-art fabrication tools exist. These tools will hardly be abandoned unless a novel technology emerges, which clearly outperforms the established ones that are making steady progress.

The attribute of potential out-performance has been assigned to quantum information technology with facets such as unprecedented computation power or secure information transfer [3]. For that purpose spins have to be operated as quantum bits which can hold information for a sufficiently long time [1]. Due to extensive experience with semiconductors in higher-dimensional systems this requires the localization of spins in order to enhance the spin coherence. Localization can be achieved by the three-dimensional (3D) confinement provided by quantum dots [4]. Another possibility, not in the center of interest for this paper, is the localization by defects [3]. Furthermore, methods for controlled manipulation of the spin excitations need to be developed during the time in which the coherence is maintained [5]. Within this limit the number of possible operations should be maximized, for which optical or microwave pulses may be employed. Control by microwaves will also not be discussed here, even though some assessment will be given in the following.

Anticipating a couple of results to be described in more detail below, we want to mention a few general features of quantum dot coherent spin manipulation derived from the results of other research groups as well as our own activities. If spins are to be operated through laser pulses exciting an exciton complex, quantum dot spins benefit from the large oscillator strength of the related optical transition, allowing efficient manipulation by lasers emitting pulses of picosecond (ps) duration or even shorter [6]. This is considerably faster than addressing spins with microwaves, where the manipulation times cannot be shorter than nanoseconds [7]. On the other hand, microwave pulses, being generated electrically, can be controlled much better than laser pulses, with respect to their properties such as duration, intensity, etc., allowing most likely more complex manipulations than by optical means [8].

These tools need to be contrasted with the coherence times. For defect-bound spins coherence times up
to milliseconds (ms) have been demonstrated, even at room temperature, so that microwave techniques may be sufficient [9]. For quantum dot spins, on the other hand, the spin coherence is limited to microseconds at cryogenic temperatures, unless sophisticated protocols for decoupling from the spin environment are implemented; see below. For higher temperatures a fast drop of the coherence times is observed so that the spin excitations become less attractive for quantum information [10]. While cryogenic technology becomes more and more easy to handle, the microsecond limitation might prove microwaves to be too slow to obtain a sufficient number of desired operations. This could be improved by extending the spin coherence through decoupling from the nuclear spin bath, but it is not clear whether a manipulation can be implemented on top of the multi-pulse decoupling protocols [11]. If at all possible, this might require mostly the application of laser pulses for operating on the spins. In that way, GHz operation frequencies may also be possible, becoming compatible and competitive with current technologies for information processing and transfer.

Here we want to give a comprehensive description of our recent works on electron or hole spins confined in quantum dot ensembles, studied by pulsed laser spectroscopy. The dot ensemble aspect is a specific feature of the activities, while most other groups working in the field concentrate on single-dot activities. Ensemble activities have recently gained renewed interest, because the ensemble might allow a collective enhancement of interactions between quantum bits, as is required for entanglement for example [12].

The paper is organized according to the individual steps required for a coherent control of carrier spins. After presenting details of the dot structures and the experimental setups in Section 2, Section 3 describes the optical orientation of electron and hole spins. Spin mode locking is introduced in Section 4. This phenomenon has allowed accurate measurement of spin coherence times (Section 5) for which we focus here on their modification by applied magnetic fields. In Section 6 we discuss the coherent rotation of spins by laser pulses. The paper is concluded in Section 7 by a summary also giving perspectives for future studies. Generally we focus on features complementary to results presented in previous publications.

2 Experiment

The studied samples contain multiple layers of self-assembled (In,Ga)As/GaAs quantum dots, grown by molecular beam epitaxy on (001)-oriented GaAs substrates. The dot layers were separated from each other by 80-nm-wide barriers such that direct electronic coupling by tunneling does not occur. To estimate the dot density, a sample of uncovered-surface quantum dots was prepared under identical conditions. An atomic force micrograph of this sample is shown in Fig. 1, from which the dot density in each sheet can be calculated to be around $10^{10}$ cm$^{-2}$. Furthermore, the dot sizes are about 30 nm in diameter and 7–8 nm in height. Overgrowth, as required to obtain optically active structures, leads to intermixing of the dot and barrier materials, reducing the In contents in the dots. Also, a change of the dot dimensions might be expected, which is however not observed: high resolution transmission electron micrographs of an as-grown sample indicate very similar dot dimensions as compared to the uncapped sample.

The ground-state emission of the as-grown samples is located at around 1.0–1.1 μm at cryogenic temperatures. Figure 1 shows photoluminescence spectra of an as-grown and an annealed sample. In order to shift the emission into the excitation wavelength range of Ti:sapphire lasers and the detection sensitivity range of Si-based diodes, thermal annealing was performed for 30 s at temperatures around 950 °C, leading to a band-gap increase through out-diffusion of indium atoms from the dots into the barriers. As a consequence the ground-state emission occurs at around 900 nm with slight variations from sample to sample. The concomitant flattening of the confinement potential is evidenced from the reduction of the energy splitting between different confined shell emission lines, which decreases from about 60 meV to about 25 meV, as seen in Fig. 1. This splitting is mostly determined by the lateral confinement, which may become more shallow not only by the band-gap increase, but also by an increase of the dot diameter. The main size increase is, on the other hand, expected predominantly along the growth direction where the In gradient is largest; any changes of the dot height or diameter, whose increases reduce confinement and lower quantization energies, are, however, overpowered by the In diffusion in their effect on the band gap [13]. One also sees that the full width at half maximum (FWHM) of the emission lines changes drastically by the thermal annealing. Being about 50 meV for the as-grown sample, it is reduced to $\sim 10$ meV in the annealed
sample, demonstrating good ensemble homogeneity. Further optical properties of these dots can be found in Ref. [14].

Two sets of samples were studied, different with respect to the resident carriers in the dots:

1. In the first set an average occupation of each dot by a single electron was targeted. For that purpose, the structures were n-modulation doped by introducing sheets of Si dopants 20 nm below each of the 20 dot layers. The Si-dopant density in each sheet was chosen to be roughly equal to the dot density. From the spin dynamics studies we could estimate that this resulted in about half of the quantum dots being singly charged. We note that some of the dopant electrons may contribute to compensating p-type doping from carbon impurities.

2. In order to study hole spins, we exploited the background doping from the just-mentioned resident carbon acceptors that are characteristic for GaAs-based material. To exploit this doping nominally undoped samples were fabricated, in this case containing only 10 layers of quantum dots, also separated by 80 nm barriers. Here the spin dynamics studies reveal as well that about half the dots in the ensemble are loaded by a single hole.

The samples were immersed in the liquid helium insert of cryostats equipped with superconducting coils for generating magnetic fields. Dominantly single-coil systems for field strengths up to 7 or 10 T were used. Also, a vector magnet was employed in the experiments, consisting of three split coils oriented orthogonally to each other. By coordinated adjustment of the current through each coil, the total magnetic field strength \( B \) can be fixed at a certain value up to 3 T, while its orientation is variable over the whole sphere. Consequently, the optical alignment does not have to be changed when the orientation between sample and field is rotated for measurements depending on that orientation. Particularly in spatially inhomogeneous systems, it is demanding to address the same sample position in a series of measurements. Furthermore, a rotation of the sample in a single split-coil system leads to the fact that the optical axis is no longer normal to the sample plane. Irrespective of problems with optical adjustment and light scattering, in polarization-sensitive studies, such as pump–probe, the polarization of the beams inside the sample will be considerably different from circular or linear for large tilt angles between magnetic field and sample growth directions, as determined by Fresnel’s equations. Using a vector magnet, the optical alignment remains unchanged, while the field direction is adapted as demanded.

The spin dynamics is addressed by time-resolved pump–probe techniques [15]. In the simplest version of the experiment a circularly polarized pump pulse is used to generate spin polarization along the heterostructure growth direction, parallel to the optical axis, which is taken as \( z \)-axis. To obtain detailed insight into the spin dynamics, typically a magnetic field normal to the optical axis (for fixed orientation taken along the \( x \)-axis) is applied due to which the spins precess about the field. The precession of the ensemble magnetization is monitored by a linearly polarized probe pulse train, split off from the same laser. For this beam either the rotation of the polarization plane (the Faraday rotation) or the ellipticity of polarization is measured after passage through the sample. Technically, the pump and probe pulse trains are taken from a Ti:sapphire laser emitting 2 ps pulses at a rate of \( \sim 76 \) MHz, corresponding to about 13.2 ns pulse separation. For varying the repetition rate, a pulse picker can be installed by which the pulse separation is increased by at least an order of magnitude. The delay between pump and probe is adjusted by sending the pump beam over a mechanical delay line where a retroreflector’s position can be tuned with high accuracy leading to possible time resolutions below 50 fs, sufficiently accurate for our spin dynamics studies. The probe’s polarization detection was performed by a pair of balanced photodiodes, interfaced with a lock-in amplifier.

In an extended version of the experiment, the pump pulse train could be split into two independent pump pulse trains with adjustable temporal separations between them, obtained by another mechanical delay line. Also, the circular polarization of the pulses in each train could be adjusted. However, even in this setting the central wavelength of all pulses is the same. To become more flexible in this respect, a setup was also realized in which two Ti:sapphire lasers were integrated; see Fig. 2. One of the lasers is used as master oscillator for the other, slave, oscillator, from which a synchronization of the two lasers with an accuracy of 100 fs can be obtained. The wavelengths of the two lasers are independently tunable; the other pulse parameters are identical to those mentioned above. Using this configuration, the wavelengths of pump and probe can be varied relative to each other. Furthermore, experiments with pump and probe resonant but with an additional laser as required for optical spin rotation also become possible.

3 Optical spin orientation

3.1 Spin polarization by periodic pump pulse application Before an optical excitation starts the spins in the quantum dot ensemble are randomly oriented. The spin orientation can be achieved by applying circularly polarized pump pulses, which transfer their angular momentum to the spins, exploiting optical selection rules according to the conduction- and valence-band structure. Generally all corresponding protocols rely on this principle, irrespective of details of the implementation. We focus here on the fast orientation using the laser systems mentioned above. For any application it would most likely be required that the total time for this process is below a nanosecond, in order to be competitive with existing information technologies.

In principle, the laser oscillators could also have been adjusted such that they emit pulses with about 100 fs duration. The pulses are Fourier-transform-limited so that their spectral width scales inversely with the pulse duration. From \( g \)-factor studies it is known that the \( g \)-factors of electrons and holes vary considerably with the optical transition energy across the photoluminescence spectra shown in Fig. 1. To avoid excitation of a strongly inhomogeneous spin ensemble, we
therefore used our lasers not in the femtosecond configuration, but in the ps configuration, as described in the previous section. The 2 ps pulses correspond to a spectral width of 1 meV. The spectrum of such pulses is shown in Fig. 1 by the blue trace.

We excited the quantum dots resonantly with the ground-state transition, leading to the formation of negatively or positively charged excitons, depending on the residual charge, in the s-shell of the quantum dots. We note that spin orientation is also possible for non-resonant excitation into an excited quantum dot shell, for example. However, in this case the spin initialization becomes more complex due to the relaxation of carriers into their ground states [16]. Moreover, the spin mode-locking signal, which is described below, is considerably weaker than for the resonant case [17].

The optically injected exciton complexes have long coherence times, which may be even only radiatively limited, but in any case they are much longer than the laser duration [18]. Therefore, a coherent description of the excitation process has to be applied, in which a laser pulse is characterized by its area defined by

$$\Theta = \frac{2}{\hbar} \int d \cdot E(t) \, dt, \tag{1}$$

where $d$ is the matrix element for the transition from the resident carrier to the correspondingly charged exciton and $E(t)$ is the electric field of the laser [15]. The scalar product of these two quantities is integrated over the pulse duration, from which after division by Planck’s constant $\hbar$ the dimensionless quantity $\Theta$ is obtained. For $\Theta$ there exists the demonstrative geometrical interpretation of it being the polar angle on the Bloch sphere, of which the south (north) pole represents the resident carrier (the exciton complex). Before excitation the Bloch vector points towards the south pole. By applying a laser pulse, the Bloch vector is rotated by the angle $\Theta$ towards the north pole, where it becomes located if the pulse area is $\pi$.

Without orientation by the pulse, the spins of electrons and holes can be arbitrarily oriented. To obtain a splitting into two levels, a magnetic field is applied, here with fixed orientation along the $x$-axis. Taking the heterostructure growth direction $z$ as the spin quantization axis with the basis states $|\uparrow\rangle$ and $|\downarrow\rangle$ pointing along (“spin up”) or against (“spin down”) the $z$-axis, an arbitrary resident electron spin state can be written as

$$\alpha|\uparrow\rangle + \beta|\downarrow\rangle, \tag{2}$$

where for normalization reasons $|\alpha|^2 + |\beta|^2 = 1$. In thermodynamic equilibrium the spins are oriented parallel or antiparallel to the field, so that $\alpha = \pm \beta$ and $|\alpha| = |\beta| = 1/\sqrt{2}$. 

Figure 2 Scheme of one of the used experimental arrangements for pump–probe studies. In this case two synchronized Ti:sapphire oscillators, each pumped by a frequency-doubled Nd:YAG laser, were implemented, to vary, for example, the pump and probe beam photon energies independent of each other (as shown) or to introduce an additional beam in degenerate pump–probe studies. The pump is sent over a mechanical delay line with a retroreflector to vary the delay between pump and probe. The power of both laser pulse trains could be adjusted by filters; clean linear polarization was achieved by sending them through Glan prisms. For the pump, circular polarization was arranged by inserting a $\lambda/4$ retarder into the optical path. The polarization of this beam could be modulated between $\sigma^+$ and $\sigma^-$ by a photo-elastic modulator (PEM), operated typically at a few tens of kHz. Both beams were sent almost parallel along the optical axis onto the sample (with a slight angle in between, to avoid scattered laser light from the pump) and focused with lenses. The sample is inserted into a 3D-vector magnet for independent variation of the field orientation relative to the sample growth direction which in all cases can then be parallel to the optical axis. After passage through the sample the probe beam is sent through a $\lambda/4$ retarder for measuring ellipticity or Faraday rotation, respectively. Then the probe is split by a polarizing beam splitter cube, with subsequently each beam being sent onto one of the diodes of a balanced photodetector that is connected to a lock-in amplifier.
Similarly, a resident hole spin state is given by

\[ \gamma|\uparrow\rangle + \delta|\downarrow\rangle. \tag{3} \]

with an equivalent definition of the spin vectors \(|\uparrow\rangle\) and \(|\downarrow\rangle\) as for the electron and the corresponding normalization condition \(|\gamma|^2 + |\delta|^2 = 1\). We note that for simplicity we use the term “hole spin,” even though it would be more appropriate to call it a total angular momentum state composed of the orbital and the spin angular momenta. If the ground state were a pure heavy hole, the two states would have \(z\)-component \(J_{h,z} = \pm 3/2\) of the total angular momentum, whereas the electron is a pure spin state with \(S_{e,z} = \pm 1/2\).

If we assume without loss of generality that the pump pulse is \(\sigma^+\) polarized, then, due to selection rules, it would lead in an undoped quantum dot to the excitation of an electron–hole pair with spin configuration \(|\downarrow\uparrow\rangle\), to an extent that is determined by the pulse area \(\Theta\) only, without further restriction. The resulting superposition state of the unexcited semiconductor vacuum and the excited exciton complex is given by

\[ \cos\left(\frac{\Theta}{2}\right)|0\rangle - i\sin\left(\frac{\Theta}{2}\right)|\downarrow\uparrow\rangle. \tag{4} \]

Subsequently, the carrier spins precess about the magnetic field with their respective \(g\)-factors. For uncharged dots electron and hole spins may become coupled by the exchange interaction while this interaction is zero for charged dots due to the spin-singlet character of one carrier species after optical injection of an electron–hole pair. If the exchange interaction exceeds the Zeeman energies of electron and hole spins, the total exciton angular momentum precesses, while for the reversed relation electron and hole spins precess independently. In any case the induced spin polarization lives only until the radiative decay of the exciton occurs.

If the hole were to have pure heavy hole character, the \(g\)-factor would be zero or at least close to zero [19], so that no appreciable precession would occur. This is because the dots still resemble planar systems with a much larger dot diameter as compared to the height. We find, however, clear signatures of hole precession in the experimental data (see below). The assignment is based on the corresponding \(g\)-factors on the order of 0.15 in the quantum dot plane. This indicates that confinement and strain in the quantum dots lead to valence-band mixing, due to which the hole ground state also contains light hole components with \(J_{h,z} = \pm 1/2\). Figure 3 shows the distribution of the hole \(g\)-factor in the quantum dot plane, for which the magnetic field orientation was changed by the vector magnet. Considerable variations with orientation are observed, from which apparently a twofold symmetry pattern can be deduced with the main axes along \([110]\) and \([1\bar{1}0]\).

The values range from 0.08 to 0.23. On the contrary, for the electron spins an almost symmetrical pattern is found with an average value of 0.535 at this particular transition energy. A closer inspection shows an anisotropy on the order of 2 \% of the mean value. Further details of the \(g\)-factor tensors can be found in Ref. [20].

In presence of resident carriers the excitation process becomes more elaborate due to the influence of Pauli blocking. The spin component of the resident carrier that is parallel to the optically injected electron (for \(n\)-doped dots) or the optically injected hole (for \(p\)-doped dots) blocks the excitation process. Only for the counter-oriented component is excitation allowed, so that for a dot structure with a resident electron, the following electron–trion superposition state is generated:

\[ \alpha \cos\left(\frac{\Theta}{2}\right)|\uparrow\rangle + \beta|\downarrow\rangle - i\alpha \sin\left(\frac{\Theta}{2}\right)|\downarrow\uparrow\rangle. \tag{5} \]

The unexcited electron components will give rise to a net electron spin contribution with strength \(|\beta|^2 - |\alpha|^2 \cos^2(\Theta/2)\). From the trion there is no electron contribution, as the two contained electrons form a singlet, though there is a signal from the hole spin [15].

A corresponding Faraday rotation trace of precessing spins recorded on \(n\)-doped quantum dots is shown in Fig. 4 for \(B = 3\) T. The impact of the hole spin can be seen only at short delays below 1 ns after application of the pump pulse at time zero. There also an exciton contribution from undoped quantum dots shows up, which vanishes on similar time scales as the hole signature. For the quantum dots under study the radiative decay time was measured to be about 0.5 ns for both neutral and charged excitons, explaining the disappearance of corresponding contributions of optically injected carriers after these times [15].

However, oscillations in the Faraday rotation signal are observed much longer than during the presence of excitonic complexes, evidencing that spin coherence has been initiated for the resident electrons. The precession of the component that has remained unexcited due to Pauli blocking continues beyond the trion-component decay. There are two reasons...
for that spin polarization: (i) assuming a zero hole $g$-factor, the hole-in-trion spin would remain fixed while the electrons are in a spin-singlet state corresponding to zero total spin. The non-excited component of the electron spin precesses until stochastic radiative recombination of the trion occurs. The contained electron would be returned with an orientation parallel to that of the hole spin. Relative to the already precessing spin’s orientation, the parallel component of the returned electron would amplify the resident electron spin polarization. Repeating this enhancement of spin orientation through our periodic pulsed excitation protocol, the electron spin becomes fully polarized [21]. (ii) Taking the hole spin precession into account, the exciton can recombine with either of the two electrons in the spin-singlet state, so that after periodic excitation — and even more so in an ensemble — the trion contribution becomes randomized and only the residual spin contributes. Periodically repeated excitation reduces the component that can be excited into a trion so that also with hole spin precession electron spin polarization builds up [15].

For long delays an exponential decay of the signal is observed, which beyond 1 T becomes accelerated with increasing field strength. This decay arises from the variation of the resident carrier $g$-factors in the optically excited quantum dot ensemble, which is still sizable despite of the narrow-band excitation by ps pulses. At lower fields below 0.5 T variations of the nuclear field are also influential. The resulting spread of the precession frequency leads to the carrier spins in the ensemble running out of phase, after their initial orientation. This dephasing is typically described by the characteristic time $T_{2}^*$ of an exponential damping factor.

To obtain insight into the damping dynamics, we have varied the magnetic field orientation in the quantum dot plane. The resulting dephasing times as function of the field direction determined from corresponding Faraday rotation traces are shown in Fig. 5 for both electron and hole spins. Obviously, there is no strong dependence of the electron spin dephasing time on the field direction, while for the hole spins the variation is considerable. The behavior very much reflects the variation of the carrier $g$-factors, as shown in Fig. 3. This supports that the spin dephasing is indeed mostly determined by $g$-factor variations. In particular, the dephasing time distribution of the hole spins resembles the anisotropy of the corresponding hole $g$-factor. Notably, the dephasing time is shortest for the directions of small $g$-factors, showing that the associated variations are more prominent than the effect of variations along the large $g$-factor direction, for which one might expect a faster dephasing due to the faster spin dynamics. However, the relative variations for large $g$-factors are apparently less important than for small $g$-factors at the applied field of 1 T.

In the coherent regime it should be possible to drive the transition from the electron to the trion periodically with increasing excitation power (entering the pulse area $\Theta$) in form of Rabi oscillations. From the discussion above it is obvious that the most efficient spin orientation is obtained...
for pulses with $\Theta = \pi$, as then the polarization-reducing component by the oppositely oriented electron, expressed by the second term in $|\beta|^2 - |\alpha|^2 \cos^2(\Theta/2)$, is minimized. Vice versa, the trion excitation, important for increasing the spin of the Pauli-blocked electron, is maximized.

Figure 6a shows ellipticity traces recorded for different pump powers. The signal amplitude rises starting from low excitation powers until at a certain power level a maximum is reached, after which a drop of the signal strength is observed. Panel (b) gives the ellipticity amplitude, determined from exponentially damped fits to the data with harmonic functions, versus pump power. The data are indicative for a strongly damped Rabi oscillation-like behavior, where a maximum is reached but beyond the corresponding power no distinct further oscillations with minima and maxima can be observed. The strong damping is expected from the variation of the transition matrix elements in the inhomogeneous optically excited dot ensemble. These inhomogeneities translate directly into variations of the pulse area $\Theta$ for Rabi oscillations.

The electric field in $\Theta$ is proportional to the square root of the pump power, so that the top axis in Fig. 6b shows the pulse area corresponding to the powers given at the bottom axis. The conversion factor between the two quantities has been chosen such that the power at which the signal reaches maximum amplitude is equal to $\pi$. For complications with this assignment, see Ref. [22]. Here the geometrical interpretation of $\Theta = \pi$ is that the Bloch vector is inverted on the Bloch sphere, with the south pole being the unexcited resident electron where the Bloch vector resides before pulse application, and the north pole being the trion. The red line shows a fit to the data proportional to $\sin^2(\Theta/2)$ corroborating the interpretation in terms of Rabi oscillations. Coherent rotations of the Bloch vector up to an angle of $\pi$ can be achieved in the dot ensemble, whereas for larger angles the effects of inhomogeneities become too strong. For many applications $\pi$ rotations are, however, sufficient.

In case of spin orientation of the holes the argument is quite similar [23]. The superposition state generated by optical excitation is here given by

$$\gamma|\uparrow\rangle + \delta \cos \left(\frac{\Theta}{2}\right)|\downarrow\rangle - i \delta \sin \left(\frac{\Theta}{2}\right)|\downarrow\uparrow\rangle,$$

where now the spin-up hole remains unexcited for $\sigma^+$-polarized light. Thereby this component gets pumped by the repeated laser pulse application and the subsequent decay of the positively charged exciton component. Corresponding ellipticity traces are shown in Fig. 7a. The hole spin precession is recognizable from the low-frequency oscillation, which is, however, at positive delays, superimposed by exciton and electron spin precession signals. A pure hole spin precession signal is observed at negative delays, whose origin is the spin mode locking, to be described in the next section, showing the same precession frequency as after the pump. The hole spin precession amplitude is strongly damped due to the inhomogeneity of the hole in-plane $g$-factor inducing a fast post-pump dephasing and rephasing only shortly before the pump pulse arrives. Therefore, only up to one spin precession period is observed on both sides of zero delay [24].

The amplitude of the mode-locked hole spin precession signal scales similarly with power as the positive delay signal. This amplitude as function of pump power (bottom axis) is shown in Fig. 7b. Also, here a strongly damped Rabi-oscillation behavior is observed, where after reaching maximum spin polarization a decay towards a constant level is obtained. For fitting the data we have used Eq. (32) of Ref. [22], derived by considering the specifics of the mode-locked signal formation. This analysis shows that maximum amplitude does not occur for pulse area $\pi$ as for the positive delay signal, but for an area of about $1.5\pi$. 

\[ \gamma|\uparrow\rangle + \delta \cos \left(\frac{\Theta}{2}\right)|\downarrow\rangle - i \delta \sin \left(\frac{\Theta}{2}\right)|\downarrow\uparrow\rangle, \]
times. Using the most efficient pulses with area long as the pulse separation is well below the spin relaxation leads to fast generation of spin polarization close to unity, as theoretical calculations show that the repeated periodic excitation [21]. With the used lasers this corresponds to a time of 0.7 ns. For the first pump we selected a pulse area of \( \pi \). For the second pulse, also with area \( \pi \), we chose the same circular polarization and applied it in one case at a maximum of the ellipticity signal, so that the spin polarization is maximum, from which we would ideally expect no further amplification of spin coherence: at the moment when pump 2 comes in, the resident electron spins point upwards, so that absorption of the \( \sigma^- \)-polarized pump 2 should be Pauli-blocked. In the other case we applied the second pump in a minimum of the ellipticity signal, so that in the ideal case the second pulse should annihilate the coherence generated by pump 1. Pauli blocking is not effective in this case as the electron spins are pointing downwards, so that a \( \pi \) pulse should be able to convert the Bloch vector completely into a trion.

The corresponding results are shown in Fig. 8. Panel (a) shows the case when pump 2 comes in at maximum ellipticity signal generated by pump 1. The two top traces show the effect of pump 1 and pump 2 alone. The comparison shows that the two spin polarizations induced by one of the pulses without action of the other one are very similar, as expected from their identical parameters. The lower trace shows the signal when both pumps are acting, in comparison to the calculated sum of the two upper signals (labeled as sum signal). The latter would be expected if the two pumps would address independent spin ensembles. The recorded signal is smaller, as expected because the pumps address nominally the same spins. On the other hand, the signal is stronger than if only one pump is used. In particular, one sees that the second pump injects exciton complexes whose signatures have disappeared only about 1 ns after its incidence. If one compares the signals at about 2 ns delay and later, still a net signal from newly generated spin coherence remains. This shows that the mentioned inhomogeneities cannot be neglected. For

3.2 Spin polarization by pump pulse doublets
Theoretical calculations show that the repeated periodic excitation of singly charged quantum dots, as described before, leads to fast generation of spin polarization close to unity, as long as the pulse separation is well below the spin relaxation times. Using the most efficient pulses with area \( \pi \), more than 99% polarization can be reached after application of a dozen pulses [21]. With the used lasers this corresponds to a time of slightly more than 150 ns, about an order of magnitude below the transverse spin coherence time \( T_2^* \); see below. Using lasers with shorter resonator length, the pulse repetition rate can be easily increased into the GHz range, so that spin orientation times of 10 ns should be feasible.

Once polarization has been maximized by repeated excitation we would in the ideal case not expect further enhancement of the spin polarization by application of additional pulses. However, the results presented so far have shown that in an ensemble variations of the dipole transition matrix element are important. Furthermore, after spin orientation variations of the precession frequency due to g-factor dispersion lead to signal dephasing. Also, the variation of the laser power in the Gaussian-shaped laser spot leads to variations of the pump pulse area for dots with different locations in the spot area. These power variations could only be avoided if pulses with flat-top spatial profiles were used. They could be formed by spatial light modulators, for example.

Therefore, we have tested the impact of an additional laser pulse applied at a time when on one hand exciton complex decay has largely occurred, while on the other hand the effects of dephasing are not too destructive for the signal formed by the resident carriers. We concentrated on the n-doped quantum dot sample and chose delays of about 0.6–0.7 ns. For the first pump we selected a pulse area of \( \pi \). For the second pulse, also with area \( \pi \), we chose the same circular polarization and applied it in one case at a maximum of the ellipticity signal, so that the spin polarization is maximum, from which we would ideally expect no further amplification of spin coherence: at the moment when pump 2 comes in, the resident electron spins point upwards, so that absorption of the \( \sigma^- \)-polarized pump 2 should be Pauli-blocked. In the other case we applied the second pump in a minimum of the ellipticity signal, so that in the ideal case the second pulse should annihilate the coherence generated by pump 1. Pauli blocking is not effective in this case as the electron spins are pointing downwards, so that a \( \pi \) pulse should be able to convert the Bloch vector completely into a trion.

The corresponding results are shown in Fig. 8. Panel (a) shows the case when pump 2 comes in at maximum ellipticity signal generated by pump 1. The two top traces show the effect of pump 1 and pump 2 alone. The comparison shows that the two spin polarizations induced by one of the pulses without action of the other one are very similar, as expected from their identical parameters. The lower trace shows the signal when both pumps are acting, in comparison to the calculated sum of the two upper signals (labeled as sum signal). The latter would be expected if the two pumps would address independent spin ensembles. The recorded signal is smaller, as expected because the pumps address nominally the same spins. On the other hand, the signal is stronger than if only one pump is used. In particular, one sees that the second pump injects exciton complexes whose signatures have disappeared only about 1 ns after its incidence. If one compares the signals at about 2 ns delay and later, still a net signal from newly generated spin coherence remains. This shows that the mentioned inhomogeneities cannot be neglected.

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Figure 7 Panel (a): Ellipticity trace recorded at \( B = 1 \text{T} \) as function of the delay between pump and probe, measured on quantum dots with resident holes. The pulse separation is \( T_R = 132 \text{ns} \). The low-frequency oscillation from hole spin precession is superimposed by strong oscillations, reflecting the electron-in-trion spin precession in charged dots as well as the exciton spin precession in non-charged dots within the ensemble. The hole is the only component contributing to the mode-locked signal at negative delays before the pump. The inset shows a series of ellipticity traces in the mode-locking delay range recorded for varying pump powers. Panel (b): Amplitude of the mode-locked hole spin signal at negative delays as function of the applied laser pump power (bottom axis), determined from the data in the inset of the upper panel. The rising part of this dependence is fitted by a theoretical formula for the strength of the mode-locking signal, from which the conversion of pulse power into pulse area at the top axis is obtained.
example, due to dephasing a fraction of quantum dots can be excited again, and in these dots the spin polarization becomes re-established. Furthermore, dots located in the periphery of the first pump spot are exposed to an excitation density below π, so that the second pump is expected to enhance their polarization.

In Fig. 8b the corresponding data are shown for the case when pump 2 comes in at an ellipticity minimum. Here the joint action of the two pumps (bottom trace) clearly leads to a weaker signal, whose strength is even below the strength of the signal that is obtained by adding the signals induced by pump 1 or pump 2 alone (the top traces), being in antiphase relative to each other. This latter fact highlights the impact of dephasing on the signals, as without dephasing the two signals should ideally exactly compensate each other.

4 Spin mode locking In the previous section we have mentioned the signal at negative delays. This coherent spin precession signal appears after the spin ensemble, originally oriented by a pump pulse, has completely dephased and before a subsequent pump pulse can re-orient the spins. In low magnetic fields the dephasing at latest occurs a few nanoseconds after spin initialization. As discussed in detail in the literature, this signal arises from synchronization of the precessional motion of the carrier spins with the periodic laser excitation. This has been demonstrated to be possible for both electron and hole spins in correspondingly doped quantum dots [24, 25].

Here only the very basic features shall be discussed again: due to the finite spectral width of the laser of 1 meV, a quantum dot ensemble having different ground state optical transition energies within the laser bandwidth is excited. For electrons, for example, the g-factor is determined largely by the band gap, so that a number of spins with continuously varying g-factors also become oriented [26, 27]. For hole spins the dependence of the g-factor on the band gap is more complex; still a distribution of spins with varying g-factors is polarized. Within these distributions, particular sets of spins are distinguished. For these spins the precession frequencies \( \omega_i \) of electrons or holes \( (i = e, h) \) are multiples of the laser repetition rate. This means that those spins perform an integer number of precession revolutions about the magnetic field within the pulse separation \( T_R \). This synchronization condition thus reads

\[
\omega_i = \frac{g_i \mu_B B}{\hbar} = N_i \frac{2\pi}{T_R}, \quad i = e, h. \tag{7}
\]

Here the \( g_i \) are the electron and hole g-factors along the magnetic field, respectively. The \( N_i \) are integers, which for \( B = 1 \text{T} \) are about 100 for the electrons, while they are only 20–30 for the holes. For this phase synchronization we introduced the term “mode locking” of the spins by the periodic excitation, being well known for the longitudinal optical modes in laser resonators. The mode locking is achieved there for example by exploiting the optical nonlinearity provided by the optical Kerr effect and leads to pulsed light emission. At first the term “mode locking” therefore seems not very well substantiated due to the seemingly missing nonlinearity; fulfillment of the condition (7) appears to be not initiated, but on the contrary rather arbitrary. However, a detailed inspection of the spin mode locking shows that there are a number of feedback mechanisms from the optical excitation which influence the synchronization and will be presented in the following.

The consideration so far suggests that only a small fraction of spins – those which exactly fulfill the synchronization condition – contributes to the mode-locked signal. The
distribution of quantum dot transition energies convoluted with the laser pulse represents the density of spin states that in principle are eligible for the spin coherent response and determine the signal at positive delay times. In this smooth density of states the spins fulfilling Eq. (7) represent sharp, $\delta$-function-like spikes. All spins located in between these spikes are expected not to contribute to the signal at negative delays. However, the situation is more complex because of two factors:

(1) For the electrons the hyperfine interaction with the lattice nuclei leads to a focusing of the spins which are not synchronized right at the start of laser excitation onto the locked modes. Due to this interaction flip-flops between electron and nuclear spins can occur by which after a couple of such processes a nuclear magnetic field may arise, which adds to the external magnetic field such that phase synchronization can occur [28]. The illumination is important for the focusing, as the mismatch between electron and nuclear spin splitting would prevent flip-flops. However, the optical excitation lifts this restriction during pulse action. When the phase synchronization is reached, the electron spins are oriented such that at pulse arrival Pauli blocking prevents further excitation so that the nuclear spin configuration is stabilized by the mode locking, maintaining it therefore. While this works well for electrons, for the holes the hyperfine interaction is reduced by about an order of magnitude so that the nuclear contribution by mode locking is small. This difference becomes pretty obvious from Figs. 4 and 7a, where the hole spin mode-locked signal is clearly much weaker than that of the electron. For the electrons ultimately all spins that are optically excited contribute to mode locking as becomes evident from the negative and positive delay signals having comparable strengths, with variations depending in particular due to the different measurement methods.

(2) Also, a second mechanism adds to the nonlinear feedback of the illumination onto phase synchronization. A detailed theoretical analysis of the mode-locking phenomenon shows that the amplitude depends strongly on the features of the optical excitation protocol. Details of this analysis are given in Refs. [28, 29]. In short, the density of mode-locked states resembles sharp spikes only for low pump power areas $\Theta$ and large pump pulse separations $T_R$ at a given spin coherence time. When increasing the pump pulse area the mode widths increase significantly, so that – roughly speaking – a larger fraction of quantum dots is synchronized with the laser excitation from the beginning until at high excitation powers beyond $\pi$ dephasing effects become important.

4.1 Single-mode precession The importance of the experimental parameters in the mode-locking regime shall be illustrated in the following by comparing Faraday rotation traces recorded at two specific magnetic field strengths for varying pump powers. For this illustration we also tried to minimize the effects of dephasing due to $g$-factor inhomogeneity. Generally the number of precession modes within a spin ensemble excited by a pulse of spectral width $\Delta \Omega$ is obtained by dividing $\Delta \Omega$ through the separation of adjacent mode-locked frequencies [30]:

$$M = \frac{\Delta \Omega}{2\pi/T_R} = \frac{\Delta g, \mu_B BT_R}{\hbar}.$$  

Here we used that $\Delta \Omega$ is translated into a distribution of precession frequencies $\Delta \omega_i$, according to the underlying distribution of carrier $g$-factors $\Delta g_i$. One should note here that this equation is a simplified form as it disregards the nuclear magnetic field, mostly due to random fluctuations (in our case with a strength of about 10 mT), which prevent lowering the total magnetic field acting on the spins below this value, even when the external field is switched off.

Based on this equation, one may seek experimental parameters in which the optically oriented spins are pushed into a single phase-synchronized precession mode, in which they all add up constructively to a macroscopic magnetization. The options to reduce the number of modes towards unity are obvious: reduction of the pulse separation and the magnetic field, while the dispersion of the $g$-factors may be changed only by turning to samples designed in another way. Our studies of quantum dots with significantly different structural and compositional properties revealed, however, quite similar dispersion of the electron $g$-factor with transition energy. One could also try to use longer pulses, as long as they are still Fourier-limited, so that their spectral width is reduced accordingly, resulting in excitation of a more homogeneous spin ensemble. However, in that way one would lose efficiency in spin polarization; see Ref. [31].

Corresponding studies show that for the quantum dots under study a pure single-mode operation is possible for hole spins, but not for electron spins due to the effective nuclear magnetic field acting on them [24, 32]. Still, one can end up in a regime where the precessional motion in the ensemble is dominated by either a single or a pair of mode-locked modes. Switching between these two regimes can be obtained by fine tuning the magnetic field strength through which the narrow spectrum of excited precession frequencies is shifted relative to the frequency comb of the phase-synchronized modes [30].

To end up in the single precession mode regime we reduced the pulse separation, 13.2 ns as emitted from the laser oscillators, to half of this value, by splitting the beam and rejoining the two parts after one of them made a detour of 6.6 ns. The magnetic field was reduced to the range of 0.3–0.6 T. Smaller fields have no further effect due to the effective nuclear magnetic field, see above. In this low-field regime the effects of dephasing due to $g$-factor inhomogeneities are also minimized, because the translation of these inhomogeneities into precession frequency dispersion is minimized.

Corresponding measurements are shown in Fig. 9 for the n-doped quantum dot sample. Here the magnetic field strength was varied around 0.5 T. In case of a dominant
Figure 9  Faraday rotation traces recorded with the n-doped sample at two different magnetic field strengths of 530 and 540 mT. In addition the pump power was varied, increasing from 1 to 5 and further to 10 mW. To approach the single precession mode regime, the separation between pump pulses was also halved, from 13.2 to 6.6 ns. We also want to attract here attention to the fact that, while at low pump powers the mode-locked signal is weaker than the signal at positive delays, for high pump powers the mode-locked signal dominates. This result is strongly influenced by choosing Faraday rotation as measurement tool, for which the spin distribution has to be asymmetrical relative to the central laser wavelength in order to detect a signal at all. This asymmetry is apparently fulfilled in a better way for the mode-locked spins than for the entirety of spins excited by the pump.

single mode ensemble precession, we would expect a harmonic oscillation that is almost undamped because of the suppressed dephasing. This is the case that we observe at 540 mT for a weak pump excitation power of 1 mW. When reducing the magnetic field to 530 mT, the response of the spin ensemble is strongly changed. Now two phase-synchronized modes contribute about equally, which leads to a node in the signal right between the two pump pulses at a delay of 3.3 ns, as around this time the contributions of the two modes to the Faraday rotation signal compensate each other.

This behavior at 530 mT is obviously maintained when increasing the pump power, as shown by the top traces in Fig. 9. However, the time at which the node occurs shifts slightly to smaller delay times. This might have several reasons. For example, the optical excitation induces nuclear magnetic field changes to keep the electron spins in mode locking. This nuclear field might redistribute the spins such that they no longer contribute with equal strengths as seen for low excitation powers. The increased excitation power might also lead to stronger excitation of additional phase-synchronized side modes beyond the two central modes, whose contributions might be asymmetrical because of a different number of quantum dots which can contribute to these additional features.

The effects of more intense excitation become much more pronounced when moving back to 540 mT, where at low powers a pure undamped sinusoidal oscillation was found.

Upon increasing the power to 5 mW and further to 10 mW damping emerges until the appearance of the signal is very similar to the case in which two dominant spin precession modes contribute, as the signal minimum is located around 3 ns. This could be explained again by a nuclear shift of the optically excited spin density such that two modes in the frequency comb contribute. Furthermore, the more efficient excitation of additional phase-synchronized modes could also become relevant.

4.2 Bursts of mode-locked spin coherence

Application of pump pulse doublets was discussed in connection to spin initialization. However, use of such pump doublets can also have other implications. Figure 10 shows ellipticity (panel a) and Faraday rotation traces (panel b) recorded on a p-doped quantum dot sample, revealing the dynamics of either resident hole or electron spins, respectively, see Ref. [24]. Now the delay between the two pump pulses was increased to \( \sim 2.2 \) ns, such that dephasing of the signals initiated by the first pump has occurred before the second one comes in. Beyond the pump-induced signals further strong signals occur at times where there is no pump to orient spins on the background of dephased spins. These signals observed for both electron and hole spins occur at about \( \sim 2.2 \) and 4.4 ns, showing a clear correspondence with the pump pulse separation.

Indeed, the origin of these signals is somewhat similar to that of the mode-locking signal, where the phase synchronization requires spins to perform an integer number
of precession revolutions within the separation between the pulses in the pump pulse train taken directly from the laser, in our case 13.2 ns. Only then can they contribute to the mode-locking signal [29]. Now an additional period is superimposed onto this excitation protocol, which requires the spins to perform in addition an integer number of revolutions within the pulse separation time in the pump doublet. From this we can derive the commensurability condition, that the separation $T_D$ between the two pulses within a doublet is a rational number times the separation $T_R$ between two pump pulse doublets:

$$T_D = \frac{m}{n} T_R$$  \hspace{1cm} (9)

with $m$ and $n$ being positive integers. Then the spins come into phase again around every delay which is a multiple of $T_D$, leading to a burst in the spin coherent signal amplitude. The rise and decay times of the signal around the delay times of maximum signal are identical to those for the signal at the times when one of the pumps hits the sample, because they are determined by dephasing of the same spins. For our particular case in Fig. 10, Eq. (9) is fulfilled for the integers $m = 1$ and $n = 6$.

5 Measurement of spin coherent signal With the knowledge that we have established now about the mode-locked spin coherent signal, we also want to discuss an important point showing that considerable care needs to be exercised in interpreting data recorded through measuring either ellipticity or Faraday rotation. We will first consider a homogeneous spin system. The ellipticity is proportional to the absorption of the system, so that the signal becomes maximum when the exciting laser is in resonance with the monitored optical transition, i.e., for a pulsed laser when the central wavelength coincides with the transition wavelength. Vice versa, the Faraday rotation signal is proportional to the derivative of the absorption, so that the signal strength should be minimum if the laser is on-resonance.

So far we have described data recorded for on-resonance conditions. Therefore, it is not surprising that the ellipticity signal rendered a strong signal. For the Faraday rotation studies it seems contradictory to the expectation, though. To expect a signal here, an asymmetry is needed so that the laser no longer coincides with the zero of the Faraday rotation spectrum. Here the nuclear focusing process can also become decisive, as it leads to a shift of the resonance of the spin-split resonance [32]. The study of an inhomogeneous ensemble is even more important here, because the optical excitation might occur asymptomatically in the density of states. Obviously, the degree of this asymmetry determines the signal strength [33].

To obtain insight into this, we have performed Faraday rotation studies with degenerate pump and probe, where, in one case, the laser photon energy is tuned to the maximum of the photoluminescence spectrum, which itself has a rather symmetrical shape for the ground-state emission. In the other case the energy was tuned to the high-energy flank of this spectrum, where the signal amplitudes are found to be higher than on the low-energy flank. This might arise from the existence of doubly charged quantum dots on the low-energy side, which contribute to the photoluminescence but not to the spin coherent signal due to the spin-singlet character of the two electrons in the quantum dot ground shell.

Figure 11a shows ellipticity traces for different magnetic fields with pump and probe degenerate on the photoluminescence maximum. The photon energy was kept fixed in this series, as the diamagnetic shift of the ground-state emission is well below $50 \mu$eV in the studied field range, so that a laser tuning is not necessary. Aside from the increasing precession frequency and the much stronger signal for positive delays, the ellipticity amplitude evolves rather smoothly with increasing magnetic field, demonstrating a slight drop except for some rise at around 3 T, while for the mode-locked signal first a slight drop occurs, followed by a small increase; see Fig. 11b. But, generally, the signal strengths show no
more smoothly with a continuous drop, showing, however, a small. Tuning the probe to lower or higher energy relative to this position, the mode-locked signal decreases continuously with increasing field and reduces the spin orientation efficiency.

Figure 12a, on the other hand, shows the corresponding traces for Faraday rotation measurements and the resulting signal amplitudes versus magnetic field. The signal for positive delays is considerably weaker than in ellipticity, which is maximum, corresponding to maximum asymmetry, it then drops almost to zero at around 4–5 T, in particular for the mode-locked signal. This means that at this field the spin distribution has to be nearly symmetrical relative to the center excitation wavelength. This asymmetry is apparently re-established on increasing the field strength further to $B = 7$ T, before the amplitude drops again, partly also because of the reduced spin initialization efficiency (see above).

Figure 12, on the other hand, shows the corresponding traces for Faraday rotation measurements and the resulting signal amplitudes versus magnetic field. The signal for positive delays is considerably weaker than in ellipticity, which might be expected from the consideration of signal strengths given above. For negative delays, on the other hand, the signal amplitudes are comparable. Even more striking, the dependence of the signals on magnetic field is much more pronounced in Faraday rotation, for both positive and negative delays. After the pump, first a drop occurs up to about 3 T, before the signal rises again with variations by a factor of up to 3. The mode-locked signal, on the other hand, varies more smoothly with a continuous drop, showing, however, a slight rise at 3 T. The drop is as well much stronger than the change observed in ellipticity.

The detection of Faraday rotation is surprising at first sight, as described above, because we excite in the maximum of the symmetrical photoluminescence emission. However, an asymmetry might become relevant due to an uneven distribution of singly charged quantum dots for which the electron-to-trion transition can occur across the laser spectrum. Even if that were not the case, an asymmetry might build up due to the nuclear focusing effect, as shown in Ref. [32]. Varying the magnetic field, the shift of the spin precession mode spectrum relative to the phase-synchronized frequency comb will certainly change the asymmetry required for detecting a signal, leading to the observed considerable variations.

From this consideration we expect that the Faraday rotation signal will behave differently with respect to its strength (for which a natural absolute scale is hard to define upon changing the photon energies) and its variation with magnetic field (which can be compared for different settings), if pump and probe are tuned away from the photoluminescence maximum. Tuning to the high (or low) energy flanks should inherently lead to an asymmetry different from that of the maximum emission excitation. As a result, the field-induced asymmetry change is also expected to be different.

This is confirmed by degenerate Faraday rotation measurements where the photon energy is tuned to the energy where the ellipticity signal becomes strongest, because the fraction of singly charged dots is apparently maximum there. The Faraday rotation signals and the resulting amplitude field dependence are shown in Fig. 13. The behavior for the positive and negative delay signals, similar to each other, is somewhat reminiscent of the behavior in Fig. 12. However, the nonlinear variation of the signal amplitude with magnetic field is much more pronounced. While at 1 T the signal is maximum, corresponding to maximum asymmetry, it then drops almost to zero at around 4–5 T, in particular for the mode-locked signal. This means that at this field the spin distribution has to be nearly symmetrical relative to the center excitation wavelength. This asymmetry is apparently re-established on increasing the field strength further to $B = 7$ T, before the amplitude drops again, partly also because of the reduced spin initialization efficiency (see above).

To obtain more insight, we have used a two-laser oscillator experiment, which allows one to tune the photon energy of pump and probe independently. Corresponding studies are shown in Fig. 14, where the pump energy was fixed at 1.3955 eV, while the energy of the probe beam was tuned around this energy. From comparison of the spectra, it is obvious that the amplitudes of the signal right after the pump and the signal of mode-locked spins before the pump show opposite trends at the chosen 2 T field strength. In particular, for the degenerate case the mode-locking signal is maximum, while the pump-initiated signal at positive delays is quite small. Tuning the probe to lower or higher energy relative to this position, the mode-locked signal decreases continuously with some peculiarity at about 0.5 meV separation with...
signal strengths close to zero. In contrast, the positive delay signal increases for this detuning, reaching maxima at the two peculiarity points before dropping smoothly to zero with increasing energy separation of the probe from the pump.

The variation of the Faraday rotation amplitude at positive delays resembles very much the behavior that one would expect from the initial consideration which was shown to be valid for 4 T [33]. An asymmetry of the probe relative to the pump excitation is needed to observe a signal and the variation with probe energy corresponds to a good approximation to the dispersive shape squared. On the other hand, the antiphase behavior of the negative delay signal indicates that in the degenerate case the probe meets a particularly asymmetrical distribution of phase-synchronized precession modes at 2 T. This overview of exemplary results demonstrates that one has to analyze the chosen experimental situation carefully if one wants to extract data from the measured amplitudes, in particular for Faraday rotation experiments. On the other hand, for measurements of precession frequencies or also dephasing times the consideration of these parameters is less important.

5.1 Spin coherence times In Refs. [25, 28] we have discussed in detail that the mode-locking phenomenon allows one to obtain accurate values for the spin coherence time $T_2$, even though the measurements are performed on spin ensembles subject to dephasing. This is because of the selectivity of the mode-locked frequency comb that serves as a filter for the spin precessional motions. In detail, the precessional motions of the spins that pass this frequency filter come into phase again when approaching the next pump pulse, as they all perform an even number of spin revolutions within the pulse separation time and their contributions add up constructively.

However, this can happen only as long as the coherence of the individual spins in each quantum dot is maintained. As soon as elastic, or even more detrimental, inelastic spin...
scattering, changing the phase of the spin precession, sets in, the mode locking becomes suppressed. Therefore, one can measure the transverse spin relaxation or spin coherence time $T_2$, which is at least at finite magnetic fields considerably shorter than the longitudinal relaxation time $T_1$, by monitoring the mode-locking amplitude as function of the pulse separation. Once this separation becomes in the range of the $T_2$ times the amplitude is expected to drop. This is the behavior that is also found in experiments. Figure 15 shows the ellipticity amplitude as function of the separation (black solid circles) for the n-doped sample. A drop of signal is observed when the pulse separation is increased from 0.1 to 0.5 $\mu$s. The black solid line shows a fit to the data by an exponential function, from which we determine the decay time to be 0.48 $\mu$s for the chosen experimental parameters ($B_L = 1$ T). Note that this value can vary under different experimental conditions (e.g., pump power), but it is always in the range of $1 \mu$s at temperatures below 10 K [25].

In Ref. [34] it was observed that the application of a small longitudinal field along the optical axis in addition to the transverse field leads to a drastic increase of the correlation times in hole spin measurements. These times are very close to the spin coherence times of the hole spins, measured independently from hole spin mode locking. The tentative explanation of this increase was the “calming down” of the otherwise random nuclear spin bath which prolongs the hole spin coherence in spite of the weak interaction with nuclei. Therefore, we wanted to explore whether such a longitudinal field component can increase the spin coherence time of electrons. The applied field strength along the optical axis was $B_L = 150$ mT, about an order of magnitude stronger than the random nuclear field acting on the electron spins in these quantum dots. If relevant this should be sufficient to reduce the nuclear spin fluctuations. Figure 15 also shows the dependence of the mode-locking amplitude for this situation (red open circles) together with the corresponding fit (red dashed line). Clearly, the data basically coincide with those for zero longitudinal field so that the fit also gives the same decay time.

Surprisingly, we thus find that the additional longitudinal field does not lead to an increase of the spin polarization decay time as measured by mode locking. The origin of this behavior is not fully understood yet and needs to be studied further. First, the longitudinal field does not change the precession of the electron spins about the transverse field. It also does not change the frequency comb. Therefore, the question arises of whether the mode-locking experiment gives the “true” spin coherence time in this experimental configuration. The limitation of the previously discussed spin coherence times is the result of fluctuations in the nuclear spin system which is frozen up to times in the $\mu$s range, but then starts to melt. Decisive for changing the phase of the spin precession about the transverse field are the nuclear fluctuations along this direction and they might not be affected by the longitudinal field. However, further studies are required to work out the underlying mechanisms.

Also, the coherence time of the hole spins has been determined through the mode-locking technique. Doing so, we find $T_2$ times in the microsecond range, very much comparable to the values found for electron spins. This is a surprising finding, since the interaction with the lattice nuclei has been identified as the limitation for the electrons, in particular the altering of the nuclear spin configuration on this time scale. For the holes, the hyperfine interaction was found to be about an order of magnitude weaker, as evidenced also from the relative strength of the corresponding signals at negative delays. Therefore, longer coherence times were expected for the hole spins, which could not be confirmed [35, 36].

For completeness, we also show the temperature dependence of the electron and hole spin coherence times. The data are shown in Fig. 16. The electron spin coherence time is constant up to almost 20 K. Above this value a fast drop into the ns range occurs over a 20 K range. Considerably
more sensitive is the hole spin coherence, which is constant only up to about 8 K and then a steep drop to nanoseconds occurs.

These findings are on first hand somewhat disappointing since any application of quantum dot spin coherence requires cryogenic cooling conditions. On the other hand, the self-assembled dot samples do not have to be inserted in liquid helium; placement in cooled exchange gas is sufficient. These conditions can now be rather simply achieved in closed-cycle cryostats.

6 Optical spin rotation In the previous section we have shown that an electron spin ensemble can be almost completely driven into a single mode precession regime where to a good approximation they are in a dephasing-free subspace. This stimulates the idea to use this ensemble representing a quasi-macroscopic magnetization as a quantum bit (qubit), which exhibits particular robustness. A single spin might get scattered out of this subspace, but the rest of the spin ensemble would still carry the complete information about the quantum state. We also want to recall that in ensembles the interactions between spins as required for entanglement are potentially enhanced [12].

In order to substantiate the ensemble qubit idea, we have to demonstrate, however, that in spite of the inhomogeneity arbitrary coherent operations, rotating the magnetization vector across the Bloch sphere, are possible for a spin ensemble as well as for a single spin. Note that the Bloch sphere is now different from the one used above for the transition from vacuum/electron to neutral/charged exciton. Here a spin Bloch sphere is used which is defined by the two electron spin states, with the $|\downarrow\rangle$ and $|\uparrow\rangle$ states being the south and north poles, respectively.

Again we decided that the rotations shall be achieved by ps-laser pulses, as also here the required time should be much shorter than the spin coherence time. First we consider pulses with a photon energy in resonance with the electron–trion transition. Some indications of how to achieve a rotation of spins by laser pulses are obtained by considering again the superposition state in Eq. (5). For simplicity we assume a $\sigma^+$ polarization of the rotation pulse. Application of such a pulse with arbitrary area $\Theta$ generates a superposition of electron and trion, which is not our target as in that way the quantum bit subspace would be exited. This can only be avoided if pulses with $\Theta = 2\pi$ are applied, for which the trion component vanishes. If such a pulse is applied an interesting feature is observed as the initial state $\alpha|\uparrow\rangle + \beta|\downarrow\rangle$ is converted into $-\alpha|\uparrow\rangle + \beta|\downarrow\rangle$. This means that the Pauli-blocked electron spin component remains unchanged, while the excited component becomes phase-inverted. This is a well-known feature from quantum mechanics where under a rotation operation by an angle of $2\pi$ a spin-1/2 particle picks up a phase of $\pi$.

In our experimental situation the spin is precessing about the magnetic field. However, this precessional motion can be neglected to a good approximation during the rotating pulse application, because the pulse duration is about two orders of magnitude shorter than the period for a full precessional revolution at 1 T. If during the motion the Bloch vector points towards the south or north pole, the rotation pulse has no effect, as the spin-down electron is unexcited due to Pauli blocking and the spin-up electron is left in its pure state, picking up only an arbitrary total phase of $(-1)$. If, on the other hand, the spin vector is in the equatorial plane ($|\alpha| = |\beta| = 1/2$), the $2\pi$ pulse leads to an inversion of the spin vector. This means that the spin vector is rotated by $\pi$ around the optical axis. The same happens if the spin vector is oriented along an arbitrary direction in between south and north poles: the pulse rotates the spin by $\pi$ in the plane perpendicular to the optical axis.

The question is, how to realize other rotation angles. In principle this can be achieved by choosing other pulse areas than $2\pi$. But, since we would leave the quantum bit subspace by applying such other pulse areas this is no option to change the rotation angle. However, other angles can be achieved if the photon energy of the rotation pulse is detuned from being resonant with the trion transition. Qualitatively this can be understood in the following way: detuning reduces the geometrical phase that the electron spin picks up while the trion remains unexcited [38].

Experimental data for adjusting the laser pulse parameters to obtain spin rotations are shown in Fig. 17, which gives ellipticity traces recorded for zero detuning of the rotation pulse photon energy relative to pump and probe. Different pulse powers were applied in order to identify the proper pulse power of $2\pi$, for which, due to the $\pi$ rotation of the spin about the optical axis, the phase in the signal should
be shifted as well by $\pi$. The bottom trace gives a reference trace without rotation pulse, recorded at 0.29 T. The moment of rotation was chosen such that the spin vector lies in the equatorial plane of the Bloch sphere, having performed 2.75 revolutions about the magnetic field after initial orientation. From top to bottom the rotation pulse power and hence its pulse area was increased. For low powers no phase change relative to the reference is observed; the signal only becomes weaker. This weakening is most pronounced for a pulse area of $0.9\pi$ and is an effect of leaving the quantum bit subspace and exciting a trion component, by which the initial spin component becomes randomized through spontaneous decay.

Increasing the pulse area beyond $\pi$ leads to an increase of the signal again with a rise in time after the rotation pulse hit until a maximum is reached before decay sets in, most likely because of dephasing. This echo-like signal appearance at around 2.2 ns in Fig. 17 will be discussed in more detail below. For the highest shown rotation pulse power (the green trace) not only the signal becomes maximum but also the phase shift of $\pi$ is clearly seen. Subsequently, we fix the rotation pulse area at this level, as only then is the pulse action minimally destructive for the electron spin coherence.

Figure 18a shows the ellipticity traces when the rotation pulse is detuned from the pump towards lower photon energies, while the rotation pulse area is $2\pi$. The strength of the applied magnetic field was 0.5 T in order to roughly double the precession frequency in comparison with Fig. 17, highlighting the spin dynamics while still being to a good approximation in the single mode precession regime. A spin precession reference trace is shown by the bottom trace. The rotation pulse is applied when 3.75 revolution periods after initial orientation. Increasing the pulse area beyond $\pi$ leads to an increase of the signal again with a rise in time after the rotation pulse hit until a maximum is reached before decay sets in, most likely because of dephasing. This echo-like signal appearance at around 2.2 ns in Fig. 17 will be discussed in more detail below. For the highest shown rotation pulse power (the green trace) not only the signal becomes maximum but also the phase shift of $\pi$ is clearly seen. Subsequently, we fix the rotation pulse area at this level, as only then is the pulse action minimally destructive for the electron spin coherence.

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the detuning, where the $\pi/2$ rotation towards an orientation parallel to the field is expected, the amplitude does not drop to zero any more, but the amplitude is still about half of that for rotation angles close to zero. This is a consequence of the inhomogeneity of the spin ensemble indicated already by the dephasing. Therefore, the phase also does not jump sharply at 0.58 meV detuning by $\pi$ when moving across the $\pi/2$ rotation angle. Instead, it more or less changes smoothly, and in the 0.58 meV case the effective phase shift is $\pi/2$ indicating that the spin components with larger rotation angle and those with smaller rotation angle than the nominal one basically compensate each other.

### 6.1 Spin echoes

Let us now come to the non-monotonic evolution of the spin precession amplitude after rotation, as observed in Figs. 17 and 18a. When the rotation pulse has hit the sample, the precession amplitude first increases, then reaches a maximum, and subsequently drops again. Rise and decay occur on similar time scales and are comparable to the dephasing time. With the rotation pulse coming in at a delay of $\tau$ relative to the pump, the maximum occurs exactly at $2\tau$. This behavior is very much indicative of a spin echo.

In fact, this is exactly the behavior expected for an inhomogeneous spin ensemble. When the rotation pulse arrives, dephasing of the spins has occurred with some spins having precessed beyond the equatorial plane, while others not having reached it yet. Through the rotation pulse they are precessed beyond the equatorial plane, while others not. The dephasing of the spins has occurred with some spins having undergone inhomogeneous dephasing. Therefore, the phase also does not jump sharply when moving across the $\pi/2$ rotation angle. Instead, it more or less changes smoothly, and in the 0.58 meV case the effective phase shift is $\pi/2$ indicating that the spin components with larger rotation angle and those with smaller rotation angle than the nominal one basically compensate each other.

Note that, for obtaining an ideal echo, the rotation angle should be $\pi$, which is obtained for zero detuning. However, as long as there is an inversion of the total magnetization, echo formation should occur. It obviously should not happen when the rotation angle is clearly below $\pi/2$, so that also the inhomogeneity does not cause some spins to be rotated by more than $\pi/2$. These expectations are met in Fig. 14, where the corresponding photon energy detunings exceeding 0.58 meV no indication for an echo is observed.

A close up of an echo recorded at $B = 0.29 \, \text{T}$ is shown in Fig. 19 (the black trace). There one sees that the echo has two contributions, a high-frequency one due to electron spins and a weaker low-frequency one, arising from hole spins. This can well be seen after decomposing the trace into the two contributions (lower red and blue traces for hole and electron spin echoes, respectively). The sample on which the echo was recorded was nominally n-doped. However, this result shows that there is a fraction of quantum dots carrying a resident hole spin. This finding also underlines the necessity to move in perspective to quantum dots with controlled charging through gates, for example.

### 7 Summary

In summary, we have discussed various aspects of our quantum dot spin dynamics studies based on pump–probe spectroscopy. The specifics of the approach is performance of these studies on ensembles of singly charged dot structures. In that way, a particular robustness of the spin quantum states may be achieved and the interaction between spins might be enhanced. However, the effects of spin inhomogeneities are unavoidable as evidenced by the dephasing of the coherent spin signal. On the other hand, these inhomogeneities open possibilities as they facilitate the mode-locking phenomenon, for which the distribution of $g$-factors with the optical transition energy is required. Making use of the frequency comb provided by the mode locking, deteriorating aspects of the inhomogeneities can at least partly be overcome. Moreover, by targeted laser excitation the variations within one ensemble may be used to define specific spin sub-ensembles each of which may serve as a macroscopic quantum bit and which can interact among one another.

Based on these results, a variety of problems emerges that can be addressed in future studies. In spite of the progress in spin manipulation with laser pulses, the limitations of this technique have become apparent as well. In addition to the quantum dot inhomogeneities, the potential of laser manipulation is limited by the restricted control of pulse properties. Here potential is given by using chirped pulses, for example, which might make the manipulation more reliable with respect to the variations of the dot properties [41]. Another strategy is the combination of optical methodology with microwave techniques, which might drastically open operation possibilities.

Beyond the technical aspects, quite a few problems concerning spin physics also need to be addressed. A particular
example concerns the interaction between spins as required for spin entanglement, for example. The inhomogeneous broadening of the optical transition energies allows one to select distinct spin ensembles by ps-laser pulses with different photon energies, such that there is no spectral overlap of the pulses. In a recent work, we have shown that the spin orientation of two such ensembles leads to interaction effects between them [42]. This is shown in Fig. 20 displaying the spin precession of one ensemble of spins without and with another spin ensemble precessing. The initial spin orientations of the two ensembles were chosen to be parallel to each other.

Clearly, a phase shift is seen for the two spin ensembles precessing to a good approximation parallel to each other, despite small differences in their central electron spin g-factors. This phase shift which increases continuously with delay time must result from an interaction between the two spin subsets. Detailed studies have shown that the interaction can be well described by a Heisenberg-like form with an effective coupling constant on the order of a μeV. Still, the microscopic origin is unclear and needs to be elaborated.

Furthermore, this interaction will lead to entanglement of the two spin ensembles. However, many questions remain such as to what extent this entanglement can be exploited. For that purpose, it has to be studied how robust the coherence of the entangled states is, to what extent the entanglement can be scaled, whether the coherent states can be manipulated coherently, etc.

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