Hyperfine interaction of electron and hole spin with the nuclei in (In,Ga)As/GaAs quantum dots

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(Dated: May 11, 2009)

The population dynamics of bright and dark excitons confined in (In,Ga)As/GaAs quantum dots have been studied as function of magnetic field by two-color pump-probe spectroscopy at cryogenic temperatures. The dark excitons are stable on a few nanoseconds time scale unless the magnetic field induces a resonance with a bright exciton state. At these resonances quasi-elastic spin flips of either electron or hole occur which are initiated by hyperfine interaction with the lattice nuclei. From the relative strength of these resonances the hole-nuclear interaction is estimated to be six times weaker than the one of the electron.

PACS numbers: 78.55.Cr, 78.67.Hc

The spin dynamics of electrons and holes in quantum dots (QDs) has attracted considerable interest recently as it strongly differs from the dynamics observed in higher dimensional systems. At cryogenic temperatures the interaction with the lattice nuclei has been revealed as limiting factor for the spin coherence in QDs while all other interactions become inefficient. Typically the hyperfine interaction is described by a Fermi-like contact Hamiltonian. Therefore it has been believed that only the electrons in the conduction band with s-wave Bloch functions are subject to this interaction. In contrast, the interaction of holes with their p-wave function character has been thought to be negligible, potentially leading to comparable or even longer coherence times than those of the electrons, despite of strong spin-orbit interaction in the valence band. This has been doubted recently in theoretical works, which show that the dipole-dipole interaction between the hole spin and the nuclei spins may be as strong as the electron hyperfine interaction.

Very lately indications to that end have been found in experiment, but still the strength of this dipole-dipole interaction is under debate. It would be beneficial to have access to the nuclear interaction strength of electron and hole spins on the same sample under the same experimental conditions. This is typically not the case for different samples which have been prepared with n- or p-doping or whose charge occupation is changed by gates, as in these cases dot structure and carrier distribution, respectively, may vary.

Here we provide such a measurement by studying the spin relaxation from dark to bright ground state exciton in (In,Ga)As/GaAs quantum dots. The spin relaxation is weak unless a dark exciton state comes into resonance with a bright state, as in this case quasi-elastic scattering with the nuclei can occur. Two resonances are observed which can be attributed to an electron and a hole spin flip with the nuclei, respectively. This confirms that the hole spin hyperfine interaction is important, even though it is found to be about six times weaker than for the electron.

The experiments were performed by a two-color pump-probe technique using two synchronized, independently wavelength-tunable Ti:sapphire lasers. The lasers emit linearly polarized 1.5 ps pulses at a repetition rate of 75.6 MHz with a temporal jitter between the two pulse trains well below 1 ps. We used one laser as a pump, exciting carriers in the GaAs barrier, whereas the other probe laser was used to test the populations in the QD ground state. The temporal delay between pump and probe was adjusted by using a micrometer-precise mechanical delay line. The resulting time-resolved differential transmission signal (TRDT) is detected by a pair of balanced Si-photodiodes connected to a lock-in amplifier, by which we take the difference between the probe beam sent through the sample with and without pump action.

The sample was grown by molecular beam epitaxy and contains 10 layers of nominally undoped (In,Ga)As/GaAs QDs, separated from each other by 100-nm-wide barriers. The structure was exposed to postgrowth thermal annealing to shift the QD emission into the sensitivity range of Si detectors. From studies of QDs with similar processing parameters, we estimate the dot dimensions to be 5-6 nm height along growth direction and approximately 30 nm diameter normal to the sample.

In the experiment, the pump excitation density into the GaAs barrier at 1.55 eV photon energy was $I_0 =10$ W/cm$^2$. The probe density was chosen to be ten times weaker $I_0/10$ at an energy matching the QD ground state transition (e.g., 1.37 eV at $T=10$ K). The experiments were performed in magnetic fields $B = 7$ T applied either in longitudinal Faraday configuration (parallel to sample growth direction and optical axis) or in transversal Voigt configuration (perpendicular to the optical axis). The fields were generated by an optical split-coil magnetocryostat in whose sample chamber the temperature was varied down to 5 K.

The pump-excited QD-photoluminescence (PL) for $T =10$ K is shown in Fig. 1(a). The solid line corresponds to excitation density $I_0$, whereas the dotted line was taken at $100I_0$. At high densities, the PL spectrum shows several emission features reflecting the QD confined level structure. At the weak pump densities,
A ground-state exciton is formed by an electron with spin \( S_{e,z} = \pm 1/2 \) along the optical axis, and a hole with angular momentum \( J_{h,z} = \pm 3/2 \), assuming a pure heavy-hole character (see below). The exchange interaction couples electron and hole spin, resulting in total angular momenta \( M = J_{h,z} + S_{e,z} = \pm 1 \) and \( \pm 2 \), corresponding to bright and dark excitons, respectively. These states are split from each other by the exchange interaction \( \delta_0 \). Earlier studies on similar QDs gave \( \delta_0 \sim 100 \mu eV \).

Fig. 1 (b) shows a typical DT transient at \( T = 10K \). The non-resonant pump excitation at \( t = 0 \) excites carriers which quickly relax towards the dot ground state leading to a fast rise of the DT signal on a ps time scale. The subsequent time evolution shows two components decaying on two different time scales. The first component shows a fast drop with a 0.5 ns time constant, because of which we attribute it to bright excitons, as the same time is observed for the emission decay in time-resolved PL (not shown). The slow component decays on as time of about 8 ns, so that a fraction of this population is still present when the next pump pulse hits the sample. Therefore we associate this population with dark excitons, which are efficiently formed for excitation into GaAs where the photoexcited carriers and in particular the hole may undergo fast spin flips while relaxing towards the QD ground states. In our ensemble we therefore observe QDs with bright and dark exciton occupation.

As has been well established, the typical energy scale for the exciton fine structure given by the exchange interaction between electron and hole and the Zeeman interaction of these carriers in magnetic field is on the order of 100 \( \mu eV \) (see also above), which is considerably smaller than the thermal energy in experiment at \( T = 10 K \), equal to about a meV. Therefore the system is comparatively hot, i.e. spin-orbit interaction mediated carrier spin flips involving phonons may be considered as efficient. However, the experiment reveals dark exciton lifetimes in the ns-range, showing inefficient scattering with phonons due to the significant mismatch of the fine structure energies and the energy of acoustic phonons with a wavelength comparable to the QD size.

The dark exciton assignment is confirmed by experiments in transverse magnetic field shown in Fig. 1 (c). In this configuration the carrier spins precess about the field, so that dark excitons are periodically converted into bright states and vice versa. In a quantum mechanical picture, the Zeeman interaction breaks the rotational symmetry about the growth direction, leading to a mixing of the exciton states which are bright and dark at \( B = 0 \), so that the bright exciton oscillator strength is distributed among the four exciton states and all of them become optically active. This should lead to a strong reduction of the dark exciton lifetime, as confirmed by Fig. 1 (c). Shown are contour plots of the DT transients (along the vertical scale) as function of the applied Voigt magnetic field up to 7 T in steps of 0.2 T.

The dark exciton decay continuously decreases with increasing magnetic field, and for field strengths beyond \( \sim 5 T \) has vanished completely. The upper scale of Fig. 1 (c) gives the ratio of the bright exciton radiative decay time of 0.5 ns to the electron spin precession time which is proportional to \( B \). The dark exciton background has in effect vanished when the electron perform about 20 revolutions about \( B \) within the decay time.

Here we are interested in the evolution of the dark exciton background when a Faraday field is applied. The exciton fine structure is described then by the effective spin Hamiltonian, following the notations in Ref. 13.

\[
H_X = \mu_B \left( g_{e,z} S_{e,z} + \frac{g_{h,z}}{3} J_{h,z} \right) B - \frac{4}{3} \delta_0 S_{e,z} J_{h,z},
\]

where \( \mu_B \) is the Bohr magneton and the \( g_{e,z} \) and \( g_{h,z} \) are the electron and hole \( g \)-factors along \( z \). We also neglect the coupling of electron spin and hole spin normal to \( z \), as the resulting splittings of the bright and dark excitons are negligibly small for these QDs. Very similar QDs
were investigated recently by pump-probe Faraday rotation spectroscopy, to determine the parameters of the exciton fine structure Hamiltonian with high accuracy. Besides the already used $\delta_0 = 100 \pm 10 \mu$eV, electron and hole $g$-factors of $g_{e,z} = -0.61$ and $g_{h,z} = -0.45$ were obtained.25

These parameters were used as input for calculating the exciton fine structure splitting in a Faraday magnetic field as shown in Fig. 2. This field configuration does not break the rotational symmetry so that the exciton angular momentum $M$ remains a good quantum number throughout the whole scanned field range. The $B$-linear splitting of the bright and dark excitons leads to crossings in the magnetic field dispersion: The $| -2 \rangle$-exciton, consisting of spin down hole and electron (represented by $\downarrow$ and $\downarrow$, respectively) crosses with the $| -1 \rangle$-exciton ($\downarrow\uparrow$) around 3 T. Further, it also crosses with the $| +1 \rangle$-exciton ($\uparrow\downarrow$) at approximately 4 T.

Figure 3 (a) shows DT transients as function of the magnetic field, applied in Faraday configuration. At low fields $< 1$ T the signal shows the two component behavior already discussed in relation with Fig. 1. After pump action the fast bright population decay is followed by the significantly slower dark exciton decay. Above 1 T, however, the slow decay becomes drastically shortened with increasing field up to 3 T, while for even higher fields the decay time increases again reaching times almost as long as at zero field. Apparently the decay dynamics of the dark excitons shows a resonance around 3 T: at $B = 0$ the estimated dark exciton decay time is 8 ns which is reduced to $\sim 5$ ns in resonance and then increases again to $\sim 8$ ns. The behavior of the bright exciton decay time is reversed with the dark exciton as it increases from $\sim 0.5$ ns at $B = 0$ to $\sim 0.6$ ns at $B = 3$ T and then drops again to 0.5 ns for high fields. Note that some long lived background remains even at 3 T because of the occupation of the $| +2 \rangle$ dark exciton.

The resonance exactly occurs at the field strength at which the crossing of the $| -2 \rangle$ exciton with the $| -1 \rangle$ exciton occurs. However, the resonance behavior is not symmetric with respect to the resonance field but features an asymmetry towards higher $B$. This asymmetry may indicate that also the resonance of the $| -2 \rangle$ exciton with the $| +1 \rangle$ exciton leads to a shortening of the dark exciton lifetime. In combination with the reversed behavior of the magnetic field dependence of the decay of the two exciton components, the observation of field resonant shortening of the dark exciton background can be traced to exchange processes with the bright exciton states, for which spin-flips are required.

To analyse this in more detail, Fig. 3 (b) shows the mean value of the DT traces recorded at negative delay $t < 0$ before pump pulse action. Each DT trace was normalized by its maximum; the resulting dataset was baseline substracted. By doing so, values below zero indicate a field induced reduction of the dark exciton population. Besides the main resonance at 2.9 T, clearly a shoulder is observed towards higher magnetic fields, supporting that indeed two field resonances occur. To determine the second resonance field, we have fitted the data with a
superposition of two Gaussians with the same halfwidth, resulting to 1.56 T. The Gaussian form is justified as the broadening of the resonances is inhomogeneity related. The data can be well described by this fit as shown by the solid red line. The two dashed curves give the individual resonances. The field position of 4.7 T for the solid red line. The two dashed curves give the inhomogeneity related.

Let us consider more in detail what spin flip scattering events are involved in the resonances. The dominant resonance involves the |−2⟩ and |−1⟩ excitons, which can be transferred into each other by an electron flip. We have already discussed that spin-orbit induced flips are generally negligible due to the lack of phonon states which in addition have zero density of states in the resonance case with zero energy difference between two states.16 The quasi-elastic flip can therefore be initiated only by the hyperfine interaction of the electron spin $\mathbf{S}_e$ with the nuclei $\mathbf{I}_i$, described by:

$$\mathcal{H}_e = \sum_i A_i |\psi(\mathbf{r}_{ei})|^2 \mathbf{S}_e \mathbf{I}_i,$$

(2)

where the sum goes over all nuclei in the QD electron localization volume. The interaction strength of the electron with the nucleus is determined by the hyperfine constant $A_i$ specific for each nuclear species in the dot and the electron density $|\psi(\mathbf{r}_{ei})|^2$ at the nuclear site $\mathbf{r}_{ei}$.

The second weaker resonance involves the |−2⟩ and |+1⟩ excitons, which can be transferred into each other by a hole spin flip only. Due to the quasi-elastic scattering in the resonance again this flip-process can be initiated by the nuclei only, demonstrating the importance of the hole-nuclei interaction. The only relevant mechanism in this case would be a dipole-dipole interaction which, however, cannot convert the ±3/2 heavy-hole states into each other due to the mismatch of angular momentum exchange. On the other hand, we know that in the studied QDs the in-plane hole g-factor differs considerably from zero ($g_{h,z} = 0.15$),22 showing that the hole ground state contains significant admixture of light-hole states with $J_{h,z} = \pm 1/2$. Due to this admixture, the hole-nuclei flip-flops become possible. The underlying Hamiltonian is:

$$\mathcal{H}_h = \sum_i C_i |\psi(\mathbf{r}_{hi})|^2 \left( c_i \left( I_{h,x}^2 J_{h,x} + I_{h,y}^2 J_{h,y} + I_{h,z}^2 J_{h,z} \right) \right),$$

(3)

where the sum goes again over all dot nuclei, the $\psi$ is the hole envelope wave function, and the $C_i$ are the corresponding interaction constants. The $c_i$ measure the light-hole content of the valence band ground state. Any flip-flop processes can only be initiated by the first two terms on the right-hand side. Thus, if $c_i = 0$ for a pure heavy-hole state, such processes would be suppressed.

Despite of recent observations that this dipolar coupling indeed plays an important role for the hole spin dynamics, its strength relative to the electron hyperfine interaction is still under heavy debate. From our resonances we obtain an estimate of this relative strength within the same sample: By comparing the enclosed areas of the two fit curves, we estimate that the hole-nuclear interaction is about six times weaker than that of the electrons, but still considerable, contrasted with the previous claims that it can be neglected.

We gratefully acknowledge the financial support by the Deutsche Forschungsgemeinschaft (DFG 1549/10-1).

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1 Spin Physics in Semiconductors, edited by M. I. Dyakonov (Springer-Verlag, Berlin, 2008).
2 I. A. Merkulov, Al. L. Efros, and M. Rosen, Phys. Rev. B 65, 205309 (2002).
3 A. V. Khaetskii, D. Loss, and L. Glazman, Phys. Rev. Lett. 88, 186802 (2002).
4 K. V. Kavokin, Phys. Rev. B 69, 75302 (2004).
5 W. A. Coish and D. Loss, Phys. Rev. B 70, 195340 (2004).
6 V. N. Golovach, A. Khaetskii, and D. Loss, Phys. Rev. Lett. 93, 016601 (2004).
7 J. R. Petta, A. C. Johnson, J. M. Taylor, E. A. Laird, A. Yacoby, M. D. Lukin, C. M. Marcus, M. P. Hanson, and A. C. Gossard, Science 309, 2180 (2005).
8 P. F. Braun, X. Marie, L. Lombez, B. Urbaszek, T. Amand, P. Renucci, V. K. Kalevich, K. V. Kavokin, O. Krebs, P. Voisin, and Y. Masumoto, Phys. Rev. Lett. 94, 116601 (2005).
9 Optical Orientation, edited by F. Meier and B. P. Zakharchenya (North-Holland, Amsterdam, 1984).
10 S. Laurent, B. Eble, O. Krebs, A. Lemaître, B. Urbaszek, X. Marie, T. Amand, and P. Voisin, Phys. Rev. Lett. 94, 147401 (2005).
11 D. V. Bulaev and D. Loss, Phys. Rev. Lett. 95, 076805 (2005); ibid. 98, 097202 (2007).
12 E. Tsitsishvili, R. v. Baltz and H. Kalt, Phys. Rev. B 67, 205330 (2003); ibid. 72, 155333 (2005).
13 J. Fischer, W. A. Coish, D. V. Bulaev, and D. Loss, Phys. Rev. B 78, 155329 (2008).
14 B. Eble, C. Testelin, P. Desfonds, F. Bernardoit, A. Balocchi, T. Amand, A. Miard, A. Lemaître, X. Marie, and M. Charmaré, Phys. Rev. Lett. 102, 146601 (2009).
15 I. A. Yugova, A. Greilich, E. A. Zhukov, D. R. Yakovlev, M. Bayer, D. Reuter, and A. D. Wieck, Phys. Rev. B 75, 195325 (2007).
16 Phonon scattering of carriers to higher shells and subsequent relaxation is negligible at the considered tempera-
tures.