Anomalous Hanle Effect due to Optically Created Transverse Overhauser Field in Single InAs/GaAs Quantum Dots

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We report on experimental observations of an anomalous Hanle effect in individual self-assembled InAs/GaAs quantum dots. A sizable electron spin polarization photocreated under constant illumination is maintained in transverse magnetic fields as high as $\sim$1 T, up to a critical field where it abruptly collapses. These striking anomalies of the Hanle curve point to a novel mechanism of dynamic nuclear spin polarization giving rise to an effective magnetic field generated *perpendicular* to the optically injected electron spin polarization. This transverse Overhauser field, confirmed by the cancellation of electron Zeeman splitting below the critical field, is likely to be a consequence of the strong inhomogeneous quadrupolar interactions typical for strained quantum dots.

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The spin of a conduction electron confined in a semiconductor quantum dot (QD) is a natural two-level system exhibiting a coherence time that is remarkably long for condensed matter [1–6]. For QDs based on III–V semiconductors, this coherence time is yet limited by the nonuniform hyperfine coupling to $N \sim 10^4$–10^6 QD nuclear spins [7,8]. Suppression of this decoherence mechanism requires a complete understanding of nuclear spin dynamics and fluctuations, which could be achieved by using the same hyperfine interaction along with optical manipulation of the QD electron spin. In particular, cross relaxation between an optically pumped electron spin and the nuclear spins of the QD causes dynamic nuclear polarization (DNP). The resulting Overhauser field (or ‘‘nuclear’’ field) is an effective magnetic field $\boldsymbol{B}_n$, usually parallel to the photocreated electron spin orientation and acting back on the electron spin dynamics. As recently shown in distinctive experimental configurations where the electron spin state is addressed resonantly, this feedback can lead to a drastic reduction of the nuclear spin fluctuations [9–11]. Alternatively, in an external magnetic field opposite to the Overhauser field photocreated under nonresonant excitation, the DNP feedback gives rise to a pronounced bistability regime where the nuclear field can overcome the external field of up to a few Teslas [12–16].

To further explore this intriguing system, we investigate the depolarization of electron spin by a transverse magnetic field, a configuration where the generated Overhauser field should vanish because of the Larmor precession of the nuclei about the transverse field [17]. The resulting electron depolarization due to spin precession, the so-called Hanle effect, is thus expected to be a Lorentzian curve with HWHM given by $B_{1/2} = \hbar/(|g_e|\mu_B \tau_s^\star)$ [17] where g_e is the Landé g factor of the QD electron, $\tau_s^\star$ the electron spin lifetime and μ_B the Bohr magneton. Such typical Hanle curves are indeed observed in individual, strain-free GaAs QDs under 40 kHz σ^+/σ^- polarization modulation of the excitation light, indicating the absence of Overhauser field [18]. However, recent experimental studies on ensembles of self-assembled InP/InGaP QDs [19] have revealed a 3 times broadening of the Hanle curve when the excitation polarization is kept constant. This behavior has been attributed to the existence of a strong longitudinal Overhauser field $B_{n,z}$ persisting even in the presence of a transverse field because of strain-induced quadrupolar splittings (QS) of the nuclear spin states.

In this Letter we report on the Hanle effect for electrons in self-assembled InAs/GaAs QDs. By performing spectroscopy on individual QDs, we find drastic distortions of the depolarization curves characterized by a $\sim$20 times broadening and qualitatively new features such as (i) an abrupt drop from 50% to zero of the polarization at a critical field and (ii) a pronounced hysteresis when sweeping the applied field back and forth. Our measurements show that this anomalous Hanle effect results from a strikingly new DNP mechanism characterized by the development of an Overhauser field *perpendicular* to the optical spin orientation which compensates completely the external field up to $\sim$1 T. In contrast to the conclusion of Ref. [19], we find that only a small longitudinal nuclear field subsists. Yet, the role of nuclear QS seems still essential to understand the conversion of the longitudinal electron spin polarization into a transverse Overhauser field.

We have investigated two samples (A and B) grown by molecular beam epitaxy on a semi-insulating GaAs [001] substrate and consisting of a single layer of self-assembled InAs/GaAs QDs. In sample A, the QDs are positively charged with one excess hole due to residual doping, while sample B has a diode structure enabling us to control the charge state with a gate voltage [20]. The μ-photoluminescence (PL) spectroscopy of individual QDs was carried out in split-coil magneto-optics cryostats at $T = 1.8$ K with optical setups providing a typical spectral resolution of 25 μeV.

To measure the Hanle effect of electrons (e), individual QDs charged with a single excess hole (h) are optically excited with a quasiresonant circularly polarized light propagating along the z direction. This creates positively charged excitons X^+ ("trions") consisting of 1 electron and 2 holes. Owing to optical selection rules and to the spin conservation by phonon-assisted relaxation, the spin of the photocreated electron can be prepared in state $\uparrow$ or $\downarrow$, while both holes are paired in a spin singlet state, as schematically illustrated in Fig. 1(a). The optical orientation of the e spin, $\langle S_z^e \rangle$, can be monitored by the PL circular polarization $\mathcal{P}_c$ through the relation $\mathcal{P}_c = -2\langle S_z^e \rangle$ [12,21]. In zero field, $\mathcal{P}_c$ achieved under quasiresonant excitation reaches more than 70%, as shown in Fig. 1(c). Since in InAs QDs the electron spin lifetime $\tau_s^\star$ is essentially determined by the trion radiative lifetime $\tau_r \approx 1$ ns, the half-width of the Hanle curve, $B_{1/2}$, is expected to be $\sim$30 mT by taking a typical value of $|g_e| \sim 0.5$. The exact value of g_e can be determined by fitting the characteristic X^+ Zeeman splitting into 4 lines, which are resolvable in strong transverse magnetic fields [Fig. 1(b)]. The inner (π_x polarized) and outer (π_y polarized) lines which emerge from the zero-field X^+ line are split by the sum and difference of the e and h Zeeman splittings [see Fig. 1(b)]. In accordance with previously reported $|g_e|$ values for sample A [22], we extract $|g_e| = 0.34$ and $|g_h| = 0.6$ from this measurement. We note that the identical analysis on the QD studied in sample B yields $|g_e| = 0.46$ and $|g_h| \approx 0$ [23]. The Overhauser shift Δ_n at $\boldsymbol{B}_{\rm ext} = 0$ [Fig. 1(c)] corresponds to a nuclear magnetic field $B_{n,z} = \Delta_n/(|g_e|\mu_B) = 0.35$ T experienced by the electron. We would therefore expect possible anomalies in the Hanle curve for a range of transverse magnetic fields of at most a few 100 mT.

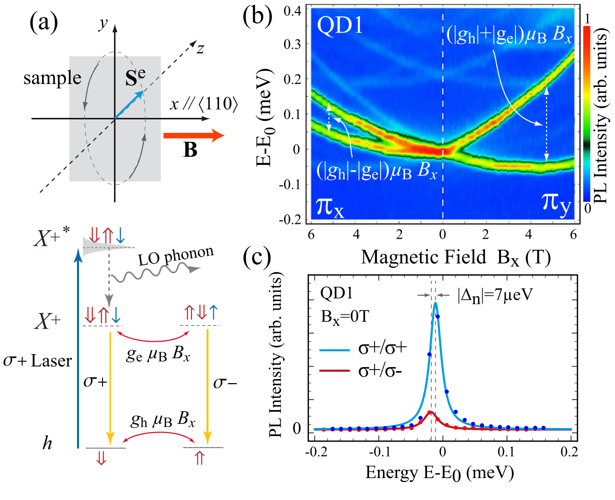

FIG. 1 (color online). (a) Experimental geometry for Hanle effect measurements and schematics of X^+ spin orientation. (b) Dispersion of the PL intensity around $E_0 = 1.3522$ eV for QD1 in a transverse magnetic field under linearly polarized excitation, measured in linear polarization π_x or π_y. (c) X^+ PL line in $\sigma\pm$ polarization at $\boldsymbol{B}_{\rm ext} = 0$ under σ^+ excitation at 1.396 eV.

Figure 2 shows typical Hanle depolarization curves for samples A and B. The discrepancy to the expected Lorentzian curve is considerable. The width of Hanle curves is increased by more than 1 order of magnitude with respect to the expected $B_{1/2}$ and their shape is clearly distorted from a Lorentzian. In sample A [Fig. 2(a)], $\mathcal{P}_c$ decreases slowly (over $\sim$0.5 T) to about half of its initial value, and then remains essentially constant until it abruptly collapses to zero. This drastic nonlinearity indicates a likely bistability of the electron-nuclear spin system which is indeed revealed by a hysteretic behavior when sweeping B_x back to zero (see QD1 and QD2). This behavior differs noticeably from the DNP nonlinearity observed in a longitudinal magnetic field which exhibits a strong asymmetry with respect to the field direction [12–16]. Here, the nonlinearity is found for both directions of

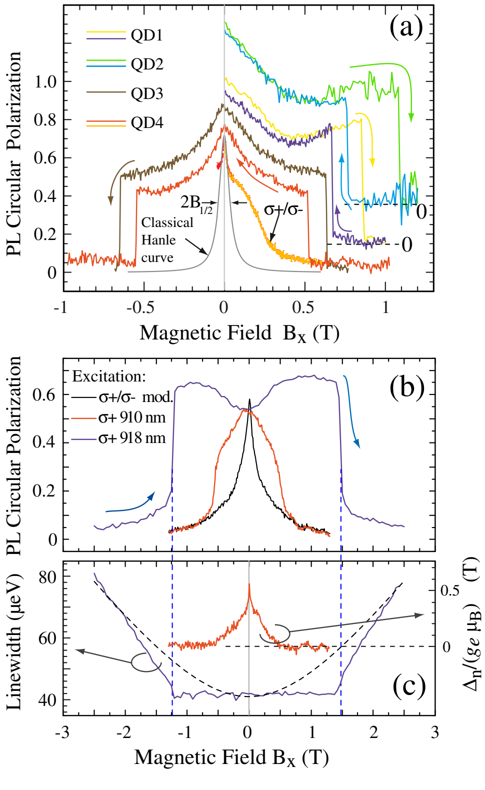

FIG. 2 (color online). (a) Hanle depolarization curves of X^+ trions in 4 different QDs in sample A, under a constant σ^+ polarized excitation. QD4 was also measured under σ^+/σ^--modulated excitation. Arrows indicate field sweep directions. Curves for QD1 and QD2 are vertically shifted for clarity. (b) Hanle depolarization curves of an X^+ trion in sample B under different excitation conditions. (c) X^+ apparent linewidth and nuclear field component $B_{n,z}$ estimated from the σ^+-σ^- splitting $|\Delta_n|$. The dashed line is the evolution of effective linewidth due to e Zeeman splitting.

the transverse field (see QD3 and QD4). Sample *B* exhibits a very similar dependence of $\mathcal{P}_c$ on the magnetic field [Fig. 2(b), red or gray curve]. In addition, we find that the qualitative behavior of the Hanle curve depends sensitively on the distinct resonances used for QD excitation as shown in Fig. 2(b). We assign these qualitative changes to the various QD excitation channels that are involved under quasiresonant excitation [24].

The anomalous Hanle curves and the magnetic field range over which these anomalies occur, suggest that the effective magnetic field experienced by the QD electron is strongly influenced by nuclear fields. This conclusion is further supported by measuring Hanle depolarization curves where the excitation polarization is modulated between σ^+ and σ^- at a few kHz—significantly faster than the time scale of DNP buildup [25] [Figs. 2(a) and 2(b)]. As a result, $\langle S_z^e \rangle$ averages to zero and nuclear fields should be strongly suppressed. We find that in this case, the strong singularities of anomalous Hanle curves indeed vanish and that $B_{1/2}$ is reduced to $\sim$0.2 T. Even under modulated excitation helicity, however, the normal Hanle line shape is still not recovered. It is known that under such excitation nuclear spin effects cannot be completely neglected. Processes like "resonant spin cooling" [17] could still affect the shape of the Hanle curve—a detailed description of these effects, however, is out of the scope of this Letter.

The fact that under constant excitation polarization, $\mathcal{P}_c$ is substantially preserved requires the total magnetic field $\boldsymbol{B}_T = \boldsymbol{B}_n + \boldsymbol{B}_{\text{ext}}$ to have a dominant *z* component ($|B_{T,z}| \geq |B_{T,\perp}|$) or a small in-plane strength ($|B_{T,\perp}| \leq B_{1/2}$). This can be achieved either by (a) a strong nuclear field along the *z* axis, such that $|B_{n,z}| \geq |B_x|$, or (b) by a nuclear field nearly antiparallel to B_x such that $|B_{n,x} + B_x| \leq \max(|B_{n,z}|, B_{1/2})$. Option (a) clearly holds in zero applied field; in the presence of a transverse external magnetic field, however, one would expect $B_{n,z}$ to vanish due to Larmor precession. As discussed in Ref. [19], quadrupolar interactions of nuclear spins in QDs could "stabilize" the nuclear spins and allow for a finite $B_{n,z}$, even in the presence of B_x. As long as $|B_{n,z}| \geq |B_x|$, the Hanle effect should therefore be suppressed giving rise to a broadening of the depolarization curve of the order of $|B_{n,z}|$.

Our experimental observations, however, rule out option (a). By measuring the energy-splitting Δ_n between the σ^+- and σ^--polarized PL lines, we indeed determine $|B_{n,z}| \sim$ 0.5 T at $B_{\text{ext}} = 0$. Since $|B_{n,z}|$ is decreasing with increasing transverse field [as evidenced by the measurement of $\Delta_n(B_x)$ in Fig. 2(c)], it cannot cause the stabilization of $\mathcal{P}_c$ for B_x up to 1 T. We therefore conclude that the observed anomalous Hanle effect is a result of an in-plane nuclear magnetic field which develops antiparallel to the applied field.

While the detailed microscopic process for the establishment of this nuclear spin polarization is still unclear, several observations qualitatively support scenario (b): (i) in the presence of a tilted magnetic field $\boldsymbol{B}_T$, $\boldsymbol{S}^e$ can acquire a finite average value along *x* [see Fig. 3(c)], which can be subsequently transferred to the nuclear spins via DNP, (ii) the nuclear spin component $B_{n,x}$ is conserved by the applied field; besides, in-plane strain in the QD lattice and alloy disorder due to Ga and In intermixing could lead to a further stabilization of the nuclear field along *x* [19,26], and (iii) compensating the external field with a nuclear field reduces the total *e* spin splitting and therefore favors a high DNP rate [15,27]. Scenario (b) is further supported by the X^+ linewidth shown Fig. 2(c) which has been deduced from the sum of σ^+ and σ^- PL lines. It remains constant (limited by spectral resolution) until the critical field of polarization collapse is reached. At this point the apparent linewidth undergoes an abrupt increase

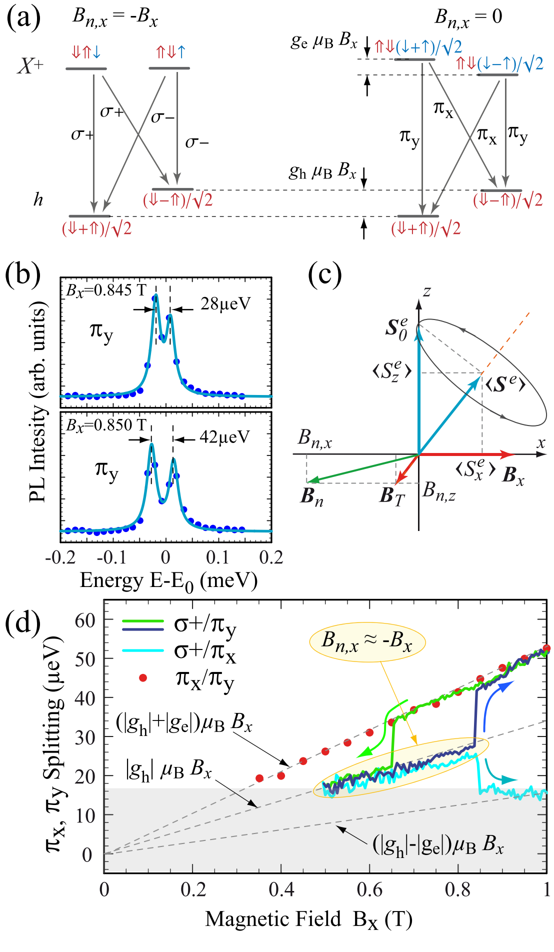

FIG. 3 (color online). (a) Schematics of X^+ optical transitions in a transverse magnetic field B_x for both special cases $B_{n,x} = -B_x$ and $B_{n,x} = 0$, assuming $B_{n,z} = 0$. (b) Fit of the QD1 X^+ PL line measured in linear π_y polarization by a Lorentzian doublet on both sides of the circular polarization collapse. (c) Diagram of *e* spin and nuclear field components leading to the experimental observations. (d) Splitting of the QD1 X^+ line measured in π_x or π_y polarization. Dashed lines indicate the *h* and $e \pm h$ Zeeman splittings. The gray-shaded area represents the resolution limit for the fit in (b).

due to the restoration of the trion Zeeman splitting when the in-plane nuclear field vanishes.

To really demonstrate the existence of an in-plane nuclear field, we measured QD1 in the basis of linear polarizations (π_x and π_y) while keeping a circularly-polarized excitation. This choice corresponds to the proper selection rules of X^+ transitions for $B_{n,x} = 0$, as illustrated in Fig. 3(a), and thus improves the resolution of their splittings. When the trion circular polarization abruptly vanishes at 0.85 T, we observe a clear discontinuity of the π_x- and π_y-polarized splittings as deduced from a double Lorentzian fit of the experimental trion line, see Fig. 3(b). The π_y splitting undergoes an increase by 14 μeV, while the π_x splitting is reduced by at least 10 μeV to ≈ 20 μeV (the validity limit of our fit). These jumps reflect the situation depicted in Fig. 3(a). Below 0.85 T, the nuclear field almost exactly compensates the applied field such that the trion splitting originates mostly from the h spin splitting $|g_h|\mu_B B_x$. Above 0.85 T, the nuclear field has essentially vanished so that the π_x (π_y) splitting is decreased (increased) by $|g_e|\mu_B B_x$. As shown in Fig. 3(d) this interpretation agrees well with the g factors determined above in a strong field (dashed lines). When the magnetic field is swept back to zero, we observe the opposite jumps yet shifted to a lower field (0.65 T) in agreement with the polarization hysteresis of QD1. As a control experiment, we also checked that under linearly-polarized excitation (i.e., no nuclear field) the π_y splitting evolves linearly as $(|g_e| + |g_h|)\mu_B B_x$.

Finally, the likely scenario for DNP in a transverse magnetic field is depicted in Fig. 3(c). Assuming a fast e Larmor precession (i.e., $|\boldsymbol{B}_T| > B_{1/2}$), the average e spin $\langle \boldsymbol{S}^e \rangle$ would be roughly aligned along the total field axis. Thereby, the nuclear field optically generated through DNP is antiparallel to $\langle \boldsymbol{S}^e \rangle$, because of the negative sign of g_e. Under the action of $\boldsymbol{B}_x$ its z component should vanish, but thanks to the nuclear QS in biaxially strained InAs QD [19,26], a finite $B_{n,z}$ component can subsist. The resulting average nuclear field $\langle \boldsymbol{B}_n \rangle$ is thus neither colinear to $\langle \boldsymbol{S}^e \rangle$ nor to the applied field. The $B_{n,z}$ component is essential in this scenario to maintain the out-of-plane component of $\boldsymbol{B}_T$. It allows for spin transfer from $\langle S_z^e \rangle$ to $\langle S_x^e \rangle$ giving rise to the in-plane nuclear field $B_{n,x}$ which countervails the applied field.

In conclusion, we have observed a spectacular broadening and hysteretic behavior of Hanle depolarization curves for electrons in InAs QDs. The analysis of these data evidences a novel mechanism of dynamic nuclear polarization characterized by a strong nuclear field almost perpendicular to the optically pumped electron spin orientation and antiparallel to the applied field. This transverse Overhauser effect is an alternative explanation to the Hanle-curve broadening observed for a QD ensemble in Ref. [19]. We suspect the strong nuclear quadrupolar shifts arising from the QD biaxial strain to play a crucial role in the establishment of this nuclear field. Further theoretical and experimental investigations are necessary to understand how this counterintuitive regime of the coupled electron-nuclear spin system develops in InAs QDs. This could reveal interesting new effects such as self-sustained oscillations of the nuclear spin-polarization [17].

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