Optical polarization of localized hole spins in *p*-doped quantum wells

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The initialization of spin polarization in localized hole states is investigated using time-resolved Kerr rotation. We find that the sign of the polarization depends on the magnetic field and the power and the wavelength of the circularly polarized pump pulse. An analysis of the spin dynamics and the spin-initialization process shows that two mechanisms are responsible for spin polarization with opposite sign: the difference of the g factor between the localized holes and the trions, as well as the capturing process of dark excitons by the localized hole states.

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I. INTRODUCTION

Localized hole spins in p-doped III–V semiconductors can have considerably long spin lifetimes[1] in the range of 100 μs and coherence times[2] on the order of μs. One possibility to localize holes or electrons consists of natural quantum dots (QDs) formed due to potential fluctuations in quantum wells (QWs).[3,4] In p-doped GaAs/AlGaAs QWs, the spin of these localized hole states has been studied using time-resolved Kerr rotation (TRKR).[5–8] Information on localized hole spins can also be obtained in n-doped QW structures by studying the recombination of an optically excited hole spin with a resident electron.[9,10] The reliable polarization of hole spins in QDs is one of the key requirements necessary to study this quantum system. Possible polarization mechanisms make use of the properties of the optically excited positively charged trion that are different than those of the bare hole, for example, the different interaction with nuclear spins[11] or the different g factor.[7]

In our study, a p-doped QW is excited with circularly polarized photons, giving rise to two competing spin polarization mechanisms that lead to polarization of an ensemble of localized holes. The two mechanisms polarize the spins with opposite signs, and their relative strength depends on the external magnetic field (B_{ext}), the pump power (P_{P}), and the wavelength (λ) of the pump beam. The first mechanism is found to rely on the difference of the trion and the hole g factors and disappears for $B_{\text{ext}} = 0$. The second mechanism remains effective for $B_{\text{ext}} = 0$ and strongly depends on P_{P} and λ. The relative strength of the two mechanisms can therefore be controlled by the properties of the pump pulse. We show that the second mechanism can be explained by a capturing of dark excitons by the localized holes.

II. EXPERIMENT

To generate and study the spin polarization, we employ TRKR.[12,13] A mode-locked Ti:sapphire laser generates 2-ps-long laser pulses with a repetition rate of 80 MHz. The laser pulses are split into a pump and a probe beam. Using a photoelastic modulator, the pump pulses are modulated between left- and right-circular polarization with a frequency of 50 kHz, generating excitons and trions in the QW with alternating spin polarization. The modulation of the photon helicity strongly reduces effects that could arise from dynamic nuclear polarization.[14] The circularly polarized laser pulses are focused onto a spot with a diameter of about 40 μm.

As an example, we consider a σ^--polarized pump pulse that directly excites localized $|\Uparrow\rangle$ holes to the $|\Downarrow\Uparrow\uparrow\rangle$-trion state [see Fig. 1(c)]. In addition to this resonant pumping, the σ^--polarized photons also create free $|\Downarrow\uparrow\rangle$ excitons. Because of the large spin-orbit interaction in the valence bands, the hole in these excitons has a higher probability to flip its spin than the electron.[15,16] This process results in the formation of dark excitons which cannot recombine optically. However, they can be captured by a localized $|\Downarrow\rangle$ hole to form a $|\Downarrow\Uparrow\uparrow\rangle$ trion.[17–19] This indirect pumping process is symbolized by the arrow marked with an X in Fig. 1(c) and has been also observed in the case of negatively charged trions.[19] When the optically excited trions recombine, they leave behind localized hole spins. The excitonic pumping process X polarizes the localized holes to the $|\Uparrow\rangle$ state, whereas the direct excitation [process σ^- in Fig. 1(c)] and subsequent recombination does not lead to a polarization of localized holes at $B_{\text{ext}} = 0$ T. At finite magnetic field B_{ext} however, the electron spin in the $|\Downarrow\Uparrow\uparrow\rangle$-trion precesses, leading to polarization of holes into the $|\Downarrow\rangle$ state after recombination. The competition between this precession-induced mechanism and the polarization by the pumping process X determines the degree I_h of hole-spin polarization after recombination of extions and trions and is the subject of this paper.

The polarization of the localized hole spins is detected by analyzing the Kerr rotation of linearly polarized probe pulses that are delayed by a time Δt with respect to the pump pulses. The probe intensity is modulated with an optomechanical chopper at a frequency of 189 Hz. The Kerr rotation Θ_{K} of the reflected probe pulses is analyzed using an optical bridge and reveals the component of spin polarization along the QW growth direction $\mathbf{z}$ at time Δt.[12] The measured signal is modulated at the frequency of both the optomechanical chopper and the photoelastic modulator, and we can use standard lock-in techniques for noise reduction.[13]

We investigate a 4-nm-wide remotely doped GaAs/AlGaAs QW with a hole sheet density of 1.1×10^{15} m^{-2} and a mobility of 1.3 m^2 (Vs)$^{-1}$ measured at 1.3 K. We concentrate on this

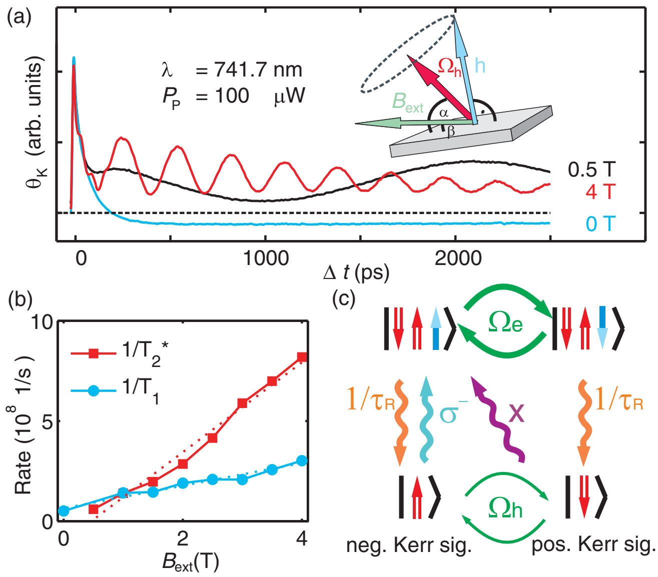

FIG. 1. (Color online) (a) Experimental TRKR signals for three values of $B_{\rm ext}$ for a probe power of 20 μW. The insert schematically shows the precession of the hole spins (h) in the tilted magnetic field $B_{\rm ext}$ about the precession axis Ω_h. (b) Hole spin relaxation rates vs $B_{\rm ext}$ as described in the text. (c) Arrows indicate the transitions between spin-polarized localized hole states (lower part) and trion states (upper part) when excited with σ^- photons. Direct excitation of trions (labeled σ^-) competes with indirect excitation X involving capturing of a $|\Downarrow\rangle$ hole by a dark $|\Uparrow\uparrow\rangle$ exciton. $1/\tau_R$ characterizes the recombination rate, whereas Ω_e and Ω_h describe precession of the trion and localized hole spin, respectively.

specific sample because the hole spin lifetime is much longer for narrow QWs, reaching up to 70 ns at a temperature below 1 K.[7] Photoluminescence from this QW at 1.6 K excited with a 633-nm continuous-wave laser displays a peak centered at 742.4 nm. The width of the peak is rather broad (15 meV), probably because of large interface roughness relative to the small QW width, leading to inhomogeneous broadening.

Figure 1(a) shows experimental TRKR signals at three different magnetic fields. At $B_{\rm ext} > 0$, $\Theta_{\rm K}$ is the sum of two exponentially decaying cosine functions and a nonoscillating exponential function. The short-lived oscillation (best seen at $B_{\rm ext} = 4$ T and $0 < \Delta t < 100$ ps) is attributed[5] to the trion spin which is determined by the spin of the electron in the trion and therefore precesses with the electron g factor g_e. We measure a decay time of $\tau_{\rm T} = 80$ ps and $g_e = 0.34$. We assume that this part of the signal decays mainly due to the recombination rate of the trions $(1/\tau_R)$;[6] that is, $\tau_{\rm T} \cong \tau_{\rm R}$. The longer-lived part of $\Theta_{\rm K}$ originates from the localized hole spins.[5] Due to the tilt angle β and the strong anisotropy of the hole g factor, the precession axis of the hole spins is tilted out of plane[7] by an angle $\alpha > \beta$ [see insert of Fig. 1(a)]. The Kerr signal is proportional to the projection of the spins of the ensemble along **z** and therefore also has a nonoscillating part. Note that for the two curves at $B_{\rm ext} = 0.5$ and 4 T, the trion and both parts of the hole signal have a positive amplitude.

The situation for $B_{\rm ext} = 0$ is strikingly different. There, $\Theta_{\rm K}$ can be described by a superposition of two exponentials with amplitudes of opposite sign. While the short-lived trion signal is still positive, the long-lived signal from the localized holes now has a negative amplitude. From this, we conclude that there must be a $B_{\rm ext}$-dependent sign change of the initialized hole polarization. We have compared Kerr signals for finite and zero magnetic fields for various λ and QWs with widths between 4 and 15 nm and always found a sign-reversal of the hole spins.

We now proceed to analyze the observed dynamics in more detail. The TRKR signal of the localized holes (neglecting the fast-decaying component from the trion spin) is described by

$$\Theta_{\rm K} = A \cdot I_h\left[a_1 e^{-\Delta t/T_1} + a_2 e^{-\Delta t/T_2^*}\cos(\Omega_h \Delta t)\right], \qquad (1)$$

where $\Omega_h = g_h \mu_B B_{ext}/\hbar$. Here, A is the λ-dependent amplitude of the Kerr rotation and $g_h \approx 0.06$ is the hole g factor. The nonoscillating and oscillating parts are proportional to the projection of the respective spin component onto **z** and are given by $a_1 = \sin^2\alpha$ and $a_2 = \cos^2\alpha$. They decay with time constants T_1 and T_2^*. The function $I_{\rm h}$ describes the effectiveness of the pump pulse to initialize hole-spin polarization. It depends on $B_{\rm ext}$, λ, and $P_{\rm P}$. The sign change of the hole-spin polarization at low $B_{\rm ext}$ is attributed to a sign change of $I_{\rm h}$.

Figure 1(b) shows the dephasing rates as a function of $B_{\rm ext}$. The decay rates were deduced from fitting Eq. (1) to $\Theta_{\rm K}$ in a time window $100 < \Delta t < 2500$ ps. After a flat region for small $B_{\rm ext}$, $1/T_2^*$ increases about linearly with $B_{\rm ext}$, which is typical for an inhomogeneous broadening of the g factor.[20] A fit of the data in Fig. 1(b) yields a slope of $k_2 = 2.2 \times 10^8$ s^{-1} T^{-1}. Also $1/T_1$ increases with $B_{\rm ext}$ with a slope of $k_1 = 5.7 \times 10^7$ s^{-1} T^{-1}.

To study the initialization at low $B_{\rm ext}$ and the sign reversal, $\Theta_{\rm K}$ was measured at fixed time delay $\Delta t = 12.4$ ns as a function of $B_{\rm ext}$ [see Fig. 2(a)]. The measured curves are well described by Eq. (1), that is, a $B_{\rm ext}$-dependent cosine function, offset by a nonoscillating part. Note that if T_2^* is comparable or longer than the laser pulse repetition period of 12.5 ns, the spin polarization created by the previous pump pulses becomes important[21] and the oscillations in $B_{\rm ext}$ deviate from a cosine shape. This is observed in our samples only at $B_{\rm ext} < 0.2$ T, in agreement with the data shown in Fig. 1(b). The sign change of I_h is very well resolved: In Fig. 2(a) we measure dips for $|B_{\rm ext}| < 0.2$ T at every integer spin rotation, whereas for $|B_{\rm ext}| > 0.3$ T we measure peaks. Between these two regimes there is a magnetic field $B_{\rm ext,0}$, where $\Theta_{\rm K} = 0$ (marked with a dot and a dashed line). At this field, the initialization of hole polarization is ineffective; that is, $I_{\rm h} = 0$. For the specific parameters $\lambda = 741.2$ nm and $P_{\rm P} = 0.2$ mW of the data shown in Fig. 2(a), we find $B_{\rm ext,0} \approx 0.25$ T. This is close to the value where T_2^* matches the laser repetition period [see Fig. 1(b)]. One could therefore think that the sign change of I_h is related to resonant spin amplification. We exclude that this is the case since $B_{\rm ext,0}$ varies strongly with $P_{\rm P}$ or λ (see below), whereas T_2^* only marginally depends on these parameters. In addition, the presence of a hole-spin polarization at the time when a new laser pulse arrives cannot lead to a sign inversion of I_h, but only to a saturation at large spin polarizations.

Figure 2(b) shows Kerr rotation data vs $B_{\rm ext}$ for different $P_{\rm P}$ (left panel) and λ (right panel). The curves are offset for clarity, and the dashed line indicates $\Theta_{\rm K} = 0$ for every curve. The

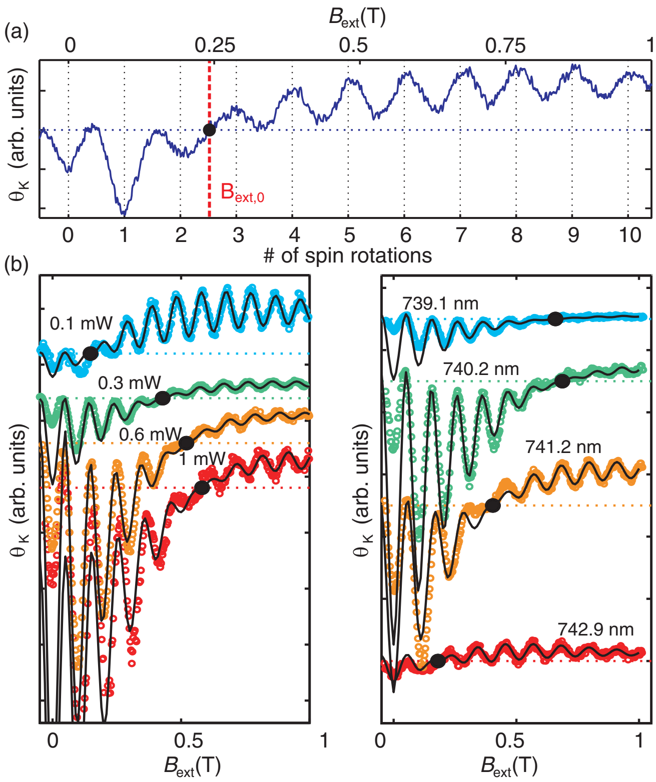

FIG. 2. (Color online) Kerr rotation measurements vs B_{ext} at $\Delta t = 12.4$ ns using a probe power of 10 μW. (a) $\lambda = 741.2$ nm and $P_{\mathrm{P}} = 0.2$ mW. The vertical line and the dot mark the value of B_{ext} where $I_{\mathrm{h}} = 0$. (b) (Left) $\lambda = 741.2$ nm and varying P_{P}; (right) $P_{\mathrm{P}} = 500$ μW and varying λ. For all measurements in (b) the fits are superimposed as solid lines.

black dots mark $B_{\mathrm{ext},0}$. We find a trend toward higher values of $B_{\mathrm{ext},0}$ for increased P_{P} or lower λ. In order to understand and fit the measured curves we now proceed to calculate I_{h}.

III. DISCUSSION

The dynamics of the initialization process involves the transfer between the four states depicted in Fig. 1(c). Ignoring a possible small polarization from the previous pump pulse, localized hole spins have the same probability to be in an up or in a down state before the arrival of a pump pulse, $p(|\Uparrow\rangle) = p(|\Downarrow\rangle) = 0.5$. Without loss of generality, we assume a σ^- pulse to arrive at $\Delta t = 0$. Due to optical selection rules, the pulse pumps $|\Uparrow\rangle$ holes into $|\Downarrow\Uparrow\uparrow\rangle$ trions with a probability p_σ. We assume pure heavy-hole states and neglect any interaction of the pump with light holes. This is justified since light holes are sufficiently far away in energy. For $B_{\mathrm{ext}} = 0$, these trions decay with the carrier decay rate $1/\tau_{\mathrm{R}}$ into the original spin state with no resulting polarization. Since $\tau_{\mathrm{R}} \ll T_2^*, T_1$, the hole spin decay can be neglected for calculating I_{h}. For $B_{\mathrm{ext}} \neq 0$, the trion spin precesses with a frequency Ω_e, whereas the spin of the localized holes precesses with a lower frequency (Ω_h) due to the smaller g factor. The recombination of such trions results in a positive I_h that increases with B_{ext}.[7]

In the experiment, we observe a $|\Uparrow\rangle$ polarization for $B_{\mathrm{ext}} = 0$, that is, a negative I_h. This observation is explained by the process X that pumps $|\Downarrow\rangle$ holes into $|\Downarrow\Uparrow\uparrow\rangle$ trions with probability p_X. Both processes (σ and X) pump localized holes into the $|\Downarrow\Uparrow\uparrow\rangle$-trion state [see Fig. 1(c)] and the total occupation probability of this state is given by $p_\sigma p(|\Uparrow\rangle) + p_X p(|\Downarrow\rangle) = 0.5[p_\sigma + p_X]$. For $B_{\mathrm{ext}} = 0$ and finite p_X, this results in a negative Kerr signal after the decay of the trions.

The system shown in Fig. 1(c) is described by an analytically solvable system of rate equations that include the precession of the spins, the dephasing of the holes, and the decay of the trions.[7] After recombination of the trions within a time scale of τ_R, the spin polarization of localized holes is described by Eq. (1) with

$$I_{\mathrm{h}} = \frac{p_\sigma}{2}\left[1 - \frac{p_X}{p_\sigma} - \frac{1 + \frac{p_X}{p_\sigma}}{\Omega_{e-h}^2 \tau_R^2 + 1}\right]. \tag{2}$$

In Eq. (2), $\Omega_{e-h} = |g_e - g_h|\mu_B B_{\mathrm{ext}}/\hbar$. We assume that p_σ and p_X are independent of B_{ext} and that the transition X is fast compared to τ_R. For $B_{\mathrm{ext}} = 0$, we recover $I_{\mathrm{h}} = -p_X$; that is, the spin-polarization is only determined by the X process. For finite p_X, I_h has a zero point at finite magnetic field ($B_{\mathrm{ext},0}$). From this point, the ratio of the two pump probabilities can be calculated

$$\frac{p_X}{p_\sigma} = \frac{\Omega_{\mathrm{e-h},0}^2 \tau_{\mathrm{R}}^2}{\Omega_{\mathrm{e-h},0}^2 \tau_{\mathrm{R}}^2 + 2}, \tag{3}$$

with $\Omega_{\mathrm{e-h},0} = |g_e - g_h|\mu_B B_{\mathrm{ext},0}/\hbar$.

For the next step, we use Eqs. (1)–(3) to fit the curves in Fig. 2(b). The spin lifetimes are modeled by $1/T_1 = 1/T_{1,0} + k_1 B_{\mathrm{ext}}$ and $1/T_2^* = 1/T_{2,0}^* + k_2 B_{\mathrm{ext}}$. The measured parameters are $g_{\mathrm{e}} = 0.34$, $\tau_{\mathrm{R}} = 80$ ps, and $B_{\mathrm{ext},0}$. The latter varies with P_P and λ, and determines through Eq. (3) the ratio p_X/p_σ. For each value of P_{P} and λ, fit parameters k_1, k_2, g_h, c_1, and c_2 are determined, where $c_1 = A p_\sigma a_1 e^{-\Delta t/T_{1,0}}$ and $c_2 = A p_\sigma a_2 e^{-\Delta t/T_{2,0}^*}$ are the B_{ext}-independent amplitudes of the nonoscillating and oscillating parts of the signal. The assumed linear increase of the spin relaxation rate with B_{ext} overestimates the Kerr signal below 0.2 T. To account for this we weighted the least-squares residuals with B_{ext}^2 for fitting the curves in the range between 0 and 1 T. The solid black lines in Fig. 2(b) are the resulting fits. The agreement between the data and the fit is very good for $B_{\mathrm{ext}} > 0.2$ T.

The fit parameters as a function of P_{P} are displayed in Fig. 3(a). The values for k_1 and k_2 match the results shown as a cross that were obtained from measurements of Θ_{K} versus Δt [see Figs. 1(a) and 1(b)]. This demonstrates that the field-dependence of Θ_{K} seen in Fig. 2 is well described by our model. Only small changes on the fit parameters are observed as a function of P_P, reflecting the notion that the pump-power dependence of I_h is the main cause for the difference between the curves in Fig. 2. For increased P_P, g_h decreases by almost 10%, which could be attributed to the high sensitivity of g_h on changes in the electrostatic confinement.[6]

From the values of $B_{\mathrm{ext},0}$, we derive the ratio p_X/p_σ. The points plotted in Fig. 3(b) are an average of the two values of $|B_{\mathrm{ext},0}|$ obtained from a magnetic-field sweep from -1 to 1 T. The black dots show the ratio as a function of P_{P} for $\lambda = 741.2$ nm. The ratio increases and saturates at a value of 0.25 for $P_{\mathrm{P}} > 0.5$ mW. By changing the wavelength and

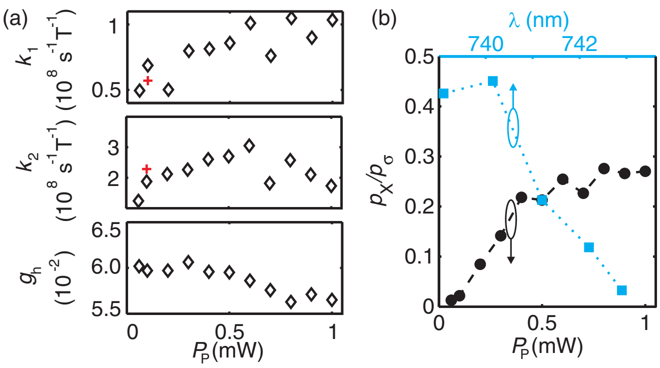

FIG. 3. (Color online) (a) Parameters of the fits vs pump power P_P as described in the text (diamonds) and for comparison fit results obtained from the data shown in Fig. 1(b) (crosses). (b) Ratio of the two pumping mechanisms p_X and p_σ as a function of P_P (dots) and λ (squares).

keeping the pump power at 0.5 mW, even larger ratios can be obtained. For λ outside the range of the data shown in Fig. 3(b), no hole signal was detected, which we relate to a decrease of the Kerr sensitivity A. The obtained p_X/p_σ ratios depend on τ_R and would be higher if τ_T were not limited by the recombination. Note that in our experiment, we cannot distinguish between direct excitation of a $|\Downarrow\Uparrow\uparrow\rangle$ trion and the capturing of a bright exciton by a $|\Uparrow\rangle$ hole. Both possibilities are included in p_σ.

With the two pumping processes show in Fig. 1(c) we can qualitatively explain the data in Fig. 3(b). The trion formation with probability p_σ requires the availability of resident holes. With increasing P_P, the average probability that the optically excited electron-hole pairs can capture $|\Uparrow\rangle$ holes decreases, and the probability for a spin flip of the optically excited holes and subsequent capturing of still available $|\Downarrow\rangle$ hole spins increases. This may explain the increase of p_X/p_σ with pump power. The saturation at $P_P = 0.5$ mW is governed by the rates describing the capturing and the spin-flip processes of the excitons. The finite number of localized holes also limits p_X for very large P_P. A shorter λ increases the p_X/p_σ ratio for the same reason: The absorption of photons increases for shorter λ, and more electron-hole pairs are generated at the same pump power. In addition, a more pronounced hole dephasing for nonresonantly pumped excitons[15] may favor process X with decreasing λ.

The oscillation in the data of Θ_K at zero field in Fig. 2(b) has a smaller amplitude than the fits. From the larger spin lifetime and the resulting resonant amplification one would expect the opposite effect. We suspect that this deviation is a result of the limitations of our model that assumes p_X to be independent of B_{ext} and neglects the time evolution of the excitonic states. The smaller peak is compatible with assuming that the formation of dark excitons is suppressed for small B_{ext}. A reason for this could be the spin dynamics of the excitons under the combined influence of both an external field and the anisotropic exchange splitting.[22,23]

To conclude, we find that localized hole spins can be spin polarized by exploiting the difference in g factor between holes and trions or via the capturing of dark excitons. These two mechanisms lead to the initialization of spin polarization of opposite signs, and their relative strength can be controlled by the magnetic field, pump power, and wavelength of the pump pulses. By changing these parameters, the size and sign of the spin polarization of localized holes can be controlled.

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