Excitation energy dependence of initial phase shift in Kerr rotation of resident electron spin polarization in a CdTe single quantum well

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The generation dynamics of a resident electron spin polarization involve the formation and transformation of the associated optically excited states depending on the excitation photon energy. Initial phase shift in the precession of a resident electron spin polarization gives the important clues to reveal the interplay between the associated excited states and resident electrons. In this work, the excitation energy dependence of the initial phase shift in Kerr rotation of a resident electron spin polarization is investigated in a CdTe/Cd\textsubscript{0.85}Mg\textsubscript{0.15}Te single quantum well under a low magnetic field. Through the careful analysis of the spin precession, the excited electron heavy-hole pairs show the unique spin dynamics when the excitation energy is increasing past the neutral exciton resonance. Meanwhile, the negative initial phase shift becomes significantly large in magnitude. The larger initial phase shift and the unique precession traces are caused by the transformation from neutral excitons to negative trions with accompanying the hole spin-flip. The exciton-to-trion transformation time is estimated experimentally to be \(\sim 160\) ps.

1 Introduction

Resident electron spin polarization (RESP) in semiconductor nanostructures has attracted considerable attention as one of key components in potential application of spin degree of freedom to electro-optic devices and quantum information processing [1–4] as well as in fundamental academic studies. Compared to the charge based devices, the spin-based electronics or the semiconductor spintronics have the potential to yield new device functionalities, to overcome the shortcoming of heating and power consumption, and to improve the performances with non-volatility and the increased data processing speed in future applications. These applications could benefit from a long spin coherence of the RESP. Because the RESP can exist after the recombination of the associated optically excited states such as negatively charged excitons (trions) and the spin coherence is not limited by the fast recombination process.

Importantly, the initialization of the RESP by resonant absorption of circularly polarized pulses is essentially dictated by the instantaneous formation of a trion state \((T)\), which is a negatively charged exciton which consists of two electrons with the paired spins (singlet state) and one hole [5]. In the past several years, the important role played by trions for the properties of RESP in the quantum wells (QWs) has been highlighted [6,7]. It is also important to have the correct scenario of the role of neutral excitons \((X^0)\), which allows for better control and study of the trion spin dynamics and RESP formation [5,8]. However, rather few groups have investigated the action of the photo-created electron-heavy holes \((e\text{-}hh)\) pairs on RESP in QWs.
systematically. The effects of the excited e-hh pairs are easy to be neglected due to their short lifetime [9]. However, we cannot rule out the potential influence of e-hh pairs on RESP, even though the exciton formation and transformation to trion make the RESP dynamics rather complicated. We have observed and analyzed the initial phase shift (IPS) phenomenon of the time-resolved Kerr rotation (TRKR) signals of the RESP component at time origin (t=0), as shown later (Fig. 1) [10]. Since the IPS gives the important clues concerning the generation processes of RESP, we consider that the experimental and theoretical studies of IPS play a crucial role in the study of RESP.

In this work, we investigate mainly the distinct negative IPS in a temporal evolution of the RESP with different photon energy in a CdTe single QW (SQW) using a TRKR technique. CdTe consists of the isotopes with I_p = 1/2, p_Cd = 25%, I_Te = 1/2, p_Te = 8% and the other isotopes with I_Cd,Te = 0, where I_p, p_i are nuclear spin and natural abundance of the isotope i, respectively. Therefore, CdTe is one of the attractive compounds in terms of smaller effects of hyperfine interaction and quadrupolar interaction induced by strain due to small abundance of the isotopes with one-half nuclear spin, compared to III-V semiconductors such as GaAs and InAs [11].

Both the exciton spin relaxation and the transformation from X^0 to T have been taken into account in our model, and the transformation time could be deduced from the measurements. Through the analysis and interpretation of RESP temporal evolutions, the contribution of the e-hh pairs to RESP dynamics has been estimated under a low applied magnetic field.

2 Experimental procedure The sample used in this study is a nominally undoped CdTe/Cd_{0.85}Mg_{0.15}Te SQW with a well width of 100 Å grown by molecular-beam epitaxy on a (100)-oriented GaAs substrate. The resident electrons are supposed to be localized in the well with a density of n_e = 2 \times 10^{10} \text{cm}^{-2}, which was estimated from the relation between X^0 and T oscillator strength in the reflectivity spectra [12]. The resident electrons can interact with optically injected e-hh pairs to form negative trion states. The n-type doping and the formation of negative trions were confirmed by TRKR measurements [10, 13].

Figure 1 shows the photoluminescence (PL) spectra measured under a continuous excitation with a photon energy of 1.650 eV at 10 K. The PL spectra can be fitted well by the sum of two Lorentz functions, corresponding to the negatively charged trion resonant transition T (at the low energy peak: 1.6110 eV) and the neutral exciton resonant transition X^0 (at the high energy peak: 1.6133 eV). Those are clearly separated by the trion binding energy of 2.3 meV, which is beneficial to the selective excitation of trions or excitons. The trions are generated through the lowest-energy optical transition in the system. From the independent time-resolved PL measurements using a streak scope, the recombination times of T and X^0 were found to be relatively short (~85 ps and ~89 ps, respectively).

In this experimental report, the dynamical evolution of the created RESP and the interplay with trions and excitons were investigated mainly via the excitation energy dependence using the TRKR technique. A circularly polarized pump pulse was incident along the QW growth direction (z) and the created carrier spin polarization was monitored. A magnetic field was applied in the x-direction (Voigt geometry). In this case, the spin polarizations were prepared orthogonal to the applied field and started to precess with Larmor angular frequency \( g_0 \mu_B B / \hbar \) in the y-z plane immediately after optical initialization. Here, \( g_0 \) is an inplane electron g-factor and \( \mu_B \) is a Bohr magneton (~58 \mu_0 \text{eV/T}). The value for \( g_0 = -1.468 \pm 0.003 \) of this sample was found in our previous study [11]. Since the pump and probe beams are in the x-z plane that is orthogonal to the spin precession plane (y-z plane), any extrinsic initial phase shifts of the electron spin precession, for instance, due to an oblique incidence of the pump pulse [14], do not arise.

Both pump and probe pulses were supplied by a mode-locked Ti: sapphire oscillator with a pulse width of ~2 ps and a repetition rate of 76 MHz. The spectral width of the pump and probe pulses (<1 meV) was narrower compared to the trion and exciton spectra as shown in Fig. 1. Similar to a standard TRKR setup, the linearly polarized probe pulse with a controlled delay time samples the carrier spin polarization in the central area excited by a pump pulse. A Kerr rotation of the probe polarization was measured sensitively by using a polarization bridge and double lock-in.
trion (↑↓↑) with an angular momentum of -3/2. At this moment, the resident electron ensemble loses the spin balance. Meanwhile, a nonzero net RESP comes out along the light propagation direction. After the trion recombination (the recombination time \(r_T \sim 85\) ps), the remaining electrons return to the resident electron ensemble and can change RESP ((II) in Fig. 2). Depending on the hole spin-flip time, the trion lifetime, and the precession period (Larmor frequency), the amplitude and phase of the TRKR signals originated from RESP may change intricately \([10]\). Since the lifetime of the trions is less than 100 ps, the oscillatory dumping component with a long coherence in the TRKR signal can confidently be associated with RESP. Note that the negative trions indicate a simple decay due to the short lifetime without a spin-precession since the total in-plane g-factor of the negative trion is zero in a QW. Also, since the pulse width (~2 ps) is much shorter than the precession period of RESP, the spin rotation during the pulse can be neglected. The generation process in the case of trion resonant excitation and the first observation of the negative IPS are detailed in the previous study \([10]\).

Under the existence of excitons, the electrons of excitons should affect the TRKR signals. As a result, a typical experimental TRKR signal \(S_z(t)\) can be described generally by the following function:

\[
S_z(t) = A_T \exp(-t/r_T) + A_X, h \exp(-t/r_X,h) \\
+ A_X, e \exp(-t/r_X,e) \cos(\omega_L t + \phi_0) \\
+ A_e \exp(-t/r_e) \cos(\omega_L t + \phi),
\]

where the coefficients \(A_T\), \(A_X, h\), and \(A_e\) are the amplitudes of trions’ holes, excitons’ electrons (holes), and RESP; \(r_T\), \(r_X, h\), and \(r_e\) are the corresponding total spin lifetimes; \(\omega_L\) is the electron Larmor angular frequency; \(\phi_0\) is the relative phase shift of the exciton precession, and \(\phi\) is the IPS of the RESP precession. The total spin lifetime of the electron-in-exciton \(r_X, c\) includes the recombination time \(r_T\), the time of exciton binding into a trion \(r_c\), and the spin relaxation time \(r_X, c\) (i.e., \(1/r_X, c = 1/r_T + 1/r_c + 1/r_X, e\)). Since there is no change of the trion complexes to excitons, the excitons could be generated only at the very time of the photon excitation \((t=0)\). Therefore we can conclude that \(\phi_0=0\). It is noted that the first, second, and third terms in Eq. 1 disappear rapidly due to their short lifetimes (~10 ps, ~10 ps, and <100 ps, respectively). The evolution of the first three terms is difficult to capture, while the fourth term remains over the range of a few ns. In the case of Fig. 2, the exciton component is negligible since the QW was excited selectively at the trion resonant transition. Thus, the TRKR signal can be fitted safely by the fourth term of Eq. 1 over the time range of \(t=0.3-3\) ns. For the fitting curve, the extrapolating back to zero time delay \((t=0)\) can be seen as the time delay of RESP generation or the temporal shift \(\phi/\omega_L\) (~19.6 ps) of the time origin as indicated in the inset (III) of Fig. 2. The temporal shift can be converted to IPS=0.47 rad. Therefore, the initial phase \(\phi\) of the RESP precession plays out a
Figure 3 (Color Online) (a) TRKR signals for different excitation energy (189 mT, 10 K). Two traces with the thick lines correspond approximately to the trion and exciton resonant conditions, respectively. (b) A typical TRKR signal (blue) and the fitting curve of the electron spin precession (red) at 1.6140 eV (189 mT, 10 K). (c) A TRKR signal (green) at 1.6140 eV (0 mT, 10 K).

Figure 4 (Color Online) The excitation photon energy dependence of IPSs. The solid line is a guide for eyes. The blue and green dashed lines are used to mark the resonant transition energies of trion (T) and neutral exciton (X^0), respectively. The upper figure is the PL spectra at 10 K.

3.2 Excitation energy dependence of IPS

In this report, we have performed the TRKR measurements when the excitation photon energy was scanned from 1.6059 eV (lower than the trion transition) to 1.6164 eV (higher than the exciton transition). Some of the TRKR signals are shown in Fig. 3 (a). For an easy view, the time origin (dashed line) of each signal is shifted in the figure. According to these results, we can see the contribution features of the trions and excitons to IPS of RESP over a large range of the pumping photon energy. The maximum amplitude of the RESP signal was obtained at the trion resonance. Seen from the figure, some unusual features in the range of 0-200 ps have been observed around the exciton resonance, where there is an extremely fast downward decay and an upswing.

An example of the unusual TRKR signal measured with a pump power of 8 mW at 10 K in B_x=189 mT is displayed in Fig. 3 (b) (not shown in Fig. 3 (a)) and its fitting curve by the third and fourth terms of Eq. 1 is added in the range of 0.3-3 ns. Here, the excitation energy was tuned to 1.6140 eV which was a little higher than the exciton resonant transition energy. No directly-generated trion can be assumed due to the exciton resonant excitation with a low power excitation. For all the analyses below, the condition 2π/ω_L > τ/τ_e is applicable. As shown in Fig. 3 (b), there is a clear fast drop close to the time origin (marked by an arrow) in this typical signal. The excited initial signal (X^0:↑↓) by a σ^- pulsed excitation is defined as positive (S_t = S_y > 0) at t = 0 (thus, S_L < 0 and S_E < 0). The signal goes down to below the balanced level largely from the initial spin polarization within tens of picoseconds. That is due to the spin precession of the electron-in-exciton (partly from ↑↑ to ↓↓) and a quick spin asymmetric relaxation of the heavy hole-in-exciton (from ↓↓ to ↑↑) which happens in trions as discussed in the former report [10]. After the point marked by an arrow, the TRKR signal rises again gradually due to the recombinaction of the transformed trions and excitons. Concretely speaking, the recombinaction of trions returns the electrons back to the resident electron ensemble to create additional RESP at later times. The subsequently created RESP induces a negative IPS and raises the spin polarization integrated throughout the entire trion lifetime.

To extract the time and relative amplitude of the short-living precession component in the signal, the oscillatory part of this TRKR trace has been fitted. Then, the relative amplitude A_{X}(1.6140 eV)(~84.2) could correspond to the relative number of photo-generated excitons with a decay time τ_{X}X[1.6140 eV](~57.6 ps). Under the same excitation condition, another TRKR signal obtained after removing the transverse magnetic field B_x is shown in Fig. 3 (c). The total relative number of resident electrons after that all excitons decay to trions should be estimated as its fitted long-living RESP. Therefore, A_{e}(1.6140 eV)(~15.3) was obtained from the fitting by S_z(t) = A_{X}exp(-t/τ_{X}) + A_{e}exp(-t/τ_{e}) for the range of 0.6-3 ns in Fig. 3(c). Then, the ratio between the exciton-to-trion transformation time τ_c and the electron-in-exciton spin lifetime τ_{X,e} would be
simplicity, the heavy hole spin dynamics and the spin-flips electron spin after trion generation (Fig. 5 (b)). For the between the electron-in-exciton spin (Fig. 5 (a)) and resident excitation. Figure 5 shows the different spin evolution be-
as to create the later additional RESP.

During the interval $\Delta t$, both the electron-in-exciton spin polarization and the RESP (the hollowed arrows in Fig. 5 (a) and (b), respectively) experience the precession by the phase of $\omega_{1}\Delta t$ under the transverse magnetic field. Therefore, the spins of the electron-in-all excitons would be composed of the $\uparrow$ and the $\downarrow$-components. At the same time, the heavy hole-in-exciton spins have been asymmetrically flipped ($\uparrow \rightarrow \downarrow$) as do trions in this QW [10]. Since the holes have no spin pre-

Finally, we summarize the RESP dynamics and the ori-
gin of the larger absolute IPS in the case of the exciton excitation. Figure 5 shows the different spin evolution be-
tween the electron-in-exciton spin (Fig. 5 (a)) and resident electron spin after trion generation (Fig. 5 (b)). For the simplicity, the heavy hole spin dynamics and the spin-flips of electron are skipped in this diagram. The electron-in-exciton spin and the electron-in-trion spin are in the circles with the solid and dashed lines, respectively.

In the figure, we describe that the excitons and trions are excited at the same time origin ($t=0$) corresponding to the exciton resonant and trion resonant excitations, respectively. In the case of the trion excitation (Fig. 5 (b)), the same amount of spin polarization $P_0$ for RESP and trions are created instantly after the excitation [10]. On the other hand, in the case of the photo-generated neutral excitons (Fig. 5 (a)), a certain time interval of $\Delta t$ ($\ll \pi/2\omega_{1}$) is necessary in order to affect the RESP since the excitons have to capture resident electrons with a proper spin ori-

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**Figure 5** (Color Online) Simplified electron spin dynamics: (a) The first RESP formation from the photo-generated excitons; (b) The first RESP under trion resonant excitation. The electron-in-exciton spin and the electron-in-trion spin are in the circles with the solid and dashed lines, respectively.

Estimated to be [16]

$$\tau_e / \tau_{X;e} = A_{X;e} / 2A_e = 2.76.$$  \hspace{1cm} (2)

Assuming that $\tau_{X;e}$, and using $1/\tau_{X;e} = 1/\tau_e + 1/\tau_{X} + 1/\tau_{X}^\uparrow$ with the above relation, we work out the exciton-to-trion transformation time $\tau_e \sim 157$ ps and electron-in-exciton spin lifetime $\tau_{X;e} \sim 57$ ps, which agrees well with the fitting parameter above ($\tau_{X;e} \sim 57.6$ ps). In the past work, the direct transformation from ex-

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**4 Conclusion** In summary, we systematically investigat-
ed the negative IPSs in the TRKR traces of RESP under a low magnetic field. According to these results, the significant role played by $e$-$hh$ pairs on the IPS has been indicated. Even there is no substantial change in the IPSs induced by trion spin dynamics under the low intensity excitation. By extending the excitation energy beyond the trion transition regime, the absolute value of negative IPS becomes larger and larger with increasing the excitation energy. This is because the newly formed RESP renews the spin flips to the beginning after the transformation from $X^0$ to $T$. In ad-
dition, the transformation time from \(X^0\) to \(T\) was deduced from the careful analysis of TRKR signals. More importantly, these clear effects of excitons and the dynamic physical picture of IPS are important for studying and controlling the electron spin dynamics in semiconductor nanostructures.

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