Spin dynamics of electrons in the first excited subband of a high-mobility low-density two-dimensional electron system

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We report on time-resolved Kerr rotation measurements of spin coherence of electrons in the first excited subband of a high-mobility low-density two-dimensional electron system in a $GaAs/Al_{0.35}Ga_{0.65}As$ heterostructure. While the transverse spin lifetime (T_2^*) of electrons decreases monotonically with increasing magnetic field, it has a nonmonotonic dependence on the temperature and reaches a peak value of 596 ps at 36 K, indicating the effect of intersubband electron-electron scattering on the electron-spin relaxation.

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Electron-spin manipulation is among the hottest topics in semiconductor spintronics research, which aims to realize next-generation devices via utilizing the spin degree of freedom of electrons.[1–3] Due to its important role in achieving effective spin manipulation, the spin dynamics of carriers in semiconductors has been extensively studied in last decades via many kinds of spin-sensitive spectroscopy techniques.[4–7] One of such techniques is the time-resolved Faraday/Kerr rotation (TRKR) measurement: it has been widely used to experimentally study spin dynamics in bulk materials[8,9] and quantum-confined system such as quantum wells[10,11] and a single-quantum dot.[12,13]

Most experiments to date are concerned with spin coherent processes of electrons in the ground state (GS). However, there is not much research on the spin dynamics of electrons in excited states of quantum-confined system so far.[14–16] Here we report on experimental investigation of the spin coherence of electrons in the first excited subbband (FES) of a two-dimensional electron system (2DES) with high mobility and low density. With TRKR technique, we measured both the magnetic field dependence (0–4 T) and the temperature dependence (1.5–80 K) of the transverse spin lifetime (T_2^*) of the electrons. While T_2^* decreases monotonically with an increasing magnetic field, it has a nonmonotonic dependence on the temperature and reaches a peak value of 596 ps at 36 K (magnetic field $B=1.5$ T), indicating the effect of electron-electron scattering on the electron-spin decoherence[17–19] while in this case the intersubband scattering is more important than the intrasubband scattering.

The sample is a modulation-doped $GaAs/Al_{0.35}Ga_{0.65}As$ 2DES sample with a mobility of $\mu=3.2\times10^6$ cm^2/Vs and an electron density of $n_{2D}=9.6\times10^{10}$ cm^{-2} at $T=4.2$ K.[18] The 2DES sample was mounted inside an optical cryostat with a tunable temperature from 1.5 to 300 K and a transverse magnetic field up to 10 T provided by a superconducting split coil. Photoluminescence (PL) spectrum measurement was performed with a He-Ne laser at temperature $T=2$ K and magnetic field $B=0$ T to determine the energy levels of the 2DES.

Figure 1(a) shows the PL spectrum at 2 K, which has two peaks at $E=1.519$ eV and $E=1.578$ eV. The PL peak at $E=1.519$ eV consists of a low-energy interface-exciton recombination and a high-energy bulk free-exciton band.[20] The peak at $E=1.578$ eV is attributed to the recombination of holes and electrons in the FES of the 2DES. The energy levels of the 2DES is then schematically drawn in Fig. 1(b).

After determining the energy levels of the 2DES, we performed TRKR measurements to study the dynamical processes of spin-polarized electrons in the FES of the sample. The sample was excited near normal incidence with degenerate pump and probe beams from a mode-locked Ti: sapphire pulsed laser, which has a pulse width of 150 fs, a repetition rate of 76 MHz and a tunable wavelength between 700 and 980 nm. The pump and probe beams were focused to a spot of ~100 μm in diameter. The circular polarization of the pump beam was modulated with a photoelastic modulator at 50 kHz for lock-in detection. With the magnetic field being perpendicular to the laser beams (Voigt geometry), the Kerr rotation θ (Δt) of the linearly polarized probe light pulse after a time delay Δt measures the projection of the net spin polarization along the direction of the light as it precesses about the direction of the magnetic field. The TRKR measurement of the spin dynamics of electrons in the FES was performed with photon energy $E_p=1.578$ eV and pump/probe power of 10 mW/1 mW.[21] The photoexcited electron density was about 8×10^{10} cm^{-2} (Ref. 22).

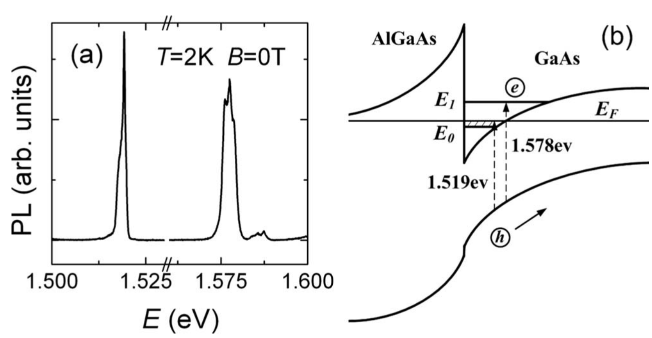

FIG. 1. (a) PL spectrum at $T=2$ K and $B=0$ T. (b) Schematical energy levels of the 2DES (not to scale). The two broken line with arrows indicate, respectively, the excitation with $E=1.519$ eV and $E=1.578$ eV.

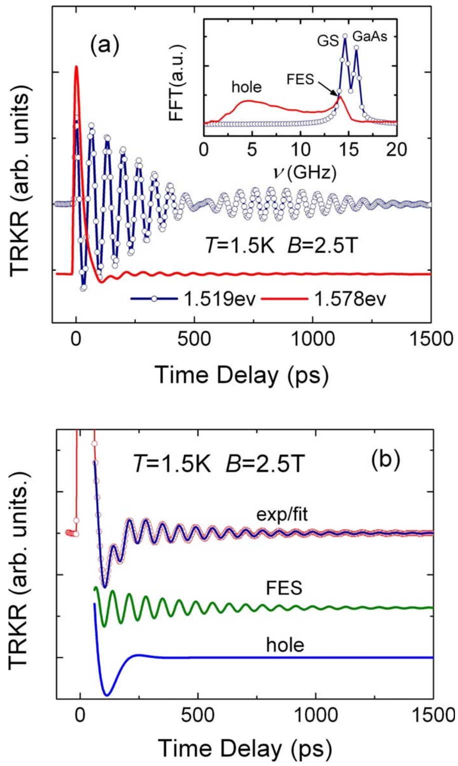

FIG. 2. (Color online) (a) TRKR traces were measured, respectively, with $E_p=1.519$ eV and $E_p=1.578$ eV at $T=1.5$ K and $B=2.5$ T. Inset: FFTs of the corresponding TRKR traces. (b) Top: the experimental TRKR trace with $E_p=1.578$ eV at $T=1.5$ K and $B=2.5$ T and the fitting curve (the solid line); middle: the extracted TRKR signal of the electrons in the FES; bottom: the extracted TRKR signal of the holes.

Figure 2(a) shows two TRKR traces measured at $T=1.5$ K and $B=2.5$ T with $E_p=1.519$ eV and $E_p=1.578$ eV, respectively. Both of them have beating features, as indicated by the two peaks in the fast Fourier transform (FFT) spectra in the inset of the Fig. 2(a). The low-frequency part in the $E_p=1.519$ eV trace is attributed to electrons in the GS and the high-frequency part is attributed to electrons in the bulk GaAs.[18] As for the $E_p=1.578$ eV case, the high-frequency part is ascribed to the spin precession of the electrons in the FES of the 2DES while the low-frequency part is attributed to the photoexcited free holes.[24] The Larmor precession frequency of the electrons in the FES is slightly samller than that in the GS because the wave function of the electrons in the FES penetrates more into the potential barrier $Al_{0.35}Ga_{0.65}As$ than that of the GS. Since the g factor of GaAs is -0.44 and the g facotr of $Al_{0.35}Ga_{0.65}As$ is $+0.5$, respectively,[25] the more spread of electron wave function is in the $Al_{0.35}Ga_{0.65}As$ barrier, the smaller the electron g factor is (in absolute value).

The FES TRKR trace at $E_p=1.578$ eV is redrawn in the top part of Fig. 2(b). The Kerr signal coming from electrons in the FES of the 2DES and the photoexcited holes can be decomposed via their distinct Larmor precession frequencies. We fit the experimental data with the following formula:

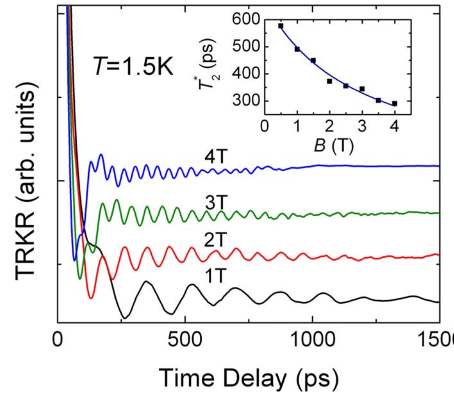

FIG. 3. (Color online) TRKR traces at different magnetic field (B) at $T=1.5$ K. Inset: the B dependence of the transverse spin lifetime of electrons in the FES (squares); the solid line is the fitting curve with Eq. (3).

$$\theta_k(\Delta t) = A_e \exp\left(-\frac{\Delta t}{T^*_{2,e}}\right)\cos(2\pi v_e \Delta t + \phi_e) + A_h \exp\left(-\frac{\Delta t}{T^*_{2,h}}\right)\cos(2\pi v_h \Delta t + \phi_h), \qquad (1)$$

where A_e (A_h) is the electron (hole) signal amplitude at pump-probe delay $\Delta t=0$, $T^*_{2,e}(T^*_{2,h})$ is the electron (hole) spin lifetime, v_e (v_h) is the Larmor precession frequency of the electron (hole), and ϕ_e (ϕ_h) is phase shift. The contributions from electrons in the FES and holes are shown, respectively, at the middle and bottom of Fig. 2(b). Their sum (the solid line in the upper panel) fits well to the experimental data. We then obtained the transverse spin lifetime T_2^* and the Larmor precession frequencies ν for both of the low- and high-frequency spin precession. The g factors of electrons in the FES and holes can be thus deduced by $\nu=g\mu_B B/h$, where μ_B is the Bohr magneton, B is the transverse magnetic field, and h is the Plank's constant. A spin lifetime of 355 ps (close to the result of other 2DES sample[26]) and a g factor of -0.405 can be obtained for the electrons in the FES. As for the holes, the spin lifetime is 49 ps and the g factor is 0.106 (Ref. 27). Hereafter we will mainly focus on the FES part.

Figure 3 shows TRKR signals at different magnetic field B at $T=1.5$ K. Fitting them with Eq. (1), we obtain the dependence of the T_2^* of the electrons in the FES on the magnetic field (see the inset). A long-lived electron-spin coherence with $T_2^*=577$ ps is found at $B=0.5$ T. It decreases monotonically to 290 ps when B increases up to 4 T.

Figure 4(a) shows TRKR traces at various temperatures at $B=1.5$ T.[28] As can be seen in this figure, with increasing temperature, the envelope of the oscillating Kerr signals decays more slowly from 1.5 to 36 K and it then decays slightly faster ever since (from 36 to 80 K). In order to figure out the temperature dependence of spin lifetime of the electrons in the FES, we measured the TRKR trace from 1.5 to 80 K in detail. Their spin lifetimes are shown in Fig. 4(b). A peak value of 596 ps is seen at 36 K in the electron-spin lifetime T_2^* and the maximum is superimposed on an

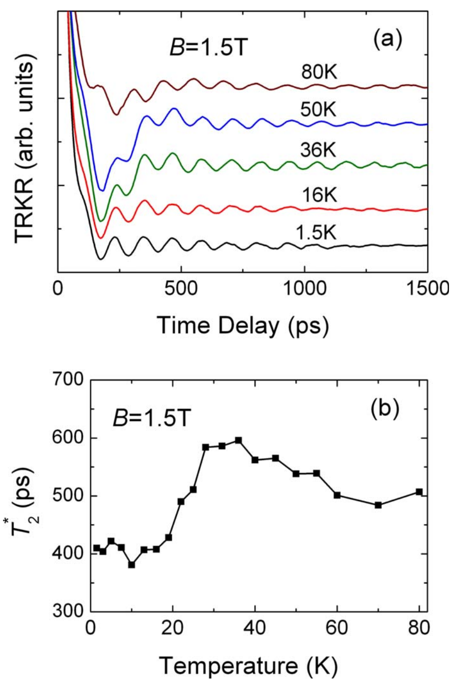

FIG. 4. (Color online) (a) TRKR traces at different temperatures (measured at $B=1.5$ T). (b) The temperature dependence of the spin lifetime of the electrons in the FES.

increasing spin lifetime background from 410 ps at 1.5 K to 507 ps at 80 K. The temperature dependence of the T_2^* of the electrons in the FES is similar to that in the GS of the 2DES.[18] However, the T_2^* of the electrons in the FES peaks at 36 K while that of the electrons in the GS peaks at 14 K (Ref. 18). The reason for the difference goes as the following.

There are two main contributions for the transverse spin lifetime of the electrons in the FES. One is the relaxation process in which the electrons in the FES relax into the GS or recombine with holes: it is an annihilation process of the electron in the FES which determines the electron-relaxation time T_1. The other is the phase smearing of electrons which remain in the FES: it determines the "pure" electron-spin dephasing time T_{22}^*. This may be expressed by the following formula:

$$\frac{1}{T_2^*} = \frac{1}{T_1} + \frac{1}{T_{22}^*} \tag{2}$$

T_1 is mainly determined by electron-phonon scattering, electron-impurity scattering, and electron-hole recombination. The high mobility of our 2DES sample ensures that the electron-impurity scattering is weak. The phonon-assisted relaxation is also weak since it is difficult for the LO and TO phonons to fulfill the conservation laws of both the momentum and the energy. The electron-hole recombination is also weak since optically generated holes are swept quickly into the GaAs buffer layer. A long T_1 can thus be expected: it could be even longer than 1 ns and does not depend much when the temperature is below 100 K.[29] Thus, the main contribution to the T_2^* dependence on the temperature (and/or the magnetic field) comes from T_{22}^*.

Some of the pump photons generate electrons and holes in bulk GaAs which then diffuse into the GaAs/$Al_{0.35}Ga_{0.65}As$ interface (in the ground state) and buffer layer, respectively. The GS electrons may affect the spins of FES electrons in three aspects. First, a higher spin polarization of the GS electrons may increase the spin lifetime of FES electrons: it comes from the Hartree-Fock contribution of the Coulomb interaction.[30] Second, an increasing GS electron density may increases the relaxation time T_1 in Eq. (2) and thus increases the spin lifetime of the FES electrons. Third, a higher density of 2DES (both in GS and FES) means a higher local electrical field perpendicular to the GaAs/$Al_{0.35}Ga_{0.65}As$ interface, a bigger spin-orbit coupling and thus a shorter spin lifetime.

Here may be the mechanism of the field dependence of T_2^*: since the electron density of the sample is very low, there may be puddles of electrons with different density, thus leading to an inhomogeneous g factor (like the case in localized two-dimensional holes and the electrons in quantum dots[10,31]) and a spread of Larmor precession frequencies in a transverse magnetic field, $\Delta\nu=\Delta g \mu_B B/h$, where Δg is the inhomogeneity of the electron g factor.[32] Note that with a fixed Δg, the higher the magnetic field, the faster the spin dephasing.[5] The relationship can be described by $1/T_{22}^*=1/T_{22}^*(0)+\Delta g \mu_B B \sqrt{2}\hbar$,[31] where $T_{22}^*(0)$ is the zero-field pure spin dephasing time. Equation (2) can be expressed as

$$\frac{1}{T_2^*} = \frac{1}{T_1} + \frac{1}{T_{22}^*(0)} + \frac{\Delta g \mu_B B}{\sqrt{2}\hbar}. \tag{3}$$

The solid line in Fig. 3(b) shows a $1/B$ fit to the T_2^* data, from which we get $T_1=1698$ ps and $T_{22}^*(0)=1100$ ps. The spin-relaxation rate determined by the D'yakonov-Perel' (DP) mechanism is $\tau^{-1}=\langle \mathbf{\Omega}(\mathbf{K})^2\rangle \tau_{\mathrm{p}}(\mathbf{k})$, where $\tau_{\mathrm{p}}(\mathbf{k})$ is the momentum relaxation time.[5] The electron-phonon scattering has a temperature dependence of $\tau_{\mathrm{eac}} \propto T^{-3/2}$ (Ref. 33). An increasing temperature leads to a stronger momentum scattering and a shorter momentum scattering time τ_{p}. This in turn induces an increasing T_2^* via the DP mechanism. Consequently, there is an increasing T_2^* background with rising temperature. In a high-mobility low-density 2DES, electron-electron Coulomb scattering dominates τ_{p} at low temperature. It contributes to the T_2^* peak at 36 K shown in Fig. 4(b) with similar arguments in previous theoretical and experimental work.[17–19] The peak is significantly shifted toward higher temperature, in contrast with the similar peak in our previous work on the electrons in the GS.[18] Such a difference may be due to the difference between the electron-electron scattering mechanism in these two cases. The intrasubband electron-electron scattering is dominant when the electrons are excited to the GS while the intersubband electroelectron scattering is domimant when the electrons are excited to the FES since the electron density of the FES is very low. The items of electron distribution function appears in the $1/\tau_{e\text{-}e}$

expression would be dramatically different in the two cases, inducing the different temperature dependence of $1/\tau_{e\text{-}e}$.[34]

In summary, we have experimentally studied the spin dynamics of electrons in the first excited subband in a high-mobility low-density 2DES. While T_2^* decreases monotonically with an increasing magnetic field at 1.5 K, it has a nonmonotonic dependence on the temperature in a magnetic field of 1.5 T and reaches a peak value of 596 ps at 36 K, indicating the effect of intersubband electron-electron scattering on the electron-spin decoherence.

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