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Tuning of the Electron Spin Relaxation Anisotropy via Optical Gating in GaAs/AlGaAs Quantum Wells *

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The carrier-density-dependent spin relaxation dynamics for modulation-doped GaAs/$Al_{0.3}Ga_{0.7}As$ quantum wells is studied using the time-resolved magneto-Kerr rotation measurements. The electron spin relaxation time and its in-plane anisotropy are studied as a function of the optically injected electron density. Moreover, the relative strength of the Rashba and the Dresselhaus spin–orbit coupling fields, and thus the observed spin relaxation time anisotropy, is further tuned by the additional excitation of a 532 nm continuous wave laser, demonstrating an effective spin relaxation manipulation via an optical gating method.

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Studying the electron spin dynamics and coherence in semiconductor nanostructures has evoked considerable interest, due to its potential applications in spintronic devices and quantum information processing technologies.[1] To achieve the novel spintronic devices, electron spin relaxation in many types of materials, especially in low dimensional Ⅲ–Ⅴ group semiconductor heterostructures, has been studied extensively both theoretically and experimentally.[2–5] It is generally believed that the electron spin relaxation in GaAs-based quantum wells (QWs) is governed by the D'yakonov–Perel' (DP) mechanism,[2,5,6] resulting from the spin–orbit coupling effect. In the QW structure there are mainly two kinds of spin–orbit coupling induced effective fields corresponding to different sources of structural inversion asymmetry: the Dresselhaus field caused by the bulk inversion asymmetry (BIA) and the Rashba field resulting from the structure inversion asymmetry (SIA).

In a two-dimensional electron system, the momentum along the growth direction is quantized due to the confinement with the electron energy expectation value proportional to the mean square of the electron momentum normal to the QWs plane $\langle k_z^2\rangle$ with $\langle k_z^2\rangle = (\pi/d)^2$, where d is the quantum well thickness, and k_x^2 and k_y^2 are smaller than $\langle k_z^2\rangle$. For the QWs grown along $z\|[001]$ direction, the effective spin–orbit coupling field $\boldsymbol{\Omega}(k)$ is a vector sum of the corresponding Dresselhaus and Rashba terms,[6–9]

$$\begin{aligned}\boldsymbol{\Omega}(k) &= \boldsymbol{\Omega}(k)_{\rm BIA} + \boldsymbol{\Omega}(k)_{\rm SIA} \\ &= \frac{\beta}{\hbar}(-k_x, k_y, 0) + \frac{\alpha}{\hbar}(k_y, -k_x, 0), \end{aligned} \quad (1)$$

where the Dresselhaus field $\boldsymbol{\Omega}(k)_{\rm BIA}$ is cubic in respect to the electron quasimomentum k, and the Dresselhaus parameter β is given by $\beta = \gamma\langle k_z^2\rangle$, γ is the spin splitting parameter, and α is the Rashba parameter.

It is obvious that the total effective field along the in-plane directions of [110] and $[1\bar{1}0]$ is different, and thus the spin relaxation will exhibit anisotropy along the two orientations, and the spin relaxation anisotropy can be tailored by tuning the relative strength of the Dresselhaus and the Rashba fields.

It was first predicted in theory by Averkiev *et al.* in zinc-blende heterostructures[10] and then it was confirmed experimentally through the steady Hanle effect method.[11] Since then, many experiments by means of time-resolved measurements have been carried out to study the spin relaxation anisotropy by manipulating the spin–orbit coupling. Among these experimental studies, electric gating manipulation is one of the common methods to be adapted to manipulate the electron density and Fermi energy, and thus, the strength of the effective Rashba spin–orbit field.[12–14] However, this method will involve the complex device processing and further degree of the structural inversion asymmetry breaking when tuning the background electron density in QWs.

In this Letter, an optical method is utilized to efficiently tune the background electron density instead of using an electric gating to avoid additional modification of the structural inversion asymmetry. The spin relaxation time and its in-plane anisotropy have been studied as a function of the optically injected electron density. The spin relaxation time anisotropy can be further tuned by the additional excitation of a 532 nm cw laser, demonstrating an effective method of spin relaxation manipulation via an optical gating method.

The sample used in our experiment is a δ-modulation doped multiple GaAs/AlGaAs QWs grown by molecular beam epitaxy on the [001]-oriented semi-insulating GaAs substrate, and the structure is shown in Fig. 1(a). A 200-nm-thick GaAs buffer layer is first grown on substrates followed by repeating 10 periods of the 156 nm AlGaAs barrier layer, and the 15 nm GaAs well layer, lastly a 20 nm

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GaAs cap layer is deposited on the top. The aluminum component for the AlGaAs barrier layer is 30% with Si δ-doping concentration about $1.4 \times 10^{13}\,\mathrm{cm}^{-2}$ at both sides. However, the δ-doping location is asymmetric, the up-side doping is located 70 nm away from the GaAs well layer, and the down-side doping is located at 86 nm away from the GaAs well layer. Our room-temperature Hall measurement indicates that the sheet carrier density for our designed sample is $6.8 \times 10^{12}\,\mathrm{cm}^{-2}$ at 300 K, and decreases to $1.6 \times 10^{12}\,\mathrm{cm}^{-2}$ when cooled down to 150 K. The carrier density of $\sim 10^{11}\,\mathrm{cm}^{-2}$ is thus expected, though the Hall results at 8 K are unavailable due to the bad Ohmic contact at low temperature.

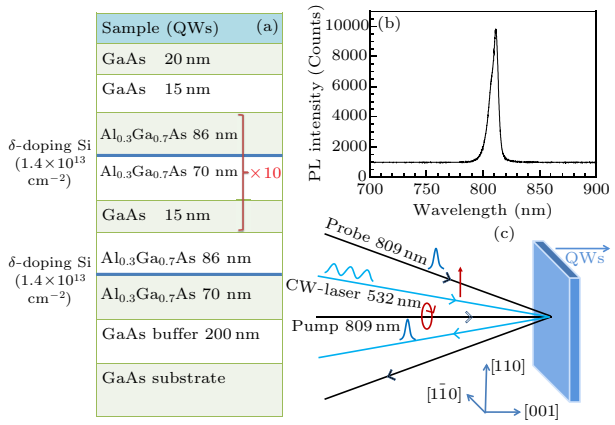

Fig. 1. (a) Schematic diagram of the GaAs/$Al_{0.3}Ga_{0.7}As$ quantum wells structure. (b) The PL response measured at 8 K for the studied GaAs/$Al_{0.3}Ga_{0.7}As$ quantum wells. (c) The layout of TR-MOKE setup with the control beam of a 532 nm cw laser.

The time-resolved magneto-Kerr rotation (TR-MOKE) measurements are performed with a Ti:sapphire laser system (Chameleon Ultra II, Coherent Inc.) which provides 150 fs pulse with repetition rate of 80 MHz. The pump beam is incident normally on the sample surface with the wavelength tuned to be 809 nm, being nearly resonant with the band gap of the studied GaAs QWs sample at low temperature, based on the photoluminescence response measured at 8 K as exhibited in Fig. 1(b). The probe beam is directed onto the sample at an angle around 30° with respect to the surface normal. The polarization of the pump beam is adjusted to be circularly polarized and the probe beam is linearly polarized. The sample is mounted within a Janis closed-cycle optical cryostat which is located between the two poles of an electromagnet. The electromagnet can provide an in-plane magnetic field of about ±1 T. Our experiment is carried out at 8 K with the in-plane magnetic field of 0.9 T applied along either [110] or [1$\bar{1}$0] direction by rotating the sample. Thus the electron spin, initially polarized along the growth direction of the quantum well upon optical pumping, can be forced to precess into the (110) or (1$\bar{1}$0) plane. The third laser beam, served as 'optical gating' by using a 532 nm cw laser, is applied onto the sample surface at nearly normal incidence, as schematically shown in Fig. 1(c). The Kerr rotation signal is collected by a sensitive optical bridge balanced Si photodetector combined with a lock-in amplifier.

Figure 2(a) shows the TR-MOKE signals measured at 8 K with pumping intensity of 45 mW (corresponding to an optically excited carrier concentration of $6.2 \times 10^{16}\,\mathrm{cm}^{-3}$) for the in-plane magnetic field applied along [110] and [1$\bar{1}$0], respectively. The TR-MOKE signals can be well fitted with an exponentially damped cosine function

$$S(t) = A\exp(-t/T_2^*)\cos(\omega t + \phi) + S_0, \qquad (2)$$

where A is the Kerr signal amplitude, t is the delay time between pump and probe beams, T_2^* is the electron spin relaxation time, ω is the electron Larmor precession frequency, ϕ is the initial phase, and S_0 is the Kerr signal background offset. The extracted electron Larmor precession frequencies for two directions are nearly identical. However, the extracted spin relaxation time as long as ~424 ps is observed with the field applied along [1$\bar{1}$0] direction, longer than that of [110] direction (~286 ps), which demonstrates a clear in-plane spin relaxation anisotropy in [001]-oriented GaAs QWs. The observed spin relaxation anisotropy was previously studied and attributed to the interference of the Rashba and the Dresselhaus spin–orbit couplings in the two-dimensional electron gas heterostructure.[10,15]

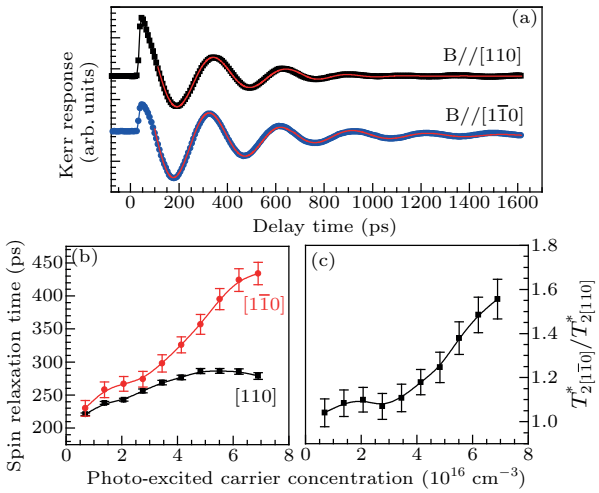

Fig. 2. (a) Time-resolved Kerr rotation signals measured at 8 K with field of 0.9 T applied along different crystallographic orientations. (b) Pumping intensity dependence of the in-plane electron spin relaxation times T_2^* measured at 8 K and the in-plane field of 0.9 T for different crystallographic orientations. (c) The optical pumping intensity dependence of the in-plane spin relaxation time anisotropy deduced from the result in (b).

Figure 2(b) shows the dependence of electron spin relaxation time T_2^* on the optically excited carrier density, which is measured at 8 K with field applied along different crystallographic orientations. Apparently, the spin relaxation time along both orientations is observed to increase with the increasing pumping intensity. For the GaAs-based QWs, the extensive studies in the past have revealed that the electron spin

relaxation is dominated by the DP mechanism,[2–5] in which the spin relaxation time is inversely proportional to the electron momentum relaxation time. Therefore the spin relaxation time T_2^* will be extended due to the enhanced momentum scattering when increasing electron density. Furthermore, the spin relaxation anisotropy ratio of $T_{2\,[1\bar{1}0]}^*/T_{2\,[110]}^*$ is observed to increase monotonically as presented in Fig. 2(c), which results from the relative strength modulation of the Rashba and the Dresselhaus fields by increasing the optically pumped electron density.[16–19] The Rashba field strength is known to be proportional to $\alpha_0 \frac{2\pi e^2 n}{\varepsilon}$,[15,18–20] where α_0 is the second spin–orbit constant determined by both bulk spin–orbit interaction parameters and properties of heterointerfaces, e and n are the elementary charge and carrier density respectively, and ε is the dielectric constant. The Dresselhaus field strength is known to be proportional to $\langle k_z \rangle^2 \propto n^{2/3}$.[20,21] Thus both the Rashba and the Dresselhaus fields are enhanced with the increasing optically pumped electron density.[7] The increasing spin relaxation anisotropy as observed in Fig. 2(c) is a consequence of the relative strength modulation for the Rashba and the Dresselhaus fields by the increased electron density. From the electron density dependent trend of spin relaxation anisotropy as observed in Fig. 2(c) and the above analyses, we can speculate that $|\alpha/\beta| < 1$ for the investigated GaAs QW structure in this work. For the DP spin relaxation mechanism, the anisotropic spin relaxation time can be expressed as[10,18]

$$\frac{1}{T_2^*} = \frac{3}{4} C\beta^2 \Big[1 + \Big(\frac{\alpha}{\beta}\Big)^2 + \frac{2\alpha}{3\beta}\sin(\varphi)\Big], \qquad (3)$$

where C is a constant, and φ is the angle of magnetic field orientation relative to the [100] axis. Therefore we can evaluate the absolute value of $|\alpha/\beta|$ for different pumping densities based on the measured value of $T_{2\,[1\bar{1}0]}^*/T_{2\,[110]}^*$. The value of $|\alpha/\beta|$ is thus estimated to increase from about ∼0.1 to ∼0.4 by increasing optically pumped electron density from $0.8 \times 10^{16}\,\mathrm{cm}^{-3}$ to $6.9 \times 10^{16}\,\mathrm{cm}^{-3}$.

To further tune the background carrier density through an optical method, a third optical beam (control beam) is introduced to adjust the electron density in GaAs QWs by using a cw laser with wavelength of 532 nm. Prior to TR-MOKE investigation, the Hall measurements are first performed at 150 K and above (at which temperatures the Ohmic contact is still good enough) by varying the excitation power of a cw laser, to check the capability of carrier density tuning by shining the cw laser. Our Hall result at 150 K shows that the carrier concentration exhibits threefold enhancement when increasing the cw laser power from 4 mW to 50 mW. Greater enhancement of carrier concentration by shining the cw laser at lower temperature is expected. Figure 3 shows the typical TR-MOKE result measured at 8 K with the magnetic field of 0.9 T applied along [110] axis and optically excited carrier density of $2.8 \times 10^{16}\,\mathrm{cm}^{-3}$ by varying the cw laser excitation power. It is seen apparently that the electron spin relaxation behavior exhibits strong dependence on the cw laser excitation. Figure 4(a) shows the dependence of the extracted electron spin relaxation time on the electron density further modulated by the cw control beam. It can be clearly seen that the spin relaxation time presents a non-monotonic dependent behavior as a function of the cw laser excitation power. The injected electron density under the cw laser excitation can be estimated to be $4.5 \times 10^{15}\,\mathrm{cm}^{-3}$ at excitation power of 1 mW. The observed maximum spin relaxation time appears at the cw laser excitation power of 10 mW (corresponding to the photo-excited electron density of $\sim 4.5 \times 10^{16}\,\mathrm{cm}^{-3}$). The non-monotonic dependence of electron spin relaxation time on the increased electron density injected by the cw laser agrees with the previous studies in GaAs[5,22,24,27] and the CdTe[23,28] two-dimensional system in which the electron spin relaxation is mainly governed by the DP mechanism.[5,22–27] Since the electron–electron scattering time τ_p exhibits the non-monotonic dependence on the electron density and temperature in the low electron density (non-degenerated) and high electron density (degenerated) regimes as $\tau_p \propto n^{-1}T^{2/3}$ and $\tau_p \propto n^{2/3}T^{-2}$,[5,24] respectively, the spin relaxation time T_2^*, which is inversely proportion to τ_p under the DP mechanism, is thus proportional to the electron density as $T_2^* \propto nT^{-2/3}$ (in the low electron density regime) and $T_2^* \propto n^{-2/3}T^2$ (in the high electron density regime), respectively. Therefore the spin relaxation time in the low electron density and high electron density regimes has different density dependent behavior. In the low electron density regime, the spin relaxation time increases with the increasing electron density under the additional excitation of the cw laser. Oppositely, in the high electron density regime, the spin relaxation time decreases with further excitation of the cw laser.

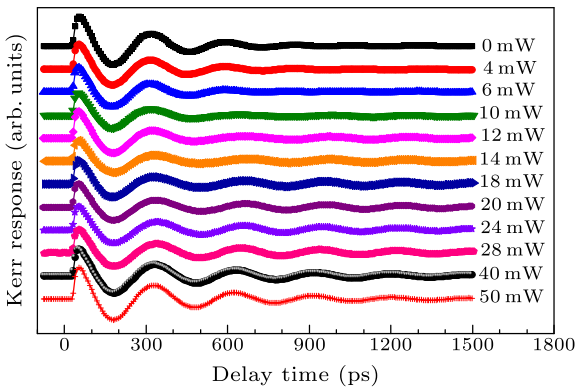

Fig. 3. TR-MOKE responses measured at 8 K for different excitation powers of the cw-laser with the magnetic field of 0.9 T applied along the [110] axis.

Figure 4(b) shows the obvious spin relaxation anisotropy modulation via the third cw laser beam excitation. It is observed that the anisotropy ratio of $T_{2\,[1\bar{1}0]}^*/T_{2\,[110]}^*$ increases from 1.8 to 2.4 when the

photo-excited electron density increases from $\sim 10^{16}$ to $\sim 10^{17}\,\mathrm{cm}^{-3}$ by the cw laser tuning. As it has been mentioned that the Rashba and the Dresselhaus field strengths can be modulated by tuning electron density, the strength of both fields can be enhanced by increasing electron concentration. Since the anisotropy of in-plane spin relaxation is the largest when $|\alpha/\beta| = 1$,[11] the enhanced anisotropy with increasing electron concentration from our measurement implies that the absolute value of the difference between the Rashba and the Dresselhaus spin–orbit coupling strengths decreases with increasing the electron concentration, as a result of different degrees of their response to the electron density tuning. Based on the measured electron spin relaxation anisotropy ratio presented in Fig. 4(b), the ratio of the Rashba and the Dresselhaus spin–orbit coupling parameters $|\alpha/\beta|$ is estimated to increase from ~ 0.5 closely to ~ 1 by further varying excitation density of the cw laser. Though the absolute value of $|\alpha/\beta|$ is hardly evaluated just based on the anisotropic spin relaxation time when $|\alpha|$ is nearly equal to $|\beta|$ by Eq. (3), since theoretically the in-plane spin relaxation time will become infinitely long at this special case, nevertheless, our experimental observations demonstrate that the relative strengths of the Rashba and the Dresselhaus spin–orbit coupling fields can be effectively tuned via an optical gating method, and consequently the spin relaxation anisotropy in a conventional quantum well structure is efficiently tuned. The optical gating realized in this work is simple and convenient, and does not require complex device processing as involved in the electric gating technique.

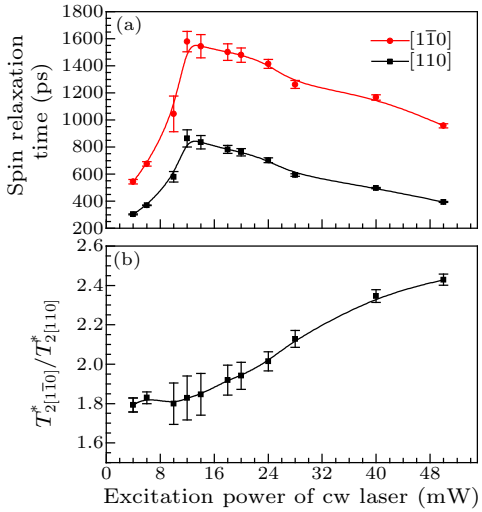

Fig. 4. (a) The in-plane electron spin relaxation time T_2^* measured at 8 K for the magnetic field of 0.9 T applied along different orientations of the sample. (b) The cw laser excitation power dependence of the spin relaxation time anisotropy deduced from the result in (a).

In conclusion, the in-plane spin relaxation anisotropy has been studied in the [001]-oriented modulation doped GaAs/$Al_{0.3}Ga_{0.7}$As QWs. The non-monotonic dependence of electron spin relaxation time on the increased electron density injected by the third cw laser confirms the dominant DP mechanism responsible for the spin relaxation in GaAs QWs. Moreover, the obvious spin relaxation time anisotropy can be efficiently tuned via an optical gating method by the additional excitation of a cw laser beam, reflecting the relative strength tuning of the Rashba and the Dresselhaus fields with the photoexcitation electron density. Our experimental observations demonstrate a simple and convenient method to efficiently tune the relative strength of the Rashba and the Dresselhaus spin–orbit coupling fields through a purely optical method without further modulation of the sample's structural symmetry.

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