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To cite this article: L. H. Teng *et al* 2008 *EPL* **84** 27006

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Density dependence of spin relaxation in GaAs quantum well at room temperature

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received 25 June 2008; accepted in final form 8 September 2008

published online 2 October 2008

PACS 72.25.Rb – Spin relaxation and scattering

PACS 78.67.De – Quantum wells

PACS 71.10.-w – Theories and models of many-electron systems

Abstract – Carrier density dependence of electron spin relaxation in an intrinsic GaAs quantum well is investigated at room temperature using time-resolved circularly polarized pump-probe spectroscopy. It is revealed that the spin relaxation time first increases with density in the relatively low-density regime where the linear D'yakonov-Perel' spin-orbit coupling terms are dominant, and then tends to decrease when the density is large and the cubic D'yakonov-Perel' spin-orbit coupling terms become important. These features are in good agreement with theoretical predictions on density dependence of spin relaxation by Lü *et al.* (*Phys. Rev. B*, **73** (2006) 125314). A fully microscopic calculation based on numerically solving the kinetic spin Bloch equations with both the D'yakonov-Perel' and the Bir-Aronov-Pikus mechanisms included, reproduces the density dependence of spin relaxation very well.

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Spintronics is an intriguing and growing field which aims to incorporate the spin degree of freedom into the traditional electronics [1,2]. In this field, the spin relaxation time is one of the most important basic quantities, especially for the design of spin-based devices. Typically, for example, ultrafast spin relaxation process is needed for spin-dependent optical switch [3–5], while long enough spin lifetime is required when dealing with quantum information storage as well as spin transport [6–8]. Investigations on spin relaxation in various materials and structures have been carried out both experimentally and theoretically, revealing that spin relaxation can be affected/manipulated by various factors, such as temperature [9–16], initial spin polarization [17], carrier and/or impurity density [6,9,11–14,16,18–21], magnetic field [10,11,15,17], drift electric field [22,23], and so on. Among these factors, the carrier density is one of the most basic quantities which can be easily controlled by gate voltage and/or optical excitation power. Thus investigation on density dependence of spin relaxation is necessary.

It is well known that for zinc-blende semiconductors such as GaAs, the D'yakonov-Perel' (DP) mechanism is the leading mechanism of spin relaxation [24]. A femtosecond time-resolved Faraday rotation measurement of *n*-type bulk GaAs has shown that the spin relaxation time decreases with carrier density [20], and the theoretical calculation [11] is consistent with this result. Note that the spin-orbit coupling terms (DP terms) depend on momentum cubically in bulk GaAs [11,25]. However, due to the confinement along the growth direction, the DP terms in quantum wells include both linear and cubic terms. For example, in (001) symmetric GaAs quantum wells with small well width (so that only the lowest subband is involved), the DP terms come from the Dresselhaus terms [25]:

$$\Omega(\mathbf{k}) = \gamma(k_x(k_y^2 - \langle k_z^2 \rangle), k_y(\langle k_z^2 \rangle - k_x^2), 0), \quad (1)$$

in which $\langle k_z^2 \rangle$ stands for the average of the operator $-(\partial/\partial z)^2$ over the electron state of the lowest subband and γ is the spin splitting parameter. The relative importance of the linear and cubic DP terms depends on the well width [22], temperature [22] and carrier density [12].

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The different momentum dependences of the DP terms lead to complicated temperature and/or density dependences of spin relaxation in GaAs quantum wells. It is predicted from a fully microscopic kinetic spin Bloch equation (KSBE) investigation [13] that in the strong scattering limit, the spin relaxation time increases with carrier density when the linear DP terms are dominant and decreases with density when the cubic terms are important [12]. The underlying physics is associated with the competition between the two effects in the DP mechanism—the inhomogeneous broadening [12,22] and the counter effect of scattering on the broadening [12,22]. Density and/or temperature can affect this competition with the relative importance of each competing effect depending on which part of the DP terms are dominant. In fact, a non-monotonic dependence of spin relaxation on temperature in GaAs quantum wells was theoretically predicted in ref. [22], and was verified experimentally recently [15,17]. However, the predicted density dependence of spin relaxation [12] has not yet been verified experimentally. This work is to investigate the density dependence of spin relaxation in (001) GaAs quantum wells at room temperature. Unlike some earlier studies on two dimensional GaAs with low carrier density and at low temperature where the excitonic effects dominate [26], the present investigation is in the regime of the electron-hole plasma.

The two-dimensional sample consists of 11 periods of 10 nm thick GaAs quantum wells separated by 6 nm $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$ barriers, grown on semi-insulating GaAs substrate by molecular beam epitaxy along the (001) direction (z -axis). The substrate is removed by polishing first and then selective chemical etching for transmission measurements. The substrate-free GaAs/AlGaAs films are mounted on a piece of sapphire window. The widely used time-resolved circularly polarized pump-probe spectroscopy [27–30] is adopted to realize spin pumping and spin relaxation measurements. The femtosecond laser pulses generated from a Ti:sapphire laser oscillator have a duration of 100 fs, a half spectrum width at half maximum of 6.7 meV, and a repetition rate of 82 MHz. By passing through a standard time-resolved pump-probe setup, the pulses are split into pump and probe ones with intensity ratio of 3:1. The pump and probe beams are incident nearly normally to the sample surface and focused by a lens of 50 mm focal length on the sample to a spot size of about 30 μm in diameter. Two commercially available achromatic quarter-wave plates are inserted into the pump and probe beams, respectively, to generate co-helicity or cross-helicity circularly polarized pump and probe pulses. The differential transmission change of the probe is detected by a photodiode and measured by a lock-in amplifier which is referenced at the modulation frequency of an optical chopper that modulates the pump beam. The central wavelength of the pulses is tuned to 830 nm to excite the heavy-hole transition alone, and thus an initial degree of spin polarization of nearly 100% may be obtained [31]. In this experiment, right circularly

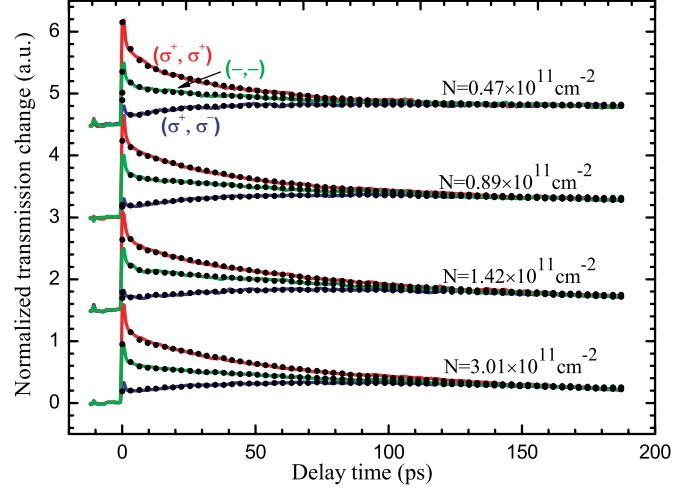


Fig. 1: (Color online) Normalized transmission change of probe beams under four typical carrier densities. The color solid curves are experimental data, while the black dots are the fittings with an elliptically polarized pump-probe absorption model. Curves labeled with (σ^+, σ^+) are taken from co-helicity pump-probe beams, with (σ^+, σ^-) are from the cross-helicity pump-probe beams and with $(-, -)$ are from collinearly polarized pump-probe beams. Different y -axis offsets are added to each set of curves for clarity.

polarized (σ^+) pump pulses create spin-down polarized electrons, while time-delayed σ^+ (left circularly polarized (σ^-)) probe pulses measure the number of spin-down (-up) electrons. Recombination of the photo-excited carriers can be detected by using linearly polarized light. In addition, a tunable optical attenuator is used to control input laser pulse energy so that the electron density N can change in a range of $0.3 \times 10^{11} \text{ cm}^{-2}$ to $4 \times 10^{11} \text{ cm}^{-2}$. The excitation density is calculated by an usual formula, $(1-R)E\alpha/(\hbar\nu S)$, with R and α being the reflectivity and absorption coefficient of the sample, respectively. Here E is the pumping energy per pulse, $\hbar\nu$ is the photon energy, and S is the area of pump spot.

The main results are shown in figs. 1 and 2. Figure 1 indicates the normalized time-delayed scanning transmission change profiles of the probe beams for four different excited carrier densities of $0.47 \times 10^{11} \text{ cm}^{-2}$, $0.89 \times 10^{11} \text{ cm}^{-2}$, $1.42 \times 10^{11} \text{ cm}^{-2}$ and $3.01 \times 10^{11} \text{ cm}^{-2}$, respectively. The profiles labeled as (σ^+, σ^+) and (σ^+, σ^-) are taken from co-helicity and cross-helicity circularly polarized pump and probe beams respectively, whereas the ones labeled by $(-, -)$ are the collinear polarization pump-probe traces which describe the recombination processes of the photoexcited carrier population. Initially, the (σ^+, σ^+) profile is stronger than the $(-, -)$ one, while the (σ^+, σ^-) profile is weaker than the $(-, -)$ one. But finally both (σ^+, σ^+) and (σ^+, σ^-) profiles tend to coincide with the $(-, -)$ profile. This just shows the relaxation of spin polarization between $|1/2\rangle$ and $|-1/2\rangle$ spin states of electrons in conduction band [27,32]. The (σ^+, σ^+) profile reflects the decay of majority spin population

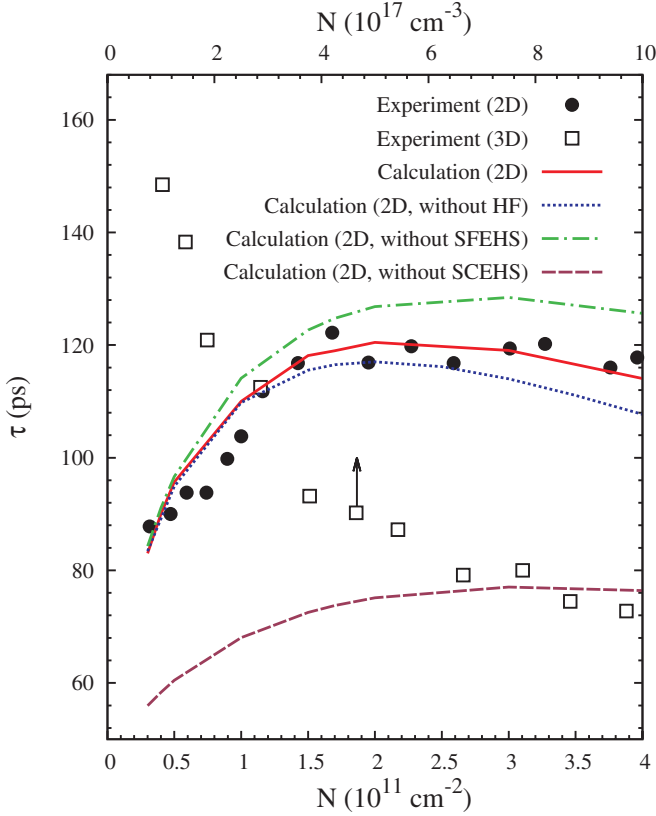


Fig. 2: (Color online) Carrier density N dependence of electron spin relaxation time τ . Dots: experimental data in quantum well (2D); open squares: experimental data in bulk material (3D). Solid curve: full theoretical calculation; chain curve: theoretical calculation without the SFEHS; dashed curve: theoretical calculation without the SCEHS; dotted curve: theoretical calculation without the Coulomb HF term. Note the scale of the bulk data is on the top frame of the figure.

directly photocreated by σ^+ pump pulses, while (σ^+, σ^-) profile rises initially toward the $(-, -)$ profile, which reflects population increase in minority spin state induced by spin flip from the majority spin state. An elliptically polarized pump-probe spectroscopic model described in ref. [32] is used to fit the time-delayed experimental profile to retrieve spin relaxation time τ . The hole spin relaxation is irrelevant because it is well known that hole spin relaxation is very fast (in sub-picosecond time scale) [30]. The results are shown in fig. 2, with dots corresponding to the results in the quantum well. In addition, a similar experiment is performed on bulk GaAs for comparison and the open squares in fig. 2 are the results. It is found that with the increase of carrier density, the spin relaxation time in bulk material decreases monotonically with carrier density, coinciding with the previous reports in n -type bulk GaAs [11,20]. However, in quantum wells the spin relaxation time first increases in the low density regime and then tends to decrease after reaching a maximum of about 120 ps at the density of $1.7 \times 10^{11} \text{ cm}^{-2}$.

In order to gain a deep insight into the experimental results of the quantum well, we performed a fully

microscopic KSBE calculation [13], which takes account of all relevant spin relaxation mechanisms (including both the DP and Bir-Aronov-Pikus [13,33] (BAP) mechanisms). The KSBEs constructed by the nonequilibrium Green function method read [13]

$$\dot{\rho}_{\mathbf{k}} = \dot{\rho}_{\mathbf{k}}|_{coh} + \dot{\rho}_{\mathbf{k}}|_{scat}, \quad (2)$$

in which $\rho_{\mathbf{k}}$ represent the density matrices of electrons with momentum $\mathbf{k}$. $\dot{\rho}_{\mathbf{k}}|_{coh}$ are the coherent terms describing the coherent spin precession due to the effective magnetic fields from the DP term and the Hartree-Fock (HF) Coulomb interaction and $\dot{\rho}_{\mathbf{k}}|_{scat}$ stand for the scattering terms. In our calculation, all the relevant scatterings, such as the electron-longitudinal-optical-phonon scattering, electron-electron Coulomb scattering and electron-hole Coulomb scattering, are explicitly included. The electron-hole Coulomb scattering is further composed of both the spin-flip electron-hole scattering (SFEHS) and the spin-conserving electron-hole scattering (SCEHS), with the former leading to the spin relaxation due to the BAP mechanism. Expressions of the coherent and scattering terms are given in detail in ref. [13].

By solving the KSBEs, we obtain the spin relaxation time as a function of photoexcited carrier density. In the calculation, the spin splitting parameter γ (as a fitting parameter) is chosen to be $21 \text{ meV} \cdot \text{nm}^3$ [2]. The initial electron spin polarization is set to be 100% following the experiment and the temperature is 300 K. The solid curve in fig. 2 is from the full calculation which reproduces the experimental results very well. The obtained results can be understood from the joint effect of the following two competing effects: i) With the increase of carrier density, the spin-conserving scattering is strengthened. This tends to suppress the inhomogeneous broadening from the momentum dependence of the effective magnetic field (the DP terms) by driving carriers to more homogeneous states in momentum space, and thus weakens the spin relaxation in the strong scattering limit [12,13,22]. ii) Both the inhomogeneous broadening and the SFEHS increase with the density. This leads to the increase of spin relaxation. As pointed out by one of the authors in refs. [22] and [12], when the linear k -dependence of the DP term is dominant, the temperature and/or density dependence of effect i) is stronger than that of effect ii), consequently the spin relaxation time increases with temperature and/or carrier density. However, when the cubic term becomes dominant, the increase of inhomogeneous broadening (effect ii) with the temperature and/or density overcomes effect i), consequently, the spin relaxation time decreases with temperature and/or carrier density. This is exactly what happens in fig. 2: When $N < 1.5 \times 10^{11} \text{ cm}^{-2}$, the linear DP term dominates and the spin relaxation time increases with carrier density. When the density goes higher, the spin relaxation time tends to decrease. However, this decrease is moderate as the contribution from the cubic DP term has not yet become dominant but

is comparable with the linear term at the present photoexcited density. This can be further seen from the chain curve where the SFEHS, *i.e.*, the BAP term, is turned off. The decrease becomes even milder. Moreover, by comparing the solid curve and the chain curve, one finds the spin relaxation from the BAP mechanism becomes stronger with the increase of photoexcited carrier (especially hole) density. However, the spin relaxation is still dominated by the DP mechanism, as addressed very recently by Zhou and Wu in ref. [13]. We further show that for intrinsic sample, due to the same electron and hole densities, the SCEHS makes marked contribution to the spin relaxation due to the DP mechanism. This can be seen from the dashed curve where the SCEHS is turned off. One obtains much shorter spin relaxation time as the counter effect of the scattering to the inhomogeneous broadening is markedly weakened by neglecting the SCEHS.

Finally we address the issue of the initial spin polarization. Due to the intrinsic two-dimensional sample, the initial spin polarization is 100% by the circular polarized laser excitation. Unlike the low temperature case where an effective magnetic field in Faraday configuration is induced by the Coulomb HF interaction and the spin relaxation is markedly reduced [17], here the effective magnetic field is very small due to the high temperature (so the electron distribution functions are much smaller than 1 for most momentums). Therefore, the effect of the HF term to the spin relaxation is marginal, as shown by the dotted curve in fig. 2 where the HF term is removed. It is further seen from the figure that with the increase of the density, the contribution of the HF term becomes noticeable and the spin relaxation is suppressed, in agreement with the previous theoretical predictions and experimental observations [17].

In summary, the carrier density dependence of electron spin relaxation in intrinsic (001) GaAs quantum wells is investigated by a femtosecond pump-probe experiment at room temperature. The spin relaxation time shows an obvious increase with density in the relatively low density regime and then a mild decrease when the density is larger, which is in good agreement with the theoretical predictions [12]. Further calculation with the fully microscopic KSBE approach reproduced the experimental results very well. It is understood that in the strong scattering limit, when the carrier density is low and thus the linear DP term is dominant, the spin relaxation time increases with density due to the increasing counter effect of the scattering on the inhomogeneous broadening. However, when the density is large enough and the cubic DP term becomes important, the spin relaxation time tends to decrease with density, thanks to the rapid increase of the inhomogeneous broadening and the enhanced effect from the BAP mechanism.

This work was supported by the Natural Science Foundation of China under Grant Nos. 10574120, 10725417,

60490295, and 60678009, the National Basic Research Program of China under Grant No. 2006CB922005 and the Knowledge Innovation Project of Chinese Academy of Sciences.

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