Study of Electron Spin Diffusion and Relaxation Dynamics by Diffraction of transient spin grating in an Intrinsic GaAs/AlGaAs Quantum Well

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Abstract. In this paper, the transient spin grating method was used to measure the attenuation rate of the diffraction signals of the intrinsic spin gratings of the intrinsic GaAs/AlGaAs quantum wells in different periods at room temperature, and the electron spin diffusion coefficient $D_s = 121 \pm 6 \text{cm}^2/\text{s}$ was obtained. The electron spin diffusion coefficients of GaAs/AlGaAs quantum well were in good agreement with that of p-type GaAs/AlGaAs quantum well, indicating that the doping type for GaAs/AlGaAs quantum well has no significant effect on the diffusion rate of spin-polarized electrons. In this paper, the widely used formula of transient spin grating diffraction signal attenuation rate was used to fit the diffraction experimental results of the transient spin grating in the intrinsic GaAs/AlGaAs quantum well. The measured electron spin relaxation time was much shorter than that measured by the saturation absorption method. The reason for the deviation in the measured spin relaxation time was analyzed. The dynamic law of transient spin grating modulation attenuation over time was derived. The decay rate formula was modified, and the modified formula was used to fit the experimental data in the transient spin grating diffraction to obtain the spin relaxation time $\tau = 123 \text{ps}$. The result was consistent with the measured electron spin relaxation time by the saturation absorption method.

1. Introduction

The transient spin grating diffraction method is a widely used experimental method [1-4]. The direction of the diffracted light generated by the probe light after the transient spin grating is different from that of the pump light and probe light, the effect of the pump light and probe light on the experimental measurement results is effectively reduced, thereby reducing the background signal. In recent years, transient spin grating technology has been widely used in the field of electron spin diffusion and electron spin relaxation dynamics. S. G. Carter et al. [5] studied the relationship between electron spin diffusion transport and relaxation processes with temperature, excited electron concentration, and excited photon energy in a n-GaAs/AlGaAs quantum well using a transient spin grating. T. Ishiguro et al. [6] applied the transient spin grating technology to measure the electron spin relaxation time of GaN, and studied the relationship between electron spin relaxation time and...
temperature in GaN at low temperature. A. R. Cameron et al. [7] used this experimental technique to study the electron spin diffusion transport process of p-GaAs/AlGaAs quantum wells at room temperature. The electron spin diffusion coefficient \( D_s = 127 \text{ cm}^2/\text{s} \) was measured and calculated, and the electron mobility \( \mu \approx 5000 \text{ cm}^2/\text{Vs} \) was obtained based on the Einstein relationship \( eD = kT\mu \).

The C. P. Weber group [8] excited the transient spin grating in the n-GaAs/AlGaAs quantum well and measured the electron spin diffusion coefficient \( D_s = 130 \text{ cm}^2/\text{s} \) at room temperature. The experimental results from Cameron's group and Weber's group showed that: n-GaAs/AlGaAs and p-GaAs/AlGaAs quantum wells have approximately the same electron spin diffusion coefficient, suggesting that the doping type of GaAs/AlGaAs quantum wells may not have a great influence on the diffusion rate of the electrons. Then, the measurement results of the electron spin diffusion coefficient of the intrinsic GaAs/AlGaAs quantum wells will be helpful to further verify the influence of the quantum well doping type on the electron spin diffusion speed.

When two beams of orthogonal linearly polarized light at a certain angle are superimposed on the sample surface, the circular polarization of the superimposed light changes periodically, resulting in the periodic change of the electron spin polarization in the sample, which is the so-called spin grating. If a linearly polarized probe light pulse with a time delay relative to the pump light pulse passes through the spin grating, the probe beam will be diffracted by the spin grating. Due to spin relaxation, spin diffusion, and electron-hole recombination, the spin polarization modulation amplitude of the transient spin grating decreases with time, and the intensity of the diffraction signal of the probe beam also decreases. Therefore, by experimentally measuring the attenuation rate of the diffraction signal of the probe beam, the information related to the electron spin diffusion and spin relaxation can be obtained. Scholars have studied the electron density grating generated by light excitation, and derived the formula for the attenuation of the modulation density of the transient density grating over time:

\[
\Gamma = \left(4\pi^2/\Lambda^3\right) D_s + 1/\tau_e \] [9],

where \( \Gamma \) is the attenuation rate of the modulation density of the transient density grating and \( \Lambda \) is the transient density grating Period, \( D_s \) is the diffusion coefficient of electrons, and \( \tau_e \) is the electron-hole recombination time. By analogy, people obtain the formula \( \Gamma_s = \left(4\pi^2/\Lambda^3\right) D_s + 1/\tau_s \) for the spin polarization decay of a transient spin grating over time, where \( \Gamma_s \) is the attenuation rate of the modulation of the transient spin grating, \( D_s \) is the electron spin diffusion coefficient, and \( \tau_s \) is the electron spin relaxation time. By this formula, after measuring the attenuation rate of a series of spin gratings with different periods, the electron spin diffusion coefficient and electron spin relaxation time can be calculated [10].

The directly and commonly used method for measuring the electron spin relaxation time is the circularly polarized light saturation absorption method. In 1990, A. Tackeuchi [11] directly observed the spin relaxation process of electrons by using a circularly polarized light saturation absorption method for the first time. They measured the electron spin relaxation time of 32ps in an AlGaAs/GaAs quantum wells at room temperature. Subsequently, the circularly polarized light saturation absorption method has been widely used in the study of electron spin relaxation kinetics [12-14]. The pump-probe method uses a circularly polarized femtosecond laser to excite the sample to cause electron spin polarization, and then uses another femtosecond laser with a certain time delay relative to the excitation light to detect the electron spin polarization. The decay dynamic curve can be measured to obtain the time constant for electron spin relaxation. It can be seen that both the transient spin grating method and the circularly polarized light saturation absorption method can measure the electron spin relaxation time, so the reliability and consistency of the measurement results of the electron spin relaxation time need to be experimentally verified.

In this paper, the transient spin grating method was used to study the electron spin diffusion process of intrinsic GaAs/AlGaAs quantum well samples. The degradation rates of the transient spin
gratings at different periods were measured, the electron spin diffusion coefficient was calculated, and the effect of the doping type on the diffusion rate of spin-polarized electrons was studied. The electron spin relaxation time was measured using the transient spin grating method and the circularly polarized light saturation absorption method. The spin grating attenuation formula was modified.

2. Experimental methods and results

The experiments were performed at room temperature. The sample used in the experiment is a GaAs/AlGaAs multiple quantum wells, which consists of 11 cycles of intrinsic GaAs (6 nm)/AlGaAs (10 nm). The substrate is removed by selective etching, and the sample is adhered on the sapphire substrate. The absorption energy level of heavy hole exciton is about 1.5 eV at room temperature. Adjust the center of wavelength of the Ti: sapphire mode-locked laser to 830 nm. The pulse width is about 150 fs and the repetition rate is 96 MHz. In a spin grating experiment, a femtosecond laser pulse from a titanium sapphire laser is split into two pumping pulses and a detection pulse by a beam splitter. The two pumping light pulses have the same intensity of 10 mW, reaching the sample at the same time and overlapping on the sample to form a spot of 100 μm in diameter. The probe light pulse reaches the sample and forms a spot on the sample whose center coincides with the center of the pump spot and has a size of 60 μm. The delay time of probe light pulse relative to the pump light pulse is controlled by a delay line. By using the wave plate on the pump and detection optical paths, pump light and detection light of various polarization states can be conveniently obtained. The chopper is set on the detection optical path. Because the intensity of the detection light is much smaller than the intensity of the pump light, which can effectively reduce the influence of the incident light scattering signal on the measurement signal. The half-wave plate on one pump optical path makes the polarization directions of the two-line polarization pump light orthogonal to each other. The two beams of pump light overlap on the sample and excite to produce a spin grating. In order to improve the signal-to-noise ratio, an analyzer with a transmission direction orthogonal to the polarization direction of the incident probe light is placed on the diffracted light path to filter out the scattered signal of the probe light.

The decay curve of the diffraction signal intensity of a transient spin grating with a period of \( \Lambda = 6.9 \mu m \) over time is shown in Figure 1. The time constant \( \tau = 16.9 \) ps can be obtained by single exponential fitting the decay curve. Figure 2 is the attenuation curve of the diffraction signal of the transient spin grating as a function of \( 2 \pi \frac{\Gamma}{\Lambda} \). Using \( 2 \Gamma = \left(8\pi^2/\Lambda^2 \right)D_s + 2/\tau_s \) as the fitting formula, the curve is linearly fitted, and the electron spin diffusion coefficient \( D_s = 121 \pm 6 \) cm²/s can be determined from the slope of the fitted straight line. This result is consistent with the measurement result \( D_s = 127 \) cm²/s of the p-GaAs/AlGaAs quantum well by the A. R. Cameron group [7] and the measurement result \( D_s = 130 \) cm²/s of the n-GaAs/AlGaAs quantum well by the C. P. Weber group [8], indicating the doping type of the quantum well has no significant effect on the diffusion rate of spin-polarized electrons. In addition, by the intercept of the fitted straight line on the vertical axis and \( 2 \Gamma = \left(8\pi^2/\Lambda^2 \right)D_s + 2/\tau_s \), the electron spin relaxation time \( \tau_s = 49 \) ps is calculated.
Fig. 1. The intensity curve of the diffraction signal of the transient spin grating with period $\Lambda = 5.8 \mu m$ over time.

Fig. 2. The relationship between the attenuation rate $2\Gamma$ of the diffraction signal of the transient spin grating and $8\pi^2D_s/\Lambda^2$. 
The kinetic curves of electron-hole recombination and electron spin depolarization measured by the saturation absorption method are shown in Fig. 3. In the saturation absorption experiment, we use excitation light with the same wavelength and intensity as the above-mentioned spin grating experiment. A single exponential fitting is performed on the $n_s+n_\tau$ and $n_s-n_\tau$ curves shown in Figure 3, by which we obtain the time constant of $n_s+n_\tau$ curve $\tau = 258\,\text{ps}$ and the time constant of $n_s-n_\tau$ curve $\tau = 47\,\text{ps}$, respectively. Then, using $1/\tau = 2/\tau_1 + 1/\tau_2$, the spin relaxation time $\tau = 115\,\text{ps}$ can be determined, which is significantly greater than $49\,\text{ps}$ which was measured by the transient spin grating method.

**Fig. 3.** Kinetic curves of electron-hole recombination and electron spin relaxation measured by the saturation absorption method.

### 3. Discussion

Obviously, different experimental measurement methods for the same physical quantity should obtain consistent measurement results. However, the spin relaxation time constants measured by the saturation absorption method and the transient spin grating method vary from each other. For this reason, it is necessary to examine the rationality of the formulas used to fit the pump-probe signal and the attenuation rate of the transient spin grating diffraction signal. For the excited region of the sample in the saturation absorption experiment, the kinetic equation for the spin-polarized electron can be expressed as [15,16]:

$$\frac{\partial n_s}{\partial t} = \frac{n_s-n_\tau}{\tau_s} + \frac{n_\tau-n_s}{\tau_\tau}$$  \hspace{1cm} (1)

$$\frac{\partial n_\tau}{\partial t} = \frac{n_\tau-n_s}{\tau_s} + \frac{n_s-n_\tau}{\tau_\tau}$$  \hspace{1cm} (2)
Combine (1) and (2):
\[
\frac{\partial}{\partial t}[n_e(x,t) - n_h(x,t)] = \frac{n_e(x,t) - n_h(x,t)}{\tau}
\]
(3)

Where \(1/\tau = 2/\tau_s + 1/\tau_r\), \(\tau_s\), and \(\tau_r\) are the relaxation time constant of the electron spin and the electron-hole recombination time constant, respectively. The solution to equation (3) is:
\[
n_e - n_h = (n_e - n_h)_{0} \exp\left(-\frac{t}{\tau}\right)
\]
(4)

(4) is the fitting formula that we used to fit the pump-probe experiment curve in the experiment.

For transient spin gratings, in addition to electron-hole recombination and electron spin relaxation, electron spin diffusion is also responsible for the decrease of the polarization electron concentration. In the electron spin diffusion region, the net spin-polarized electron concentration satisfies the continuity equation [10]:
\[
\frac{\partial}{\partial t}[n_e(x,t) - n_h(x,t)] = \frac{D_s}{\tau_s} \nabla^2 [n_e(x,t) - n_h(x,t)] - \frac{n_e(x,t) - n_h(x,t)}{\tau}
\]
(5)

The physical meaning of \(\tau\) in (5) is exactly the same as that in (3), and \(D_s\) is the diffusion coefficient of the electron spin. Compared with the formula (3), the first term of the formula (5) represents the change rate of the spin polarization. After solving this equation, we get the equation as follows:
\[
\delta(t) = \exp\left[-\left(\frac{4\pi^2 D_s}{\Lambda^2} + \frac{1}{\tau}\right)t\right]
\]
(6)

Where \(\delta(t) = (m_{\text{max}}(t) - m_{\text{min}}(t))/2\) and \(m(x,t) = n_e(x,t) - n_h(x,t)\) are the modulation amplitude of the spin grating, \(\Lambda\) is the grating constant of the transient spin grating, \(\Lambda = \lambda/[2\sin(\theta/2)]\), \(\theta\) is the angle between the two pumping lights, and \(\lambda\) is the wavelength of the pumping light. According to formula (6), the degradation rate of the modulation degree of the transient spin grating can be expressed as:
\[
\Gamma_s = \frac{4\pi^2 D_s}{\Lambda^2} + \frac{1}{\tau_s} = \frac{4\pi^2 D_s}{\Lambda^2} + \frac{1}{\tau_s} + \frac{2}{\tau_r}
\]
(7)

Equation (7) is the formula for fitting of the spin grating experimental curve, which is used to correct the widely used \(\Gamma_s = (4\pi^2/\Lambda^2)D_s + 1/\tau_s\). Because the intensity of the diffraction signal of the transient spin grating is proportional to the square of the modulation of the spin grating, the attenuation rate of the diffraction signal of the transient spin grating is as follows:
\[
2\Gamma_s = \frac{8\pi^2 D_s}{\Lambda^2} + \frac{2}{\tau_s} = \frac{8\pi^2 D_s}{\Lambda^2} + \frac{2}{\tau_s} + \frac{4}{\tau_r}
\]
(8)

The last term on the right terms of the formula (7) differs by one time from that of \(\Gamma_s = q^2 D_s + 1/\tau_s\). In addition, the formula (7) also takes into account the contribution of electron-hole recombination to the attenuation of the transient spin grating. Using equation (8) to re-fit the spin grating diffraction signal, combined with the electron-hole recombination time \(\tau_r = 258\,\text{ps}\) measured by the saturation absorption method, the electron spin relaxation time \(\tau_s = 123\,\text{ps}\) can be calculated. The result is basically consistent with the spin relaxation time \(\tau_s = 115\,\text{ps}\) measured by the saturation absorption method, which proves that the above-mentioned modification of the transient spin grating degradation rate formula is reasonable.

4. Conclusions

The transient spin grating method was used to study the dynamics of electron spin diffusion and relaxation in the intrinsic GaAs/AlGaAs quantum well. The electron spin diffusion coefficient was
measured as $D_s = 121 \pm 6 \text{cm}^2/\text{s}$, which was in good agreement with experimental measured electron spin diffusion coefficients of n-type GaAs/AlGaAs quantum wells and p-type GaAs/AlGaAs quantum wells reported in the literature, indicating that the doping type of the quantum well has no significant effect on the diffusion rate of spin-polarized electrons. The attenuation rate formula of the diffraction signal of the transient spin grating was derived. The original degradation rate formula of the spin grating was modified, and the electron spin relaxation time was obtained by using the modified degradation rate formula of the transient spin grating.

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Reference

[1] K. Chen, W. F. Wang, J. D. Wu, D. Schuh, W. Wegscheider, T. Korn and T. S. Lai, Transmission-grating-photomasked transient spin grating and its application to measurement of electron-spin ambipolar diffusion in (110) GaAs quantum wells, Opt. Expr. (2012) 20(7) 8192.
[2] K. Chen, W. F. Wang, J. M. Chen, J. H. Wen and T. S. Lai, A transmission-grating-modulated pump-probe absorption spectroscopy and demonstration of diffusion dynamics of photoexcited carriers in bulk intrinsic GaAs film, Opt. Expr. (2012) 20 3580.
[3] H. L. Yu, S. Y Fang, J. H. Wen and T. S. Lai, Measurement of electron-spin transports in GaAs quantum wells using a transmission-grating-sampled circular dichroism absorption spectroscopy, J. Appl. Phys. (2014) 116 173765.
[4] K. Jarasiunas, V. Gudelis, R. Aleksiejunas, M. Sudziu, Picosecond dynamics of spin-related optical nonlinearities in In$_x$Ga$_{1-x}$As multiple quantum wells at 1064 nm, Appl. Phys. Lett. (2004) 84 1043.
[5] S. G. Carter, Z. Chen and S. T. Cundiff, Optical Measurement and Control of Spin Diffusion in n-Doped GaAs Quantum Wells, Phys. Rev. Lett. (2006) 97 136602.
[6] T. Ishiguro, Y. Toda and S. Adachi, Exciton spin relaxation in GaN observed by spin grating experiment, Appl. Phys. Lett. (2007) 90 011904.
[7] A. R. Cameron, P. Riblet and A. Miller, Spin Gratings and the Measurement of Electron Drift Mobility in Multiple Quantum Well Semiconductors, Phys.Rev.Lett. (1996) 76 4793.
[8] C. P. Weber, N. Gedik, J. E. Moore, Orenstein J, Stephens J and Awschalom D D. Observation of spin Coulomb drag in a two-dimensional electron gas, Nature (2005) 437 1330.
[9] Prem B. Bisht, Relaxation processes using transient grating technique, Res. Chem. Intermed. (2001) 27(4,5) 539.
[10] A. Pugzlys, P. J. Rizo, K. Ivanin, A. Slachter, D. Reuter, A. D. Wieck, C. Hvan. der Wal and P. H. M. van Loosdrecht, Charge and spin dynamics in a two-dimensional electron gas, J. Phys.: Condens. Matter (2007) 19 295206.
[11] A. Tackeuchi, S. Muto, T. Inata and T. Fujii, Direct observation of picosecond spin relaxation of excitons in GaAs/AlGaAs quantum wells using spin-dependent optical nonlinearity, Appl. Phys. Lett. (1990) 56 2213.
[12] R. S. Britton, T. Grevatt, A. Malinowski and R. T. Harley, Room temperature spin relaxation in GaAs/AlGaAs multiple quantum wells, Appl. Phys. Lett. (1998) 73 2140.
[13] A. Tackeuchi, O. Wada, and Y. Nishikawa, Electron spin relaxation in GaAlAs/InP multiple-quantum well, Appl. Phys. Lett. (1997) 70 1131.
[14] D. J. Hilton and C. L. Tang, Optical Orientation and Femtosecond relaxation of spin-polarized holes in GaAs, Phys. Rev. Lett. (2002) 89 146601-1.
[15] Lai T S, Liu L N, and Shou Q, et al. Elliptically polarized pump-probe spectroscopy and its application to observation of electron-spin relaxation in GaAs quantum wells, Appl. Phys. Lett. (2004) 85 4040.
[16] Lai T S, Liu X D, and Xu H H, et al. Elliptically polarized absorption spectroscopy and observation of spin coherence in intrinsic GaAs, Appl. Phys. Lett. (2006) 87 262110-1.