Room-temperature electron spin dynamics in GaAs/AlGaAs quantum wells

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(Received 1 August 1995; accepted for publication 30 November 1995)

We have investigated the spin relaxation process of electron at room temperature in nominally undoped GaAs/AlGaAs multiple-quantum wells. The spin relaxation times are measured for different well thicknesses using time-resolved polarization absorption measurement. The spin relaxation time τ_s , is found to depend on the electron confined energy E_{1e} , according to τ_s $\propto E_{1e}^{-2.2}$, showing that the main spin relaxation mechanism at room temperature is the D'yakonov– Perel' interaction. © 1996 American Institute of Physics. [S0003-6951(96)01706-4]

Spin relaxation is of interest from the viewpoints of fundamental physics as well as possible applications of spin dependent optical nonlinearity.¹ Spin relaxation of carriers in III-V semiconductors has been studied by measuring the steady state luminescence^{2,3} and the time-resolved luminescence⁴⁻⁸ using polarization. However, in both measurements, it is difficult to obtain the spin relaxation time at 300 K, the most practical temperature, due to the low luminescence efficiency. Also, the resolution of the most timeresolved luminescence has been on the order of 10 ps. One strong candidate for the spin relaxation mechanism near room temperature is the D'yakonov-Perel' (DP) interaction.⁹ Clark et al. reported, based on the steady state measurement, that an electron spin relaxation time is dominated by the DP mechanism from 50 to 200 K for GaAs crystal.² Miller et al. described that the DP process is relevant to spin relaxation in a GaAs/Al_{0.28}Ga_{0.72}Al MQW between 70 and 150 K.³ However, they did not report on the relaxation at room temperature probably due to the low luminescence intensity. The understanding of the spin relaxation kinetics at room temperature has remained vague.

Our present concern is the mechanism for the spin relaxation of electrons at room temperature and the controllability of the spin relaxation time. Previously, we showed that the time resolved absorption measurement using spin-dependent optical nonlinearity is a powerful tool to observe spin relaxation process directly in multiple quantum wells (MQW) with a high time resolution.¹⁰ Also, we demonstrated that the spin relaxation process is applicable to GaAs quantum well etalon to realize all-optical picoseconds gate operation.¹ The similar time resolved measurements have unveiled the spin relaxation process of excitons in a GaAs heterostructure,¹¹ type II superlattices,^{12,13} and CdMnTe MQW¹⁴ with a high time resolution of 1 ps or less. In this letter, we clarify the electron spin relaxation mechanism at room temperature in GaAs QW through our investigation of the dependence of the spin relaxation time on the electron confined energy. The quality of samples is important since spin dynamics can be strongly influenced by impurities or defects. Using MQW wafers with variation of the well thickness even on the same wafer, we extract the dependence of the spin relaxation time on the electron confined energy, independent of the impurity density. Our result shows an indication of DP mechanism The DP mechanism regards the spin-flip processes as a result of the asymmetry of the constituent atoms in the III–V compound with the zinc-blende structure. The anisotropic cubic in *k* term of the conduction band dispersion relation couples conduction-electron states of opposite spins. As the electron momentum is changed by any scattering event, the magnetic field fluctuates, resulting in slowing the spin relaxation. D'yakonov and Kachorovskii^{15,16} have shown that for quantum wells, the spin relaxation time, τ_s , of nondegenerate carriers in the conduction band is given by

$$\frac{1}{\tau_s} = \frac{2(\alpha E_{1e}/h)^2}{E_g k_B T} \int_0^\infty \epsilon \tau_v(\epsilon) \exp(-\epsilon/k_B T) d\epsilon, \qquad (1)$$

where α is a numerical coefficient governing the spin splitting of the conduction band. E_{1e} is the first electron confined state in the QW, and E_g is the band gap. τ_v is the electron momentum relaxation time and is proportional to the concentration of scattering centers. Equation (1) shows that the spin flip is more efficient for narrower wells as long as E_{1e} increases with decreasing well thickness.

In nominally undoped materials, since the concentration of scattering centers randomly fluctuates from sample to sample, a plot of τ_s vs E_{1e} in a series of samples might be deceptive. We used samples whose well thickness changed even on the same wafer. The intensity of the molecular beam during sample growth differs slightly for different positions on the GaAs wafer in our molecular beam epitaxy system. The well thickness reduces as it goes from the wafer center to the wafer edge. The concentration fluctuation of the scattering centers is expected to be much smaller in the same wafer, when compared to samples obtained from different wafers. The sample we investigated is designed consisting of 120 periods of alternating 4.52-nm-thick GaAs quantum wells and 4.0-nm-thick Al_{0.51}Ga_{0.49}As barriers at the wafer center (sample A). Another sample consists of 80 periods of alternating 5.65-nm-thick GaAs quantum wells and 4.0-nmthick $Al_{0.51}Ga_{0.49}As$ barriers at the wafer center (sample B). These structures were grown on a semi-insulating (100) GaAs substrate by molecular beam epitaxy. Figure 1 shows the absorption spectrum of sample A at various points on the sample wafer. The well thickness strongly depends on its position on the wafer. The electron-heavy hole absorption

being the leading spin relaxation mechanism at room temperature.

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FIG. 1. The absorption spectrum of sample A at various points on the sample wafer. The inset shows the measurement positions on the 2 in. wafer.

peak sweeps from 792 nm (center) to 771 nm (edge) which corresponds to the difference of 30 meV in electron confined energy.

We observed the spin relaxation process using spindependent optical nonlinearity. Spin-aligned carriers are created when electrons are excited by circularly polarized light. After right circularly polarized photoexcitation, σ_{+}^{pump} , the populations of carriers $N_{-}(N_{+})$, with down (up) spin along the direction of light propagation, are probed by right (left) circularly polarized probe pulse, $\sigma_{+}^{\text{probe}}$ ($\sigma_{-}^{\text{probe}}$). The detail of the pump-probe absorption measurement is described in our previous publication.¹⁰

The observed time dependence of transmission at the heavy hole exciton peak is shown in Fig. 2. We observed a clear exponential decay for the same circular polarization $(\sigma_{+}^{\text{pump}}, \sigma_{+}^{\text{probe}})$, and a clear exponential rise for anticircular polarization $(\sigma_{+}^{\text{pump}}, \sigma_{-}^{\text{probe}})$.

To better understand the spin relaxation kinetics, we subtracted the results of the circular polarization from the result of the linear polarization, as shown in Fig. 3. The curve consists of two time constants. One is a clear exponential decay of 16 ps which seems to be the electron spin relaxation. Another one is observed as an initial sharp spike which is comparable to the present time resolution of 1 ps. The clear initial peak in Fig. 3(a) can be mainly attributed to the coherent artifact that is generated due to pump beam diffraction by the optical transient grating in the sample. However, the observed sharp peak component in Fig. 3(b) means that the fast decay component with a time constant faster than 1 ps exists, because the coherent artifact becomes small for anticircular polarization.¹⁷

In the valence band, spin numbers are not good quantum numbers due to the band mixing of the heavy hole band and the light hole band.¹⁸ At room temperature, since the holes distribute over higher energy regions due to the thermal effect, the hole spin relaxation is largely affected by the bandmixing effect. Kawazoe *et al.* observed for type II MQWs that hole spin relaxation time reduces to subpicosecond at room temperature from 20 to 100 ps at liquid He temperature.^{12,13} The observation suggests that the our initial fast decay is also due to the hole spin relaxation. Therefore,



FIG. 2. The observed time dependence of transmission at the heavy hole exciton peak at the center of the sample A wafer. $(\sigma_{+}^{pump}, \sigma_{+}^{probe})$ indicates a right circularly polarized excitation and a right circularly polarized probe. Linear perpendicular shows the linear polarized excitation and linear polarized probe perpendicular to the excitation polarization.

we can attribute the slower 16 ps decay to the electron spin relaxation of 32 ps.

In the conduction band, the DP mechanism is believed to determine the electron spin relaxation at high temperatures.^{2,3} Although the DP mechanism usually works for free electrons in the conduction band, this mechanism of free electrons would work to relax the electron spin in excitons through the fast thermalization [0.2-0.3 ps (Ref. 19)] of electrons in free or excitonic states at room temperature. Equation (1) can be simplified by neglecting the energy dependence of τ_v ; so that τ_s is proportional to E_{1e}^{-2} . Therefore, the E_{1e} dependence of τ_s would be good evidence to determine the simplified by neglecting the simplified by neglecting the simplified by neglecting the energy dependence of τ_s would be good evidence to determine the simplified by neglecting the simplified by neglecting the simplified by neglecting the energy dependence of τ_s would be good evidence to determine the simplified by neglecting the simplified by neglecting the simplified by neglecting the energy dependence of τ_s would be good evidence to determine the simplified by neglecting the sim



FIG. 3. (a) The data obtained by subtraction of linear perpendicular from $(\sigma_+^{\text{pump}}, \sigma_+^{\text{probe}})$. (b) The data obtained by subtraction of $(\sigma_+^{\text{pump}}, \sigma_-^{\text{probe}})$ from the linear perpendicular.

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FIG. 4. The dependence of the spin relaxation time on the electron energy for sample (A) (open circles) and sample B (solid circles). The curves are fitted with the least-square method.

mine the spin relaxation mechanism. The electron energies are evaluated from the e_1 - hh_1 exciton absorption wavelength, where the excitonic binding energy of 8.6 and 8.4 meV for samples A and B, respectively, are calculated from variational approach.²⁰

Figure 4 shows the dependence of the relaxation time on the electron energy. The least-square method is used for curve fitting assuming

$$\tau_s = \beta E_{1e}^{-\gamma},\tag{2}$$

where, β and γ are fitting parameters. The obtained results show

$$\tau_s = 1.4 \times 10^{-6} E_{1e}^{-2.2} \tag{3}$$

for sample A and

$$\tau_s = 1.0 \times 10^{-6} E_{1e}^{-2.2} \tag{4}$$

for sample B.

The difference in the parameter β , means that the τ_v fluctuates from sample to sample showing the difficulty to extract systematic data from the some samples grown by different runs. However, note that the exponents are the same and very close to -2, which is expected from the DP mechanism. The accordance of the exponents for both samples means that the τ_v fluctuation in a wafer is small and the common spin relaxation mechanism governs both samples. The result indicates that the DP interaction is the leading electron spin relaxation mechanism in GaAs QW at room temperature.

The present observation is comparable with the result obtained by Rousiignol *et al.* at lower temperatures. They measured the τ_s dependence on E_{1e} at 4.2 K using time resolved PL measurement and found that τ_s is proportional to $E_{1e}^{-0.6}$ for GaAs/AlGaAs QW. Their result agrees with the other reports on the point that the DP interaction is not a main spin relaxation mechanism at low temperatures.^{2,3,5,8,21} The steady state measurements show that the temperature dependence of the spin relaxation time deviates from that of the DP mechanism at lower temperatures than 50 K for GaAs crystal and for GaAs/AlGaAs MQW.^{2,3} Also, a recent time resolved PL study indicates that exchange interaction governs the spin relaxation mechanism at lower temperatures than 50 K in GaAs QW.^{5,8,21} Therefore, the clear difference in the exponents between the result of Rousignol *et al.* and ours shows that the dependence of spin relaxation time on the electron confined energy is a good criterion to determine the spin relaxation mechanism.

From the point of application, controlling spin relaxation time is very important for designing an optimum device such as an etalon.^{1,22} The present result shows that the reasonable control of spin relaxation time can be obtained by changing the well thickness according to D'yakonov–Perel' interaction. The further reduction of the spin relaxation time is possible by reducing the concentration of the scattering centers.

In summary, we have investigated the spin relaxation process for electrons at room temperature in nominally undoped GaAs/AlGaAs multiple-quantum wells. The spin relaxation times are measured for the different well thickness using time-resolved polarization absorption measurements with high time resolution of 1 ps. The dependence of the spin relaxation time on the electron confined energy is found to be proportional to $E_{1e}^{-2.2}$. The result indicates that the D'yakonov–Perel' interaction is the leading spin relaxation mechanism at room temperature.

We thank Dr. Shunichi Muto (now at Hokkaido University) and Satoshi Nakamura for their useful discussion and their help in crystal growth. We also acknowledge Dr. Teruo Sakurai and Dr. Hajime Ishikawa for their encouragement.

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