Spin Relaxation in GaAs(110) Quantum Wells

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We investigated electron spin relaxation time τ_s in GaAs/AlGaAs (110) quantum wells (QWs), in which a predominant spin scattering mechanism [D'yakonov-Perel' (DP) mechanism] for conventional (100) QWs is substantially suppressed; τ_s in (110) QWs was of nanosecond order at room temperature, more than an order of magnitude longer than that of the (100) counterpart. The mechanism responsible for the spin relaxation was examined by studying the quantized energy, electron mobility, and temperature dependences of τ_s. The results suggest that in the absence of DP interaction, electron-hole exchange interaction limits τ_s in a wide temperature range ($\sim$0–300 K).

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Availability of spin degree of freedom has become a focus of interest in semiconductor electronics [1]. One of the key factors in making use of it is the relaxation time of the electron spins τ_s, which must be sufficiently long to process information stored in the form of the polarization of spin ensembles. To find a way to extend it, it is important to understand the spin relaxation mechanisms in semiconductor heterostructures such as quantum wells (QWs) which are designed so that spins can be appropriately confined and/or transferred [2]. Although recent time-resolved optical studies have highlighted some unique aspects of the electron spin dynamics in QWs [3–6], the mechanism that governs the spin lifetime has still not been fully understood [7]. The relative importance of possible spin relaxation mechanisms depends on a number of factors such as temperature, momentum relaxation time, and carrier type and density: in undoped GaAs/AlGaAs (100) QWs, for example, the D'yakonov-Perel' (DP) mechanism [8,9] is thought to be dominant at higher temperatures [10–12], while at lower temperatures or in highly p-doped QWs the Bir-Aronov-Pikus (BAP) mechanism [13,14] is believed to be enhanced [11]. The lack of inversion symmetry in III-V compounds of zinc-blende structure like GaAs results in spin splitting of the conduction band via spin-orbit coupling, which is the driving force for the spin relaxation in the DP theory [8]. Further decrease of symmetry and large momentum imposed by quantum confinement in (100) QWs enhances the DP interaction as compared to bulk [9]. Although no experiment has been reported prior to the present work, the DP interaction is expected to depend on the directions of electron momentum and spin in the host crystal, and by selecting the confinement axis appropriately, it may even be suppressed considerably in QWs.

In this Letter, we investigated the spin relaxation mechanism for two dimensional electron gas (2DEG) in III-V QWs. We report the observation that τ_s of 2DEGs in GaAs/AlGaAs QWs can be increased to nanosecond order even at room temperature (RT) by adopting [110] axis normal to the QW planes. Our results not only provide evidence that τ_s of GaAs (100) QWs, which is typically a few tens to a few hundred picoseconds, is limited by the DP interaction, but also show that (110) QWs offer an opportunity to study spin relaxation mechanisms that cannot be approached by studies on conventional (100) QWs or on bulk GaAs.

To explore the spin relaxation mechanism, we performed a systematic study of the dependence of τ_s on the characteristic parameters such as electron quantized energy E_1, electron mobility μ, and temperature T. We prepared four different GaAs/AlGaAs QW structures grown on undoped (110) GaAs substrates by molecular beam epitaxy (MBE). They consist of 60 periods of GaAs QWs separated by $Al_{0.4}Ga_{0.6}As$ barriers. For samples A–C the well width $L_W = 7.5$ nm is kept identical with the barrier thickness $L_B = 10$–12 nm. Sample A was not intentionally doped, while for samples B and C, Si donors were uniformly doped in the QW region. The electron density n per QW and μ evaluated by Hall measurements at RT were $n = 4.1 \times 10^{11}$ cm^{-2} and $\mu = 1800$ cm^2/V s for sample B, and $n = 1.4 \times 10^{12}$ cm^{-2} and $\mu = 1100$ cm^2/V s for sample C. Undoped sample D was grown without substrate rotation during MBE growth in order to intentionally introduce nonuniformity. This provides L_W of $\sim$6–9.5 nm. QW structures on (100) substrates were also prepared for comparison. Although the optimized MBE conditions for the growth of (Al,Ga)As on (100) and (110) orientations are quite different, there was no significant difference either in the linewidth of the exciton absorption peak or in the carrier recombination time τ_r between the (110) and the (100) QWs. For optical transmission measurements, opaque GaAs substrates were selectively removed.

For measurements of τ_s, we employed a degenerate pump-probe transmission configuration using a circularly polarized light. An optical pulse of $\sim$110 fs duration was generated by a mode-locked Ti:sapphire laser at a repetition rate of 76 MHz, and divided into pump and

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probe with the intensity ratio of 10:1. The samples were excited by a normal-incident right circularly polarized (σ^+) pump pulse at the heavy-hole exciton resonance, and the nonlinear change of the absorption was detected by measuring the transmission intensities of a delayed σ^+ or left circularly polarized (σ^-) probe pulse ($I^{\sigma\pm}$). Linear polarization measurements were also carried out to check τ_r of the photoexcited carriers.

In Fig. 1(a), the traces of $I^{\sigma\pm}$ for undoped (110) QWs (sample A) are plotted as a function of the time delay Δt. Since the hole spin relaxation takes place in the subpicosecond regime and because of the very fast exciton thermalization at RT [11], $I^{\sigma\pm}$ is attributed to the transients of "spin-down" or "spin-up" electron density. The step rise of $I^{\sigma-}$ at $\Delta t = 0$ has been attributed to the spin-independent Coulomb effect [15]. One can see in Fig. 1(a) that the net spin polarization initially induced by a σ^+ pump pulse is retained over the time scale $\Delta t \sim 500$ ps (which is limited by our experimental setup). In Fig. 1(b), for comparison, we plot $I^{\sigma\pm}$ measured in a (100) QW sample with the same structure as sample A. The traces of $I^{\sigma\pm}$ in Fig. 1(b) are nearly symmetric since $\tau_s \ll \tau_r$. The polarization is seen to be fully relaxed about $\Delta t \sim 100$ ps. The degree of polarization defined by $P = (I^{\sigma+} - I^{\sigma-})/(I^{\sigma+} + I^{\sigma-})$ is calculated using the data in Figs. 1(a) and 1(b), and shown in Figs. 1(c) and 1(d) by semilogarithmic plots. As indicated by solid lines, $P(\Delta t)$ can be fitted with a single exponential $P(\Delta t) = P_0 \exp(-2\Delta t/\tau_s)$, from which τ_s is determined. In Fig. 1(c), one reads $\tau_s = 2.1$ ns for (110) QW sample A, while $\tau_s \sim 70$ ps for a (100) reference sample as shown in Fig. 1(d).

This 30-fold increase of τ_s in (110) QWs can be attributed to the fact that the spin scattering is quite anisotropic and substantially suppressed when the spin-polarization vector axis is along [110], which is consistent with the DP theory [9]. According to the theory, spin relaxation is described as $dS_i/dt = -\sum_j \Gamma_{ij} S_j$, where $\boldsymbol{S}$ is the average spin of electrons, and the indices are x, y, and z. The relaxation tensor Γ_{ij} is given by

$$\Gamma_{ij} = \tau_{s0}^{-1}(\delta_{ij}\,\mathrm{Tr}\boldsymbol{B} - B_{ij}), \tag{1}$$

where τ_{s0} is the spin relaxation time for (100) QWs, and $\boldsymbol{B}$ is a function of a unit vector $\boldsymbol{n}$ normal to the QW plane, defined as

$$B_{xx} = (n_y^2 - n_z^2)^2 + 4n_x^2(n_y^2 + n_z^2) - 9n_x^2(n_y^2 - n_z^2), \tag{2}$$

$$B_{xy} = -2n_x n_y(n_x^2 + n_y^2) - 9n_x n_y(n_y^2 - n_z^2)(n_z^2 - n_x^2). \tag{3}$$

Other components are obtained by cyclic transposition of x, y, and z. As one can see, substituting $\boldsymbol{n} = (1/\sqrt{2}, 1/\sqrt{2}, 0)$ into Eqs. (1)–(3) leads to $dS_{(110)}/dt = 0$, i.e., $\tau_s = \infty$ for (110) QWs.

The results shown in Fig. 1 arouse an interesting question: what mechanism is now responsible for the observed slow spin relaxation in (110) QWs? Although the dominant term vanishes in the two-dimensional DP interaction, small (bulklike) contribution to τ_s may still remain in (110) QWs [9]. Another candidate is the Elliott-Yafet (EY) mechanism [16], which is based on the fact that the wave functions of the conduction band (except for the band edge) are not spin eigenstates due to the spin-orbit coupling. Moreover, electron-hole scattering (BAP mechanism) [13] and/or exciton spin relaxation [17] might be of importance in this time scale.

To identify the spin relaxation mechanism, we experimentally investigated the E_1, μ, and T dependences of τ_s. The E_1 dependence of τ_s measured at several points on nonuniform (110) QWs (sample D) are shown in Fig. 2. Here, E_1 was obtained from the excitonic peak in the absorption spectra. The least squares fit results in $\tau_s \propto E_1^{-1.0}$. Figure 3(a) shows the μ dependence of τ_s of n-type (110) QWs of the same L_W but with different μ (samples B and C) at RT. Also shown in Fig. 3(a) are τ_r of (110) QWs measured by linear polarization configuration and τ_s of QWs on (100) substrates. Note that $\tau_s > \tau_r$ for these n-doped (110) QWs. Contrary to the (100) case, where $\tau_s \propto \mu^{-1}$ is observed in accordance with a previous study on μ dependence of τ_s [18], τ_s of (110) QWs increases with the increase of μ. Figure 3(b) shows the absorption spectra of undoped (A), $\mu = 1800$ cm^2/V s (B), and $\mu = 1100$ cm^2/V s (C) samples measured at RT. The degradation of excitonic peak is observed as n doping increases from undoped (A) through moderate doping (B) to heavy doping (C). Finally, Fig. 4 presents the T dependence of τ_s observed in undoped (110) QWs (sample A). As shown in Fig. 4, one observes

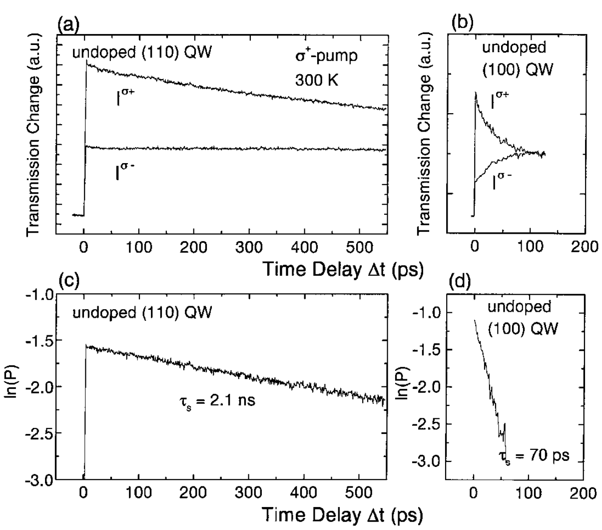

FIG. 1. Nonlinear transient of transmission $I^{\sigma\pm}$ for undoped GaAs/AlGaAs (110) QWs (sample A) (a) and (100) QWs with the same structure (b). (c) and (d) show the traces of polarization $P(\Delta t) = (I^{\sigma+} - I^{\sigma-})/(I^{\sigma+} + I^{\sigma-})$ and the spin relaxation time τ_s.

two different temperature regimes below and above $T = 20$ K. A distinct feature is that τ_s of (110) QWs *increases* with T in a power law $\tau_s \propto T^{0.6}$ when $T > 20$ K, while below <20 K τ_s almost saturates at ~ 300 ps. The inset of Fig. 4 shows the excitation intensity I^{ex} dependence of τ_s in the case of undoped (110) QWs at RT. The data shown in Figs. 1–4 were taken with I^{ex} fixed at averaged power density of 20 W/cm^2 in front of the sample, corresponding to ~ 2 mW in the inset of Fig. 4.

In the following, we examine the existing theories to identify the possible spin relaxation mechanism in (110) QWs, using the dependence of τ_s on the experimentally varied parameters. First, we recall the case of GaAs/AlGaAs (100) QWs. The theory of DP mechanism in two dimensions predicts $\tau_s \propto E_1^{-2}\tau_p^{-1}T^{-1}$ [9], where τ_p is the momentum relaxation time, while for the bulk three-dimensional DP interaction $\tau_s \propto \tau_p^{-1}T^{-3}$ [8]. It was shown experimentally that τ_s depends on E_1 as $\tau_s \propto E_1^{-2.2}$ [10] in undoped (100) QWs at RT, and $\tau_s \propto \mu^{-1}$ holds in the range $\mu > 700$ cm^2/V s in n-GaAs (100) QWs [18,19] [see also Fig. 3(a)], all in agreement with the theory. On the other hand, τ_s increases with the increase of μ in (110) QWs, qualitatively different from the case of (100) QWs. In addition, we observed positive T dependence in (110) QWs, whereas, by taking into account the T dependence of the ionized impurity and optical-phonon scatterings, the DP theory predicts negative T dependence of τ_s. From these observations, we can rule out the DP mechanism from the mechanisms responsible for the spin relaxation in (110) QWs.

Next we consider the EY mechanism. An analytic expression for τ_s determined by the EY spin scattering in (100) QWs has been derived from that for the three dimensional case [13,20] as $\tau_s \sim (9/16)(E_g^4\tau_p)/(\Delta^2 E_1 k_B T)$, where Δ is the spin split-off energy, k_B the Boltzmann constant, and E_g the energy gap. Although $\tau_s \propto E_1^{-1}$ at RT (see Fig. 2) and τ_s increases with the increase of μ, as are expected from the EY theory, the theory predicts negative T dependence. For 2DEGs, the EY results in $\tau_s \propto \tau_p T^{-1}$. Again taking into account the T dependence of the ionized impurity and optical-phonon scatterings, we expect that τ_p increases as T decreases in the high temperature regime ($T > 70$ K) for undoped GaAs QWs. Thus, the EY theory fails to explain the observed τ_s vs T shown in Fig. 4.

The third mechanism we consider is the spin relaxation due to electron-hole exchange interaction. The increase of τ_s from its undoped value of 2.1 ns (sample A) to 4.0 ns by moderate n doping (sample B) suggests the importance of Coulombic interaction on determining τ_s: the heavy-hole exciton peak at the absorption edge is seen to degrade gradually as n increases in Fig. 3(b), because of the screening of Coulomb attraction between the photoexcited carriers. The dependence $\tau_s \propto E_1^{-1}$ shown in Fig. 2 can also be understood by the fact that electron-hole interaction is enhanced by narrowing the spatial confinement of wave functions: a numerical calculation shows that $\tau_s \propto L_W^{\beta} \propto E_1^{-\beta/2}$ with $1 < \beta < 2$ [17]. We also note that τ_s of (110) QWs depends on the excitation intensity I^{ex}: as shown in the inset of Fig. 4, τ_s decreases as I^{ex} *increases*, whereas τ_s of (100) QWs is *independent* of I^{ex}. The I^{ex} dependence suggests that electron-hole interaction is important in (110) QWs.

Two theoretical frameworks have so far been proposed: the BAP interaction based on the spin-flip scattering of free electrons in a sea of spin-depolarized holes via exchange interaction [13,14], and exciton spin relaxation, where the excitonic nature of the wave function on spin-flip scattering is taken into account [17]. To examine the theoretical models, we first focus on the T dependence of τ_s at $T < 20$ K shown in Fig. 4. The saturation of T dependence of τ_s has also been observed in (100) QW: the origin of the spin relaxation in this temperature regime is considered to be BAP or exciton spin relaxation [7]. One can distinguish between the two processes (BAP or exciton spin relaxation) by examining whether or not electrons and holes have the same spin relaxation time. In the present experiment, $P(\Delta t)$ ($0 < \Delta t < 0.5$ ns) of

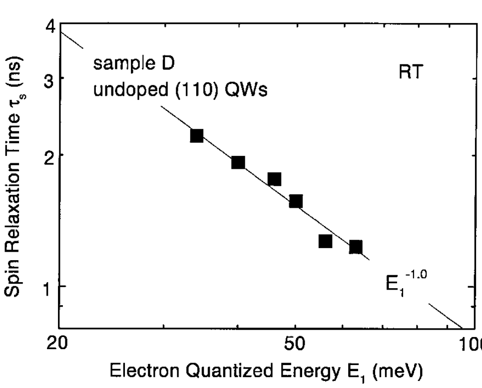

FIG. 2. Quantized energy E_1 dependence of τ_s measured at several points of undoped (110) QWs (sample D) at room temperature. The solid line is the least squares fit ($\tau_s \propto E_1^{-1.0}$).

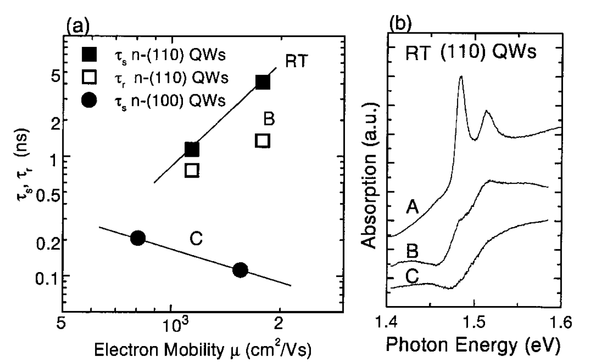

FIG. 3. (a) Solid squares are τ_s of n-GaAs/AlGaAs (110) QWs (samples B and C) plotted against μ at room temperature. Open squares indicate τ_r of samples B and C. Solid lines are guides for eyes. Solid circles are τ_s of n-GaAs/AlGaAs (100) QWs measured in the same configuration. (b) The absorption spectra of samples A, B, and C at RT.

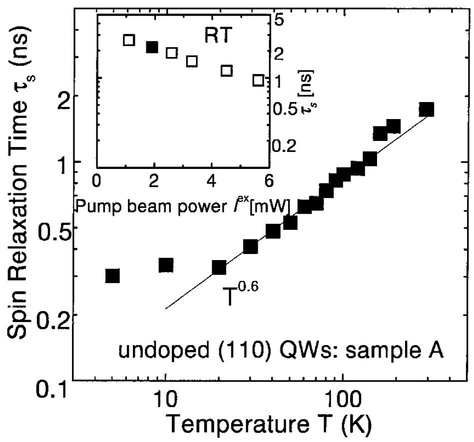

FIG. 4. The temperature (T) dependence of τ_s for undoped (110) QWs (sample A). $\tau_s \propto T^{0.6}$ is the fitting curve above 20 K. The inset shows the excitation intensity I^{ex} dependence of τ_s measured at RT. (The data in Figs. 1–4 were obtained at $I^{ex} \sim 2$ mW indicated by the solid symbol in the inset.)

sample A is a single exponential decay function. This suggests that when $T < 20$ K the exciton spin relaxation is predominant rather than the BAP process: electron spin is scattered simultaneously with that of holes.

As T increases, hole spin relaxation rate is enhanced [11], which should result in higher spin relaxation rate of excitons, contrary to what was observed in Fig. 4. Therefore, the exciton spin relaxation alone cannot explain the positive T dependence. The BAP mechanism cannot account for the positive temperature dependence either, since the T dependence of τ_s is generally predicted to be $\tau_s \propto 1/T^{\beta}$ with $\beta > 0$ [13,14].

Although two existing frameworks fail, there may yet be another scenario based on the spin scattering due to exchange interaction: if thermal ionization of excitons takes place more rapidly as T increases, i.e., the dissociation of excitons has stronger T dependence than the hole spin relaxation rate, τ_s may show positive T dependence, provided that τ_s of free electrons is reasonably long, as has been observed in undoped ZnCdSe/ZnSe QWs [2,21].

So far we have not taken into consideration the crystalline axis dependence for spin relaxation mechanisms other than the DP interaction. Although no corresponding theory has been developed, we expect that EY and electron-hole exchange relaxation rates change less drastically compared to the case of DP mechanism when [110] direction is taken as the polarization axis. We also note that 100% pure spin excitation cannot be achieved in (110) QWs due to the mixing of valence band states [22].

In conclusion, we investigated the spin relaxation mechanism of 2DEGs in GaAs/AlGaAs (110) QWs, where the DP interaction is suppressed. The unexpected temperature dependence $\tau_s \propto T^{0.6}$, observed in an undoped sample when $T > 20$ K, cannot be explained within the simple framework of EY or bulklike DP mechanism, while exciton spin relaxation seems to govern τ_s when $T < 20$ K. The observed $\tau_s(T)$ might reflect the thermal ionization process of excitons with increasing T, resulting in the increase of τ_s with T.

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