

Anomalous Spin Dephasing in (110) GaAs Quantum Wells: Anisotropy and Intersubband Effects

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(Received 29 September 2003; published 29 September 2004)

A strong anisotropy of electron spin decoherence is observed in GaAs/(AlGa)As quantum wells grown on a (110) oriented substrate. The spin lifetime of spins perpendicular to the growth direction is about one order of magnitude shorter compared to spins along [110]. The spin lifetimes of both spin orientations decrease monotonically above temperatures of 80 and 120 K, respectively. The decrease is very surprising for spins along the [110] direction and cannot be explained by the usual Dyakonov-Perel dephasing mechanism. A novel spin dephasing mechanism is put forward that is based on scattering of electrons between different quantum well subbands.

DOI: 10.1103/PhysRevLett.93.147405

PACS numbers: 78.66.Fd, 73.50.-h, 78.47.+p, 85.75.-d

The electron spin in semiconductors has recently become a focus of intense research in the context of spin electronics or spintronics. This new kind of electronics aims to utilize spin for devices with unprecedented properties [1–3]. A prime condition for the development of potential applications is the understanding of spin decoherence, i.e., the loss of spin memory, in semiconductor structures [4]. The main reason for spin decoherence at room temperature is the intrinsic spin splitting of the conduction band, which occurs in all binary semiconductors. The spin splitting, which acts as an effective magnetic field, depends on the electron's momentum and is the basis for the Dyakonov-Perel (DP) spin relaxation mechanism [5,6]. Semiconductor heterostructures are, in this context, of particular interest since spin splitting in conduction and valence band can be controlled via dimensionality and orientation of crystal axes [7]. Ohno *et al.* observed very long electron spin decoherence times at room temperature in GaAs quantum wells (QWs) grown on (110) oriented substrates that exceeded the coherence times in the usual (100) grown QWs by more than one order of magnitude [8,9]. However, slow spin dephasing in (110) QWs was demonstrated only for electron spins oriented along the crystal growth direction. The dynamics of in-plane spin was left unexplored.

The starting point for the theoretical description of the spin dynamics in (110) QWs is the Dresselhaus-Hamiltonian for binary semiconductors

$$H_{\text{spin}} = \Gamma \sum_i \sigma_i k_i (k_{i+1}^2 - k_{i+2}^2), \quad (1)$$

where $i = x, y, z$ are the principal crystal axes with $i + 3 \rightarrow i$, Γ is the spin-orbit coefficient for the bulk semiconductor, and σ_i are the Pauli spin matrices [10]. Comparing Eq. (1) with the spin Hamiltonian for a free electron in a magnetic field ($H = \frac{1}{2} \sum_i \mu_B \sigma_i B_i$), one easily recognizes that random scattering of electrons leads to an effective k dependent random magnetic field with com-

ponents in the x , y , and z directions. This random magnetic field destroys the average spin orientation of an ensemble of electrons by rotating individual spins in different directions. The DP effect increases in bulk semiconductors with temperature due to occupation of higher k states with larger spin splittings despite a motional narrowing effect at higher temperatures (the spin lifetime τ_s is inversely proportional to the momentum scattering time τ_p^*). In (110) QWs, however, the spin splitting (effective magnetic field)

$$H_{\text{DP}} = -\Gamma \sigma_z k_x [\frac{1}{2} \langle k_z^2 \rangle - \frac{1}{2} (k_x^2 - 2k_y^2)] \quad (2)$$

is suppressed in the plane of the QW via averaging of H_{spin} along the confinement direction $z \parallel [110]$ (Ref. [10]). Here, $\langle k_z^2 \rangle = \int |\nabla \psi_z|^2 dz$, ψ_z denotes the z part of the electron wave function in the lowest subband, $x \parallel [1\bar{1}0]$, and $y \parallel [001]$. An effective magnetic field exists only along the growth direction z . Electron spins oriented along z are therefore not affected by the DP mechanism. In contrast, electrons with a spin component along x or y will randomly precess around the effective field, giving rise to spin dephasing anisotropy with dephasing rates $\gamma_x = \gamma_y \gg \gamma_z$.

In the first part of this Letter we report on the observation of such a huge dephasing anisotropy with fast dephasing of electron spins oriented in the plane of a (110) QW and long dephasing times for spins along the growth direction. The sample under investigation is a symmetrically modulation doped GaAs/Al_{0.4}Ga_{0.6}As multiple quantum well (MQW) with ten wells of 20 nm thickness and an electron density of $n = 1.1 \times 10^{11} \text{ cm}^{-2}$ per well. The sample is mounted in a magnet cryostat with variable temperature. The magnetic field lies in the plane of the MQW along the y direction. Spin dephasing rates are determined by means of time and polarization resolved optical photoluminescence (PL) measurements. Spin oriented electrons are optically created in the sample along the growth direction by circularly polarized

100 fs pulses from a mode locked Ti:sapphire laser with a repetition rate of 80 MHz. The PL energy shifts with temperature from 1.523 eV at 6 K to 1.428 eV at 314 K. The excitation energy is fixed at 1.675 eV. After excitation the carrier momentum distribution rapidly thermalizes by emission of phonons and scattering with other carriers. Holes rapidly lose their spin orientation due to strong valence band mixing and k dependent spin splitting. The polarized luminescence is spectrally and temporally resolved by a synchroscan streak camera with two-dimensional readout which provides a resolution of 1 nm and 20 ps, respectively. Polarization is selected by a liquid crystal retarder and a polarizer. The optical selection rules yield a proportionality between the degree of circular PL polarization and the degree of spin orientation, with the opposite sign for heavy hole (hh) and light hole (lh) transitions [6]. Photoluminescence lifetimes increase with temperature from 500 ps at 6 K to 9 ns between 200 and 300 K, which is sufficiently long for a precise determination of spin relaxation times [11].

Figure 1 depicts the transient decay of circular polarization after excitation of about 2×10^{10} carriers per cm^2 at a sample temperature of 200 K. For $B = 0$, the average spin direction is always oriented along the z direction where the DP mechanism does not apply. We find a long spin lifetime $\tau_z = \gamma_z^{-1} = 3700$ ps as reported by Ohno [8]. For $B > 0$ we observe spin precession around the field, which leads, according to the optical selection rules, to a periodic change between σ^- and σ^+ polarized luminescence in the z direction [12]. The magnetic field forces the spin after a quarter precession period in the plane of the QW, where the effective fluctuating magnetic field rapidly randomizes the spin orientation of the ensemble of electrons. We measure for the precessing spin an average dephasing rate $(\gamma_z + \gamma_x)/2 = 1/670$ ps^{-1} which is by a factor of 5 faster than for spin in the z direction.

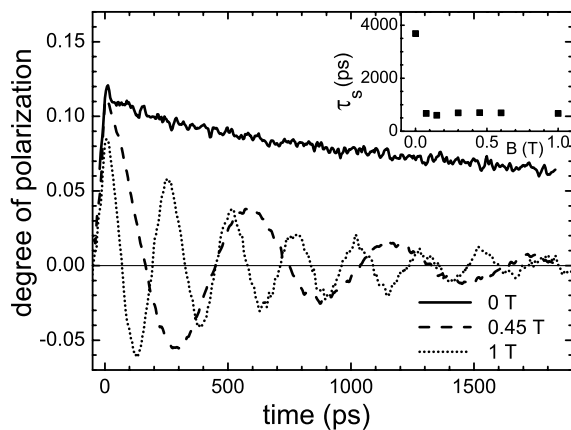


FIG. 1. Degree of polarization of the time resolved PL for magnetic fields of 0 (solid line), 0.45 (dashed line), and 1 T (dotted line) measured at a temperature of 200 K. Inset: Dependence of measured spin lifetime τ_s on magnetic field.

The dephasing rate $\gamma_x = 1/370$ ps^{-1} is even 10 times larger than γ_z . Such a strong anisotropy in a semiconductor structure has never been reported before [13]. The rapid dephasing rates γ_x and γ_y have direct negative implications for devices where the stability of spins with arbitrary orientation is important, as in qubits for quantum computation. We measure the magnetic field dependence of the spin dephasing time to verify that the fast spin dephasing is not a new dephasing effect caused by the external magnetic field or inhomogeneous broadening of the precession frequency (inset of Fig. 1). The almost constant spin lifetime for $B > 0$ clearly excludes a direct influence of the magnetic field on spin dephasing.

Next, we systematically investigate the spin lifetimes for temperatures between 6 and 314 K keeping the excitation power of the laser fixed. Figure 2 depicts the temperature dependence of the spin lifetimes at $B = 0.6$ T and $B = 0$ for the full temperature range. Two features, the weak dephasing anisotropy γ_x/γ_z (see inset) at low temperatures and the decreasing spin lifetime for $B = 0$ at temperatures above 120 K, are discussed in the following. At low temperatures the spin dephasing is governed by the Bir-Aronov-Pikus (BAP) mechanism where the randomly oriented hole spins act via exchange coupling with electron spins like a fluctuating effective magnetic field in all directions [14]. The main contribution of the effective magnetic field arises from the $1s$ exciton where the electron hole overlap is at maximum [15]. In a quantum well the z component of the effective magnetic field is dominant, which explains the anisotropy $\gamma_x/\gamma_z > 1$ at low temperatures [16]. Higher temperatures break the strong spatial correlation of electrons and holes which leads to the observed increase in spin lifetime up to 80 K. Another clear hint for the dominance of dephasing via the BAP mechanism at low temperatures is found in

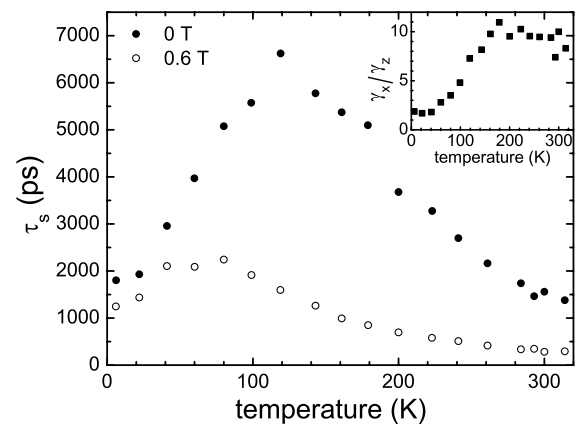


FIG. 2. Temperature dependence of spin lifetime τ_s for $B = 0$ (closed circles) and $B = 0.6$ T (open circles). Inset: Corresponding temperature dependence of spin relaxation anisotropy γ_x/γ_z .

the time resolved PL transients, where we observed a retarded subexponential decay of the degree of spin polarization. Here the BAP mechanism decreases in time due to the loss of holes by optical recombination (not shown) [17].

For $B = 0.6$ T and $T > 80$ K, the DP mechanism becomes stronger than the BAP mechanism and leads to the expected reduced spin lifetime $[(\gamma_x + \gamma_z)/2]^{-1}$. In contrast, the decrease of spin lifetime γ_z^{-1} for $B = 0$ and $T > 120$ K is very surprising, since all subbands strictly exhibit no in-plane component of the effective magnetic field. Occupation of higher subbands at elevated temperatures (about 22% of the electrons at 300 K) should also not lead to spin dephasing. This can be seen by transforming Eq. (1) into the (110) coordinate system

$$H_{\text{spin}} = \frac{1}{2}\hbar[\sigma_x\Omega_x + \sigma_y\Omega_y + \sigma_z\Omega_z], \quad (3)$$

where $(\Omega_x, \Omega_y, \Omega_z) = \Gamma/\hbar(-k_x^2k_z - 2k_y^2k_z + k_z^3, 4k_xk_yk_z, k_x^3 - 2k_xk_y^2 - k_xk_z^2)$. Since $\langle k_z \rangle = \langle k_z^3 \rangle = 0$ for all subbands, all in-plane magnetic field components Ω_x and Ω_y vanish. A simple application of the DP dephasing formula $\gamma_z = \langle \Omega_x^2 + \Omega_y^2 \rangle \tau_p^* = 0$ yields no spin dephasing [6].

In the following we resolve the puzzle by noting that scattering of electrons between subbands along with spin-orbit coupling H_{spin} constitutes a new spin relaxation mechanism. Existing theories treat spin dephasing only within the lowest subband [18–21]. Here, we sketch the principle idea for intersubband spin relaxation (ISR) and give a first estimate of its effectiveness. In the case $H_{\text{spin}} = 0$ a simultaneous intersubband and spin flip transition like $|\vec{k}, 0, \uparrow\rangle \rightarrow |\vec{k}', 1, \downarrow\rangle$ is forbidden. However, for $H_{\text{spin}} \neq 0$ the corresponding transition $|\vec{k}, 0, \uparrow\rangle + \epsilon_1|\vec{k}, 1, \downarrow\rangle + \dots \rightarrow |\vec{k}', 1, \downarrow\rangle + \epsilon_2|\vec{k}', 0, \uparrow\rangle + \dots$ is modified according to first order perturbation theory which mixes zero order terms with first order terms possessing opposite spin. Therefore, spin flip transitions become allowed with rates that are $\alpha = |\epsilon_1|^2 + |\epsilon_2|^2$ slower than the intersubband transition rate τ_{IB}^{-1} , i.e., $\tau_s \approx \alpha^{-1}\tau_{\text{IB}}$. We obtain the first order coefficient $\epsilon_1(k_x, k_y) = \langle \vec{k}, 0, \uparrow | H_{\text{spin}} | \vec{k}, 1, \downarrow \rangle / E_{\text{ISG}}$ using a spin splitting constant $\Gamma = 19.55$ eV \AA^3 [22] and an intersubband gap of $E_{\text{ISG}} = 31$ meV. The z wave functions are approximated with those of a well with infinite barriers. We find a spin lifetime $\tau_s = \langle \alpha \rangle^{-1}\tau_{\text{IB}} = 1.7$ ns, for an intersubband scattering rate $\tau_{\text{IB}} = 250$ fs and a distribution of electrons at 300 K with averaged α (effective mass $m^* = 0.0665m_0$). The estimate is in good agreement with our measurements. Though related to the Elliott-Yafet (EY) spin relaxation mechanism, ISR is unlike EY based on the Dresselhaus spin splitting and cannot be derived within existing EY theory [6]. ISR depends strongly on the occupation of higher subbands and is much weaker in narrow quantum wells where intersubband splitting is

large. This is in accord with recent measurements in narrow n -doped quantum wells where spin lifetimes of more than 10 ns were limited not by ISR but by the BAP mechanism [23]. We note that our new dephasing mechanism may also explain a result from Ohno where he found an increased spin lifetime in higher mobility (110) samples at room temperature ($\tau_s \propto \tau_p^*$ or τ_{IB} , respectively), which he could not explain with the usual DP result $\tau_s \propto (\tau_p^*)^{-1}$ [8].

Decreasing spin lifetimes for elevated temperatures have also been reported in (110) grown samples with external electric field. This field induces a Rashba term $(\Omega_x, \Omega_y \neq 0)$, which leads via the DP mechanism to spin relaxation [24]. In our sample, the Rashba term should be very weak or absent since asymmetry of the QW or a built-in electric field in growth direction is suppressed by symmetric growth and symmetric doping. Moreover, we found no experimental evidence for the presence of a Rashba contribution, which should reveal itself in spin lifetimes that strongly depend on the orientation of a tilted external magnetic field $[B_x \cos(\theta), B_y \sin(\theta), B_z]$ if $B_z \neq 0$. And even if some Rashba related spin relaxation was present in our sample, the above estimate shows that the novel ISR mechanism contributes substantially.

In the last paragraph we argue and experimentally verify that anisotropic spin dephasing modifies the usual linear relation $\omega = g\mu_B B/\hbar$ between magnetic field B and measured frequency ω of spin oscillations, where μ_B is Bohr's magneton and g is the constant Landé g factor. The time dependent degrees of spin polarization s_x and s_z for spins that precess around a magnetic field oriented along y are given by

$$\frac{\partial}{\partial t} \begin{pmatrix} s_x \\ s_z \end{pmatrix} = - \begin{pmatrix} \gamma_x & -\omega \\ \omega & \gamma_z \end{pmatrix} \begin{pmatrix} s_x \\ s_z \end{pmatrix}, \quad (4)$$

where γ_x and γ_z are the relevant components of the spin dephasing tensor. The solution for $s_z(t)$ which is measured in our experiments is

$$s_z(t) = s_0 \frac{e^{-(\gamma_x + \gamma_z)t/2}}{\cos\varphi} \cos(\omega_a t - \varphi), \quad (5)$$

where $\tan\varphi = \frac{\gamma_x - \gamma_z}{2\omega_a}$, $\omega_a = \sqrt{\omega^2 - (\gamma_x - \gamma_z)^2/4}$, and $s_z(0) = s_0$ is the initial degree of spin polarization. The effective spin oscillation frequency ω_a is slowed down by the dephasing anisotropy $\gamma_x - \gamma_z$. Figure 3(a) depicts the measured modified oscillation frequency ω_a for magnetic fields between 0.075 and 1 T along with a fit of $\omega_a(B)$. The value of $\gamma_x - \gamma_z = 1/380$ ps $^{-1}$ obtained from the fit compares very well with the expected value of $1/410$ ps $^{-1}$ extracted from the data in Fig. 1. The most significant influence of anisotropic spin dephasing is found at magnetic fields below 0.1 T. Figures 3(b)–3(d) show the measured time resolved degree of spin polarization along with the theoretical curves including an-

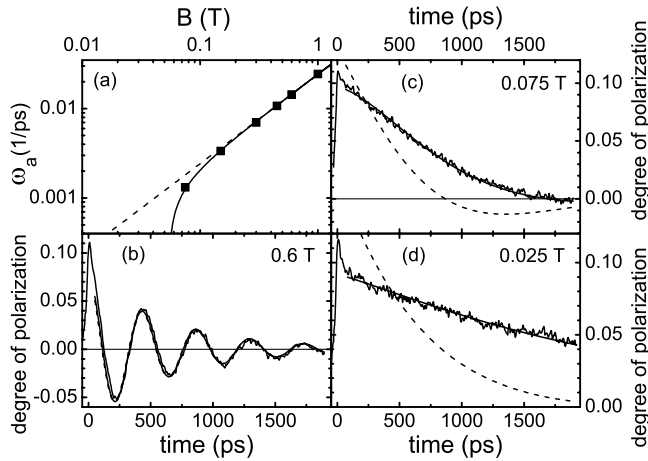


FIG. 3. (a) Measured spin oscillation frequency ω_a vs magnetic field (closed squares) at $T = 200$ K. The fitting curve includes effects of anisotropic spin dephasing (black line). The dashed line depicts for comparison the spin oscillation frequency for isotropic spin dephasing. (b)–(d) Transients of the degree of polarization of time resolved PL for magnetic fields of 0.6, 0.075, and 0.025 T. The solid lines are fits including effects of anisotropic spin dephasing. The dashed lines show fits assuming no anisotropy.

isotropy [Eq. (5)] and without anisotropy, i.e., $s_z(t) = s_0 e^{-(\gamma_x + \gamma_z)t/2} \cos(\omega t)$, where we used the initial degree of polarization s_0 as the only fit parameter. The values for the parameters used for calculating the transients were obtained as follows: γ_z by fitting the transient at $B = 0$ and g and γ_x by fitting the transients at elevated magnetic fields (0.075 to 1 T), assuming validity of anisotropic spin dephasing. While the transient at $B = 0.6$ T is matched by both theoretical curves [Fig. 3(b)], the transients for $B = 0.075$ and 0.025 T can be explained only by including anisotropic spin dephasing. We note that for the case of $|\omega| < |\gamma_x - \gamma_z|/2$, i.e., $B < 0.05$ T, spin oscillations are completely suppressed.

In conclusion, we investigated anisotropic spin dephasing in (110) QWs from 6 K up to room temperature. Spin memory at room temperature lasts for times as long as nanoseconds only if the spin is parallel or antiparallel to the growth direction. Spin coherence in the plane of the QW is rapidly lost by the DP mechanism. These results have an immediate impact on design considerations for spintronic devices. We further found that intersubband scattering constitutes a new spin dephasing mechanism that is necessary to explain reduced spin lifetimes at elevated temperatures for electron spins along [110]. We expect that this new mechanism will stimulate the development of a more general theory of spin dephasing in QWs including the influence of higher subbands. Finally we demonstrated the modification and total suppression of spin oscillations via anisotropic spin dephasing.

We gratefully acknowledge most helpful discussions with R. Winkler and financial support by BMBF and DFG (German Science Foundation).

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