Spin dynamics in high-mobility
two-dimensional electron systems

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Abstract. Understanding the spin dynamics in semiconductor heterostructures is highly important for future semiconductor spintronic devices. In high-mobility two-dimensional electron systems (2DES), the spin lifetime strongly depends on the initial degree of spin polarization due to the electron-electron interaction. The Hartree-Fock (HF) term of the Coulomb interaction acts like an effective out-of-plane magnetic field and thus reduces the spin-flip rate. By time-resolved Faraday rotation (TRFR) techniques, we demonstrate that the spin lifetime is increased by an order of magnitude as the initial spin polarization degree is raised from the low-polarization limit to several percent. We perform control experiments to decouple the excitation density in the sample from the spin polarization degree and investigate the interplay of the internal HF field and an external perpendicular magnetic field. The lifetime of spins oriented in the plane of a [001]-grown 2DES is strongly anisotropic if the Rashba and Dresselhaus spin-orbit fields are of the same order of magnitude. This anisotropy, which stems from the interference of the Rashba and the Dresselhaus spin-orbit fields, is highly density-dependent: as the electron density is increased, the cubic Dresselhaus term becomes dominant and reduces the anisotropy.

1 Introduction

In recent years, semiconductor spintronics [1, 2, 3] research has found increased interest, in part due to new materials like ferromagnetic semiconductors [4]. Among the key requirements for semiconductor spintronic devices is an understanding of the spin dephasing mechanisms in semiconductors. For GaAs, many experimental studies have focused on slightly n-doped bulk material, where extremely long spin lifetimes (≥ 100 ns) were observed [5, 6] for doping levels close to the metal-insulator transition. Even though this material has low mobility and poor conductivity properties, it was used in a number of spin injection [7] and transport [8, 9] experiments.

Relatively few studies have been performed on high-mobility two-dimensional electron systems (2DES): Brand et al. investigated the weak scattering regime and the temperature dependence of the D’Yakonov-Perel (DP) mechanism in a high-mobility 2DES [10], while Leyland et al. [11] experimentally showed the importance of electron-electron collisions for spin dephasing. Several groups
performed experiments on [110]-grown quantum wells and 2DES, in which the DP mechanism can be suppressed for spins aligned along the growth direction [12], while it remains active for other spin orientations [13, 14]. In these systems, gate control of the spin lifetime was demonstrated by Karimov et al. [15]. Theoretical studies investigating the electron-electron interaction in 2DES were performed using perturbation theory by Glazov and Ivchenko [16], while Wu et al. developed a powerful microscopic many-body approach [17] to study 2DES far from thermal equilibrium, considering all relevant scattering mechanisms, including electron-hole and electron phonon scattering [18, 19, 20, 21].

2 Theory

2.1 Optical orientation of electrons in a 2D electron system

In GaAs, the conduction band has s-like character, while the valence bands have p-like character. The light-hole (LH $J_z = \pm \frac{1}{2}$) and the heavy-hole (HH $J_z = \pm \frac{3}{2}$) valence bands are degenerate at $k=0$ for bulk GaAs. Optical excitation above the bandgap can generate electron-hole pairs through photon absorption. Due to angular momentum conservation, a circularly-polarized photon ($s=1$) may excite both, electrons with spin up (from a light-hole valence band state with $J_z = -\frac{1}{2}$) and spin down (from a heavy hole valence band state with $J_z = -\frac{3}{2}$). Due to the different probabilities of these transitions, a finite spin polarization may be created in bulk GaAs [22]. In a quantum well, the $k=0$ degeneracy of the light and heavy hole valence bands is lifted due to confinement. By resonantly exciting only the HH or LH transition in a quantum well (QW) with circularly-polarized light, almost 100 percent spin polarization may be generated [23]. If a 2DES is created within the QW by (modulation) doping, the electrons occupy conduction band states up to the Fermi energy. Optical absorption is only possible into states above the Fermi energy, and does not occur at $k = 0$. For $k \neq 0$, the valence band states in a QW consist of an admixture of heavy-hole and light-hole states and $J_z$ is no longer a good quantum number [24]. Therefore, excitation with circularly polarized light will yield a mixture of spin-up and spin-down electrons in the conduction band, depending on the excitation wavelength. Additionally, the 2D electron system is typically unpolarized in thermal equilibrium. In order to create a large spin polarization in a 2D electron system by optical excitation, the excitation density therefore has to be on the order of the 2DES density if we assume that the 2D system returns to thermal equilibrium in between excitation pulses. The initial spin polarization degree $P$, created by a short optical pulse, may be calculated using the following formula:

$$P = \frac{n_{ph}}{n_e + n_{ph}^{tor}}.$$
Here, \( n_{\text{ph}} = \xi \cdot n_{\text{tot}}^{\text{ph}} \) is the spin-polarized fraction \( \xi \) of the optically created electron density, \( n_{\text{tot}}^{\text{ph}} \) is the total electron density that is optically created, and \( n_e \) is the background electron density of the 2DES.

### 2.2 Rashba and Dresselhaus spin-orbit fields in a [001] quantum well

In crystal structures which lack inversion symmetry, like GaAs, the spin-orbit interaction may be described by an intrinsic, \( k \)-dependent magnetic field \( B_i(k) \), which causes a precession of the electron spin. Typically, a Larmor precession frequency vector corresponding to the electron precession about this internal field is defined as \( \Omega(k) = \frac{\gamma}{h} \cdot \mu_B B_i(k) \). In bulk structures, \( \Omega(k) \) is cubic in \( k \) and has the following form [25]:

\[
\Omega(k)^{\text{BIA}} = \frac{\gamma}{h} \cdot \left[ k_x (k_y^2 - k_z^2), k_y (k_z^2 - k_x^2), k_z (k_x^2 - k_y^2) \right].
\]

This term stems from the inversion asymmetry of the crystal lattice and is therefore often called bulk inversion asymmetry (BIA) term or Dresselhaus term. In a quantum well grown along the \( z \) direction, the momentum along the growth direction is quantized due to confinement. In first approximation, the expectation value is \( \langle k_z^2 \rangle = (\pi/d)^2 \), where \( d \) is the quantum well thickness. It follows that:

\[
\Omega(k)^{\text{BIA(2D)}} = \frac{\gamma}{h} \cdot \left[ k_z (k_y^2 - \langle k_z^2 \rangle), k_y (\langle k_z^2 \rangle - k_x^2), 0 \right].
\]

Typically in a 2D electron system, the in-plane momentum \( (k_y)^2 \) is smaller than \( \langle k_z^2 \rangle \). Therefore, terms cubic in the in-plane momentum are often neglected, resulting in the following approximation:

\[
\Omega(k)^{\text{Dressel}} = \frac{\beta}{h} \cdot [-k_x, k_y, 0].
\]

This is linear in the in-plane momentum \( k_\parallel \) and therefore typically called linear Dresselhaus term. Its symmetry is shown in Fig. 1 (a). Additionally, a lack of structure inversion symmetry along the quantum well growth direction causes a second intrinsic effective magnetic field, the so-called Rashba field [26].

\[
\Omega(k)^{\text{Rashba}} = \frac{\alpha}{h} \cdot [k_y, -k_x, 0].
\]

Structure inversion asymmetry (SIA) along the growth direction may be induced by different barrier materials on either side of the quantum well, single-sided or asymmetric modulation doping resulting in an effective electric field due to ionized donors, or the application of an external electric field by a gate voltage. The symmetry of the Rashba field is shown in Fig. 1 (b).
2.3 Spin dephasing/relaxation

The main mechanism for spin dephasing in GaAs bulk and quantum well structures at low temperatures is the so-called D’Yakonov-Perel mechanism [27]. It is caused by the \( k \)-dependent spin-orbit fields which cause a precession of the electron spin. In an ensemble of electrons, the \( k \) values are distributed according to Fermi statistics, causing different precession frequencies and directions for the electron spins, which leads to dephasing. Two regimes can be distinguished according to the relationship between the average precession frequency \( \bar{\Omega} \) and the momentum relaxation time \( \tau_P \):

1. weak scattering regime \((\bar{\Omega} \cdot \tau_P > 1)\): here, the electron spins may precess more than one full cycle about the spin-orbit field before they are scattered and their \( k \) value changes. A collective precession of electron spins may be observed in this regime.

2. strong scattering regime \((\bar{\Omega} \cdot \tau_P < 1)\): here, the electrons are scattered so frequently that the spin-orbit field acts like a rapid fluctuation. In this regime, the spin relaxation time \( T_2^* \) is inversely proportional to the momentum relaxation time: \( \frac{1}{T_2^*} \propto \tau_P \).

2.4 Magneto-Anisotropy

In samples where both the Rashba and the Dresselhaus terms are present and of the same magnitude, the effective spin-orbit field has to be calculated as the vector sum of the two terms:

\[
\Omega(k)_{CEF} = \frac{1}{\hbar} \cdot [(\alpha k_y - \beta k_x), (\beta k_y - \alpha k_x), 0].
\]  

(6)
Due to the different symmetry of the two contributions, the resulting combined effective field (CEF) may show a well-defined preferential direction if $\alpha = \beta$:

$$
\Omega(k)_{CEF} = \frac{\beta}{\hbar} [(k_y - k_x), (k_y - k_x), 0] \equiv \frac{\beta}{\hbar} (k_y - k_x) \cdot [1, 1, 0].
$$

In this case, the CEF points along the in-plane [110] direction for any value of $k$, as Fig. 1 c) shows. For $\alpha = -\beta$ this preferential direction becomes [101]. This means that electron spins that point along the [110] direction experience no torque and can therefore not dephase due to the D’Yakonov-Perel mechanism, which is effectively blocked for this spin orientation, while electron spins pointing along [110] or along the growth direction will experience a torque and start dephasing. This spin dephasing anisotropy was first pointed out by Averkiev and Golub [28], and recently observed experimentally [29, 30].

### 3 Sample structure and preparation

Our sample was grown by molecular beam epitaxy on a [001]-oriented semi-insulating GaAs substrate. The active region is a 20 nm-wide, one-sided modulation-doped GaAs-Al$_{0.3}$Ga$_{0.7}$As single QW. The electron density and mobility at $T = 4.2$ K are $n_e = 2.1 \times 10^{11}$ cm$^{-2}$ and $\mu_e = 1.6 \times 10^6$ cm$^2$/Vs, respectively. These values were determined by transport measurements on an unthinned sample. For measurements in transmission geometry, the sample was glued onto a sapphire substrate with an optical adhesive, and the substrate and buffer layers were removed by selective etching.

### 4 Measurement techniques

#### 4.1 Time-resolved Kerr/Faraday rotation

For both, the time-resolved Faraday rotation (TRFR) and the time-resolved Kerr rotation (TRKR) measurements, two laser beams from a mode-locked Ti:Sapphire laser, which is operated at 80 MHz repetition rate, were used. The laser pulses had a temporal length of about 600 fs each, resulting in a spectral width of about 3-4 meV, which allowed for near-resonant excitation. The laser wavelength was tuned to excite electrons from the valence band to states slightly above the Fermi energy of the host electrons in the conduction band. Both laser beams were focused to a spot of approximately 60 µm diameter on the sample surface. The pump pulses were circularly polarized by an achromatic $\lambda/4$ plate in order to create spin-oriented electrons in the conduction band, with spins aligned perpendicular to the QW plane. The weaker probe pulses were linearly polarized. The polarization rotation of the
transmitted/reflected probe beam was analyzed by an optical bridge detector. In order to separate the time evolution of the spin polarization from the photocarrier dynamics, all measurements were performed using both helicities for the circularly-polarized pump beam [31]. The TRFR measurements were performed in a split-coil magnet cryostat with a $^3$He insert, allowing for sample temperatures between 1.5 K and 4.5 K. The TRKR measurements were performed in a continuous-flow He cold finger cryostat. In this cryostat, non-thinned samples from the same wafer were used. Unless otherwise stated, the experiments were carried out at a nominal sample temperature of $T=4.5$ K.

5 Experimental results

5.1 Variation of spin polarization degree

![Graph](image)

**Fig. 2.** (a) Normalized TRFR traces for different pump beam fluence and therefore different initial spin polarization. (b) Comparison of TRFR traces for low initial spin polarization at different temperatures. The inset shows the data from (a) with a log scale. (c) TRKR traces for varying circular polarization degree of the pump laser beam. The curves have been normalized for easier comparison. Note the log scale. (d) Spin relaxation time as a function of circular polarization degree of the pump beam, extracted from the TRKR traces. ((a,b) reprinted with permission from [32]. Copyright (2007) by the American Physical Society.)

Typically, in time-resolved Faraday/Kerr rotation measurements on semiconductor heterostructures, the excitation density is kept very low to avoid heating the sample. Here, we present experiments in which the excitation density was considerable, resulting in a significant initial spin polarization. Figure 2 (a) shows a series of TRFR measurements performed without external magnetic field. In this measurement, the excitation density, and therefore the initial spin polarization, was increased by increasing the laser pump fluence. The curves typically show a biexponential decay of the spin polarization, except for the lowest curve, where a strongly damped oscillation is
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visible. The oscillatory behavior will be discussed in the following subsection. We associate the fast decay with the spin dephasing of the photoexcited holes, which typically lose their spin orientation within a few picoseconds, and the slower decay with the spin dephasing time of the electrons, $T_2^*$. It is clearly seen how $T_2^*$ is increased by more than an order of magnitude as the initial spin polarization is increased from the low-polarization limit to about 30 percent. This increase is due to the Hartree-Fock (HF) term of the Coulomb interaction (as predicted by Weng and Wu [18]), which acts as an out-of-plane, $k$-dependent effective magnetic field. This effective field lifts the degeneracy of the spin-up and spin-down states, causing a spin-flip to require a change in energy. This significantly reduces the spin-flip rate and increases the spin dephasing time. In microscopic calculations including the HF term of the Coulomb interaction, the measurements shown in Fig. 2 (a) could be reproduced with great accuracy.

In order to clearly demonstrate that the observed effect is due to the increase in spin polarization, and not caused by either a change in electron density or sample heating, we performed control experiments in which the initial spin polarization was varied while the excitation density was kept constant [33]. In order to achieve this, the circular polarization degree of the pump laser pulse was varied from almost 100 percent circular polarization to almost linear polarization, by gradual rotation of the $\lambda/4$ plate in the pump beam. By this means, the fraction of spin-polarized electrons created by the pump beam could be tuned. Figure 2 (c) shows a series of TRKR measurements performed in this way. It is clearly visible how an increase of the circular polarization degree of the pump beam, and therefore an increase of the initial spin polarization, leads to reduced spin dephasing and therefore a shallower slope in the logarithmic plot of the TRKR trace. The TRKR traces have been normalized for easier comparison. In Fig. 2 (d), the spin dephasing times extracted from the measurement series are plotted as a function of the circular polarization degree.

### 5.2 Coherent zero-field oscillation

In Fig. 2 (b), two TRFR traces taken without external magnetic field at low excitation density are shown. The upper (red) curve, showing a strongly damped oscillation, represents the same data as the lowest (red) curve in Fig. 2 (a). The lower curve in 2 (b) was taken using the same excitation density, but at a lower sample temperature of 1.5 K. Here, the damping is significantly reduced. Both traces are representative of the weak scattering regime of the DP mechanism: an ensemble of electrons with $k$ slightly above the Fermi wave vector is generated by the pump pulse. While the orientation of the effective spin orbit field for these electrons depends on the electron $k$ vector, it generally lies in the sample plane, leading to spin precession. The $z$ component of the electron spins oscillates, and the damped oscillation we observe, is the coherent superposition of these oscillations. We note that this
effect was first reported by Brand et al. [10]. Two effects contribute to the damping of this coherent oscillation:

1. The amplitude of the effective spin-orbit field depends on $k$ if both the Rashba and the Dresselhaus terms are present (see for example Fig. 1c)), leading to a (reversible) dephasing of the electron spins due to different precession frequencies.

2. Initially, the electron spins are aligned along the growth direction by the laser pump pulse, therefore the effective spin-orbit field is perpendicular to the electron spin for the whole ensemble. Precession tilts the electron spins into the sample plane, and momentum scattering changes $k$ and $\Omega(k)$, leading to different angles between the electron spins and the effective spin-orbit fields. This causes (irreversible) dephasing due to different spin precession frequencies. Momentum scattering is temperature-dependent, hence the damping of the coherent oscillation becomes more pronounced as the sample temperature is raised.

5.3 Spin dephasing in an external magnetic field

Here, we present experiments performed in small external magnetic fields applied perpendicular to the quantum well plane. The field amplitude was kept below 0.5 Tesla, where effects due to Landau quantization may be neglected. In this regime, we observe a competition between the Hartree-Fock effective out-of-plane field, created by the interaction between spin-aligned electrons, and the external magnetic field. As Fig. 3 (a) shows, the application of an external magnetic field monotonically increases the spin dephasing time for both small and large initial spin polarization. At zero and small external fields, the HF field dominates, leading to a significantly longer spin lifetime for large initial spin polarization. However, as the external field amplitude is increased, the spin lifetimes for large and small initial spin polarization start

![Fig. 3.](image-url)
to merge and become almost identical for magnetic fields of 0.4 Tesla and above. This indicates that the mean value of the HF field for the large initial spin polarization is below 0.4 Tesla.

The spin polarization, which we create optically, defines a preferential direction along the growth axis. The external magnetic field can therefore be applied parallel or antiparallel to this preferential direction. Figure 3 (b) shows that for both external field directions the spin lifetime increases monotonically, and the curve is symmetric in the external field B. This indicates that the external magnetic field cannot be used to compensate the effective Hartree-Fock field, since that is $k$-dependent.

5.4 Probing magneto-anisotropy

The magneto-anisotropy in a [001]-grown GaAs QW leads to different spin lifetimes for electron spins aligned along different in-plane directions. In our experimental setup, however, we create an out-of-plane spin polarization by the circularly-polarized pump pulse, and the detection scheme using the linearly-polarized probe beam at near-normal incidence is only sensitive to the out-of-plane component of the spin polarization. Therefore, in order to probe the in-plane magneto-anisotropy, we use an in-plane magnetic field to force the spins to precess into the sample plane and back out again. By this means, the spin lifetimes we observe, represent an average of the out-of-plane and the different in-plane spin lifetimes within the sample and therefore allow us to infer the in-plane anisotropy. In the experiment, we use two sample pieces from the same wafer, which are mounted in the cryostat with their in-plane [110] axis either parallel or perpendicular to the applied in-plane magnetic field. Figure 4 shows the experimental results: (a) without an applied magnetic field, both sample pieces (black line and red circles) show the same spin lifetime $T_2^* = 113 \pm 1$ ps, as we expect due to symmetry reasons. (b) As an in-plane magnetic field of 1 Tesla is applied, the spin lifetimes become markedly different: if the magnetic field is applied along the in-plane [110] direction, the electron spins are forced to precess into the [110] direction. The spin lifetime average between the [001] and the [110] is $T_{2[110]}^* = 127 \pm 1$ ps. If, however, the magnetic field is applied along [110], forcing the electron spins into the [110] direction, the averaged spin lifetime $T_{2[110]}^* = 204 \pm 2$ ps increases by about 60 percent. This is a clear indication of the in-plane magneto-anisotropy. From the magnetic-field dependence of the averaged spin lifetimes, both the Rashba and the Dresselhaus coefficients can be determined by comparing the experimental data to numerical many-body calculations [34, 35]. Here, we find values of $\beta = 1.38$ meVÅ and $\alpha = 0.9$ meVÅ, yielding a ratio $\frac{\alpha}{\beta} = 0.65$. The symmetry of the CEF for this ratio is shown in Fig. 4 (c): the preferential direction of the CEF along the [110] direction is clearly visible. Using these values, an in-plane spin lifetime anisotropy of 60 to 1 can be inferred from the calculations. Thus, the spin
lifetime for electron spins initially *aligned* along the [110] axis is estimated to be about 8 ns. We note that in order to observe the magneto-anisotropy by spin precession, the initial spin polarization has to be so high that the spin lifetime is long enough to allow for at least one precession cycle.

![Fig. 4](image)

**Fig. 4.** (a) Time-resolved Faraday rotation traces for two orientations of the sample without external magnetic field. (b) TRFR traces at 1 Tesla in-plane magnetic field, for two relative orientations of the sample and the magnetic field. (c) Symmetry of the combined effective field for Rashba-Dresselhaus ratio $\alpha = 0.65\beta$.

### 5.5 Breakdown of magneto-anisotropy

![Fig. 5](image)

**Fig. 5.** Calculated symmetry of the combined effective field due to Rashba and *kubic* Dresselhaus terms. The carrier density and thus the Fermi energy is increased from (a) to (c).

The spin-dephasing anisotropy in [001]-grown QWs is an approximation based on the interference of the Rashba and the *linear* Dresselhaus spin-orbit fields. In this approximation, the vector sum of Rashba and Dresselhaus fields points along the [110] direction for $\alpha = \beta$, regardless of $k$. As the
carrier density in a 2DES is increased, the cubic Dresselhaus term has to be taken into account, and the symmetry of the combined effective field (CEF) changes. Here, we show density-dependent calculations of the symmetry of the CEF as a function of the carrier density. The calculations were performed for a 20 nm wide quantum well, and for the calculations the ratio of the Rashba and Dresselhaus terms was kept constant at $\alpha = \beta$. As Fig. 5 (a) shows, for low density the orientation of the CEF remains almost perfectly aligned along the [110] direction. As soon as the density is increased to values more common for a 2DES, as in 5 (b) and (c), a deviation from this orientation can be observed for a range of $k$ values. In order to quantify the breakdown of the magneto-anisotropy, we calculate the average torque $\bar{\tau}$ acting on an electron spin $s$ pointing along the in-plane [110] or [1¯10] directions:

$$|\bar{\tau}| = \sum_k |s \times B_{eff}(k)|.$$  

Here, the sum is over all directions of $k$ for $|k| = k_F$. From these values, we can determine the (dimensionless) anisotropy $\bar{\tau}_{[110]}/\bar{\tau}_{[1\bar{1}0]}$ of the initial torque acting on electron spins pointing along [110] and [1¯10], and track its density-dependence. Figure 6 shows the results of this calculation for 15-25 nm wide quantum wells with equal Rashba and Dresselhaus terms ($\alpha = \beta$). It can be clearly seen how the torque anisotropy is reduced by an order of magnitude as the density is increased from values common for low-doped 2DES samples ($n = 1 \times 10^{11}$ cm$^{-2}$) to highly-doped 2DES ($n = 8 - 10 \times 10^{11}$ cm$^{-2}$). For comparison, the same calculation was performed for a 20 nm wide QW with ($\alpha = 0.65\beta$), the ratio corresponding to the sample investigated in our experiments. There, the torque anisotropy is significantly lower than for ($\alpha = \beta$) and remains almost constant over a wide range of densities. We note that a direct experimental observation of the magneto-anisotropy breakdown will be difficult as tuning the carrier density by an external gate voltage will also change the Rashba/Dresselhaus ratio.

**Fig. 6.** Anisotropy of the initial torque acting on electron spins pointing along the in-plane [110] and [1¯10] directions as a function of electron density. Calculations for 15 nm (red circles), 20 nm (black squares), and 25 nm (blue triangles) wide QWs with equal Rashba and Dresselhaus terms ($\alpha = \beta$) are shown, as well as the values for a 20 nm wide QW with ($\alpha = 0.65\beta$) (green stars).
6 Summary

In conclusion, we have investigated the spin dephasing in a high-mobility 2DES as a function of the initial spin polarization, showing that due to electron-electron interaction, the spin dephasing is reduced as the initial spin polarization is increased. Additionally, we studied the spin dephasing in external magnetic fields applied perpendicular and parallel to the 2DES. Here, we observed a strong in-plane anisotropy of the spin dephasing due to interference between the Dresselhaus and Rashba spin-orbit fields. This anisotropy is strongest for equal Dresselhaus and Rashba fields, yet highly density-dependent, as the cubic Dresselhaus term changes the symmetry of the combined effective field.

7 Acknowledgements

The authors would like to thank J. Zhou, J.L. Cheng, J.H. Jiang and M.W. Wu for fruitful discussion. Financial support by the DFG via SPP 1285 and SFB 689 is gratefully acknowledged.

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