Precession and Motional Slowing of Spin Evolution in a High Mobility Two-Dimensional Electron Gas

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Optical spin-dynamic measurements in a high-mobility n-doped GaAs/AlGaAs quantum well show oscillatory evolution at 1.8 K consistent with a quasi-collision-free D'yakonov-Perel'-Kachorovskii regime. Above 5 K evolution becomes exponential as expected for collision-dominated spin dynamics. Momentum scattering times extracted from Hall mobility and Monte Carlo simulation of spin polarization agree at 1.8 K but diverge at higher temperatures, indicating the importance of electron-electron scattering and an intrinsic upper limit for the spin-relaxation rate.

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Growing interest in conduction electron spin relaxation and precession in III-V semiconductors has been stimulated by possibilities for spin electronics as well as by unusual characteristics of the dominant relaxation mechanism. In this Letter we describe optical experiments on electron spin evolution in a two-dimensional electron gas (2DEG) in the high-mobility limit at low temperatures which elucidate two neglected features of the dynamics. These considerations may be relevant to applications where both high-mobility and long spin memory are required simultaneously. As first pointed out by D'yakonov, Perel', and Kachorovskii (DPK) [1,2], the dominant mechanism for relaxation of the polarization of a population of initially aligned electron spins is precession of the individual electron spin vectors ($\mathbf{S}$) during the time between momentum scattering events (τ_p^*) according to $\dot{\mathbf{S}} = \mathbf{\Omega} \times \mathbf{S}$, where the effective Larmor precession vector $\mathbf{\Omega}(\mathbf{k})$ describes the conduction band spin splitting [3] which depends on both the magnitude and direction of electron wave vector, $\mathbf{k}$. Previously, attention has focused on the collision-dominated or "motional-narrowing" regime [1,2,4], wherein $\langle|\mathbf{\Omega}|\rangle\tau_p^* \ll 1$ with $\langle|\mathbf{\Omega}|\rangle$ the average precession frequency and where spin precession is interrupted frequently by momentum scattering; spin reorientation is motionally slowed with respect to the average precession period and becomes an exponential relaxation with rate [2,4]

$$\tau_s^{-1} = \langle\Omega^2\rangle\tau_p^*. \tag{1}$$

Two other spin-relaxation mechanisms have been identified [4,5]: (a) spin-orbit induced spin flips during momentum scattering, the Elliott-Yafet mechanism, which is generally much weaker than the DPK mechanism [2,5,6]; (b) electron-hole exchange interaction, the Bir-Aronov-Pikus mechanism, which is not relevant here as we are concerned with minimal hole concentrations in n-doped material.

Consider now the neglected features of the DPK mechanism. First, it is usually assumed [2,4] that the scattering time, τ_p^*, for randomization of spin precession is effectively the same as the transport scattering time which determines the mobility, τ_p. Thus, in the collision-dominated regime, according to Eq. (1) the spin-relaxation rate will increase continuously with the scattering time and so, by assumption, as the mobility is increased. We point out that precession of an electron spin can be as effectively randomized by scattering from another electron via the Coulomb interaction as by scattering from thermal vibrations or defects, and yet the electron-electron scattering can affect the mobility only very weakly via U-processes [7]. Therefore, in general, $\tau_p^* \leq \tau_p$ and, in a high-mobility 2DEG at low temperatures, where the electron mobility and hence τ_p is determined by *extrinsic* defect scattering, the collision-dominated spin-relaxation rate [Eq. (1)] will have *an intrinsic upper limit* set by electron-electron scattering and not directly related to the electron mobility. Only for the special case of electrons at the Fermi energy at $T = 0$ K should $\tau_p^* = \tau_p$, giving spin-relaxation rate limited by extrinsic scattering, since electron-electron scattering is then inhibited by the Pauli exclusion principle [7–9]. Glazov and Ivchenko [10] have recently treated the case of a nondegenerate electron gas theoretically and shown that electron-electron scattering for a given electron concentration is even more effective for randomizing spin precession than the scattering by ionized donors which give rise to the free electrons. The second feature is that for sufficiently weak scattering we may expect breakdown of the collision-dominated regime. In an almost collision-free regime, $\langle|\mathbf{\Omega}|\rangle\tau_p^* \gg 1$, the electron spins will rotate many times between scatterings and the average z component of spin of an electron population $\langle S_z\rangle$, initially spin polarized along z, will therefore oscillate with frequency $\sim\langle|\mathbf{\Omega}|\rangle$.

To access optically this regime and to search for its signature—oscillatory spin evolution—we have made measurements on a degenerate high-mobility 2DEG at the lowest available temperature (1.8 K) and with minimal excitation density to excite and probe spin-polarized electrons into a narrow band of states near the Fermi energy (E_F), as indicated in Fig. 1. Time-resolved experimental data (see below) indeed show heavily damped oscillatory spin evolution at this temperature where we expect $\tau_p^* \approx \tau_p$ and estimate $\langle|\mathbf{\Omega}|\rangle\tau_p \approx 2$. As the temperature is increased, strongly retarded exponential decay is observed as expected for the standard collision-dominated regime, but there is a significant discrepancy between τ_p^* and τ_p, consistent with our proposal of a contribution of electron-electron scattering to the former.

We describe in detail data from a sample [11] in which the 2DEG was confined in a (001)-oriented 10 nm one-side n-modulation-doped GaAs/AlGaAs single quantum well structure grown by molecular-beam epitaxy. The structure was processed into a Hall bar field-effect transistor (FET) with transparent Schottky gate for optical measurements and *in situ* Hall measurements. In this sample we have full control of the experimental parameters; bias was set for maximum electron concentration, N_S, and mobility in the well. The same qualitative features of spin evolution have been observed for three other unprocessed samples with nominally comparable mobilities and electron concentrations. In Fig. 1(b) the photoluminescence (PL) peak (1.5643 eV) corresponds to the band-edge recombination [see Fig. 1(a)], while the coincident peaks of luminescence excitation (PLE) and electroreflectance (ER) at 1.5749 eV indicate absorption at the Fermi edge; the Stokes shift of 10.6 meV gives $E_F = 6.64$ meV and $N_S = 1.86 \times 10^{11}$ cm^{-2}. The Hall mobility [Fig. 1(c)], reaching 264×10^3 cm^2 V^{-1} s^{-1} at 1.8 K equivalent to $\tau_p \approx 10$ ps, indicates that neutral impurity or interface roughness scattering dominate at low temperatures [12]. The Hall measurements also showed N_S to be approximately constant below 100 K.

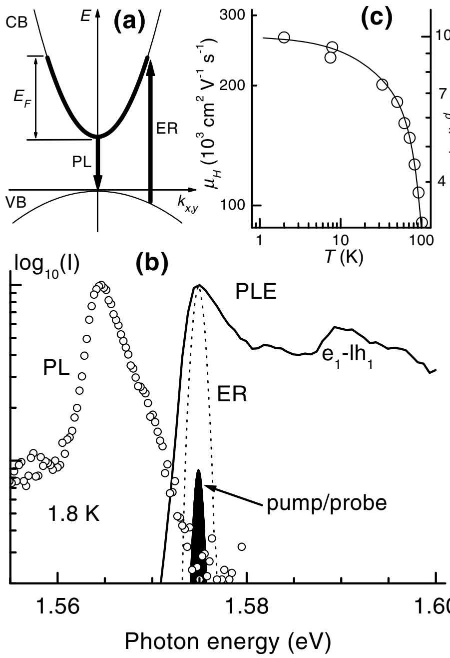

FIG. 1. (a) Schematic band structure showing electron Fermi sea and main optical transitions, (b) 1.8 K photoluminescence (PL), photoluminescence excitation (PLE), and electroreflectance (ER) spectra, and (c) Hall mobility and corresponding momentum scattering time (τ_p) of 10 nm GaAs/AlGaAs n-modulation doped quantum well. Pump/probe indicates energy and spectral width of spin evolution measurement.

The theoretical average precession frequency of electrons at the Fermi level has two contributions in this sample [3,4]; the largest is from the Dresselhaus or bulk inversion asymmetry (BIA) term for the zinc blende structure, whereas the Rashba or structural inversion asymmetry (SIA) term is about 10 times less important. For an (001) oriented well the precession vector lies in the plane of the well and in a perturbation approximation has the form [3]

$$\mathbf{\Omega}(\mathbf{k}) = \frac{2}{\hbar}\{[a_{42}k_x(\langle k_z^2\rangle - k_y^2) - a_{46}E_z^{\mathrm{eff}}k_y]\mathbf{x} + [a_{42}k_y(k_x^2 - \langle k_z^2\rangle) + a_{46}E_z^{\mathrm{eff}}k_x]\mathbf{y}\}, \qquad (2)$$

where $\langle k_z^2\rangle$ is the mean square electron wave vector along the growth direction, k_x and k_y are components of the in-plane electron wave vector, and $\mathbf{x}$ and $\mathbf{y}$ are unit vectors along (100) and (010) axes, respectively. The coefficients a_{42} and a_{46} define the strengths of BIA and SIA terms and have values of 1.6×10^{-29} eV m^3 and 9.0×10^{-39} C m^2, respectively [3]. E_z^{eff} is an effective electric field in the growth direction, which vanishes for a symmetrical structure and depends on band bending and band-edge offsets [3]. The solid curve in Fig. 2(a) shows $|\mathbf{\Omega}(\mathbf{k}_F)|$ for the BIA term alone ($E_z^{\mathrm{eff}} = 0$), and the dotted curve includes an SIA term with $E_z^{\mathrm{eff}} = 15$ kV cm^{-1}, estimated from self-consistent solutions of Poisson and Schrödinger equations for our structure. A third contribution, native interface anisotropy [4], occurs if well and barrier materials have no common atom and is zero here. The calculated average frequencies $\langle|\mathbf{\Omega}(\mathbf{k}_F)|\rangle$ at the Fermi wave vector are 0.238 and 0.241 rad ps^{-1} for $E_z^{\mathrm{eff}} = 0$ and 15 kV cm^{-1}, respectively. Combined with the measured value of $\tau_p = 10$ ps, these values give $\langle|\mathbf{\Omega}|\rangle\tau_p \approx 2.4$ at 1.8 K, indicating that a collision-dominated description is not appropriate for spin evolution at the Fermi energy, and instead we should expect oscillatory behavior. Figure 2(b) shows the calculated collision-free spin evolution; the

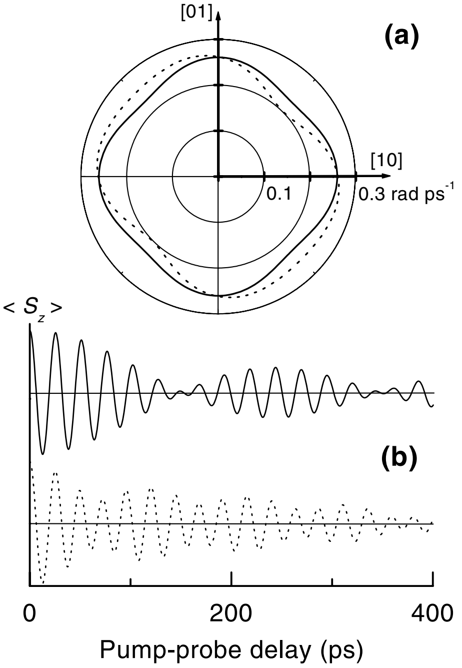

FIG. 2. (a) Polar plot of calculated precession frequency $|\mathbf{\Omega}(\mathbf{k}_F)|$ for electrons at the Fermi wave vector, and (b) corresponding predicted evolution of averaged spin z-component $\langle S_z \rangle$ in the absence of momentum scattering for $E_z^{\mathrm{eff}} = 0$ (solid curves) and 15 kV cm^{-1} (dotted curves).

weak decays and beating are associated here with the anisotropy of $|\mathbf{\Omega}(\mathbf{k}_F)|$. More accurate nonperturbative calculations [4] indicate that Eq. (2) is likely to overestimate $\langle|\mathbf{\Omega}|\rangle$, but the qualitative conclusions will not be affected.

The time-resolved optical response of the 2DEG was investigated by near-normal incidence reflection of wavelength-degenerate 2 ps circularly polarized pump and delayed linearly polarized probe pulses from a mode-locked Ti-sapphire laser. Pump-induced changes of probe reflection ΔR and of probe polarization rotation $\Delta\Theta$ were recorded simultaneously as functions of probe pulse delay using balanced photodiode detectors and lock-in techniques. For ΔR, ~10% beam splitters allowed comparison of intensities of the incident and reflected probe, and, for $\Delta\Theta$, a polarizing beam splitter gave comparison of reflected polarization components at ±45° to the incident probe polarization. The pump beam intensity was 0.1 mW focused to a 60 μm diameter spot giving an estimated photoexcited spin-polarized electron density 2×10^9 cm^{-2}, less than 2% of the unpolarized electron concentration in the 2DEG; the probe power density was 25% of the pump. Figures 3(a) and 3(b) show $\Delta\Theta$ taken at several temperatures with the laser tuned to the ER peak [see Fig. 1(b)], and so injecting electrons at the Fermi level. At the lowest temperature (1.8 K) the spin polarization evolves as a heavily damped oscillation of frequency 0.19 ± 0.02 rad ps^{-1} (see below), which corresponds well with the calculated average precession frequency (~ 0.24 rad ps^{-1}). At 10 K the evolution appears exponential, and as the temperature is increased the decay slows dramatically, as expected for the transition to a collision-dominated regime.

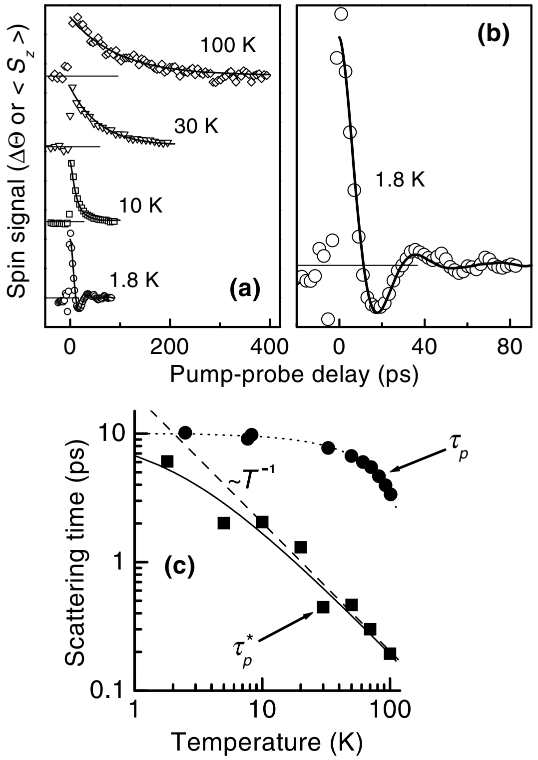

FIG. 3. (a) Polarization rotation signal, $\Delta\Theta$, proportional to $\langle S_z \rangle$, at various temperatures showing damped oscillatory behavior at 1.8 K [expanded view in (b)] and monotonic decay at higher temperatures: points are experimental; curves are Monte Carlo simulation (see text). Excitation energy as indicated in Fig. 1(b) and density ~2×10^9 cm^{-2}. (c) Momentum scattering times from mobility (τ_p) and from Monte Carlo simulation of spin evolution (τ_p^*). Solid curve is derived from the sum of scattering rates represented by an empirical T^{-1} fit to τ_p^* at high temperature (dashed line) and τ_p (dotted curve).

On the time scale of this experiment, phase-space filling by the photoexcited electrons should dominate the pump-induced changes [9]. Circularly polarized (σ^+) pump photons will create an excess population of $|S_z = -1/2\rangle$ electrons at the Fermi energy with isotropic distribution of in-plane wave vectors and an equal population of $|J_z = -3/2\rangle$ holes in the valence band. The phase-space-filling effect of the holes may be neglected since they will rapidly relax into states already blocked by the Fermi sea of electrons. $\Delta\Theta$ will therefore be proportional to the pump-induced imbalance of electron spin polarization and ΔR to the density of photoexcited electrons. We observed that ΔR was constant at each temperature following the pump pulse, indicating negligible recombination over the time scale of the spin evolution. Thus $\Delta\Theta$ follows $\langle S_z \rangle$ and is dominated by the evolution of the photoexcited electrons at the Fermi level.

Figure 2(b) indicates that the anisotropy of $|\mathbf{\Omega}(\mathbf{k}_F)|$ at the Fermi energy cannot alone explain the rapid damping of the experimental oscillations at 1.8 K, and therefore momentum scattering must play a dominant role. To simulate this, we have made a Monte Carlo simulation of $\langle S_z \rangle$ neglecting anisotropy of $\mathbf{\Omega}$ and assuming elastic momentum scattering with an adjustable rate $(\tau_p^*)^{-1}$. An initial arbitrary population of 10^5 $|S_z = -1/2\rangle$ electrons is injected at the Fermi energy with an isotropic distribution of in-plane wave vectors. These all precess at the same frequency $\langle|\mathbf{\Omega}|\rangle$ which may be adjusted in the simulation and undergo random changes of wave vector on scattering. The axis of precession of each electron is as specified by the vector $\mathbf{\Omega}(\mathbf{k})$ and may thus be changed by scattering events as the wave vector is changed. The best fit to the 1.8 K data is obtained for $\langle|\mathbf{\Omega}|\rangle = 0.19 \pm 0.02$ rad ps^{-1} (as stated above) and with $\tau_p^* = 6 \pm 1$ ps quite close to the scattering time $\tau_p = 10 \pm 1$ ps from the Hall mobility. Calculations suggest that $\langle|\mathbf{\Omega}|\rangle$ will be only weakly temperature dependent up to 100 K in our sample; initially it decreases with the chemical potential of the Fermi sea and then increases as the degeneracy temperature, $E_F/k_B \approx 70$ K, is approached. Therefore for higher temperatures we assume a constant value of $\langle|\mathbf{\Omega}|\rangle = 0.19$ rad ps^{-1} and vary τ_p^* to fit the data at each temperature. The model gives good agreement with the observed spin evolution as shown in Fig. 3(a) and 3(b) (solid curves), clearly simulating the transition to the collision-dominated regime accompanied by progressive slowing of the decay. The extracted values of τ_p^* are plotted in Fig. 3(c) (squares) along with the values of transport relaxation time τ_p from Hall measurements (circles); τ_p remains relatively constant, whereas the variation of τ_p^* indicates an additional scattering time which falls approximately as T^{-1}. Although our Monte Carlo simulation is a simplification, the difference of the times of more than an order of magnitude for most of the temperature range and their very different temperature dependences confirm that there is a fundamental distinction between the nature of scattering required to randomize spin precession and electron drift. The convergence of the two times at $T = 0$ K is consistent with expectation of inhibition of electron-electron scattering which is removed at higher temperatures due to broadening of the Fermi edge. The experimental results of Kim *et al.* [9] and also Landau Fermi liquid theory [8] suggest that at $T = 35$ K ($k_BT/E_F \approx 0.5$) the electron-electron scattering time will be $\sim$1 ps, which is sufficiently close to the measured value of τ_p^* to give further support to our interpretation. Full confirmation requires a theoretical treatment of spin relaxation in a degenerate 2DEG, at present lacking [10].

In conclusion, we have observed breakdown of the normally assumed collision-dominated regime of spin relaxation in a high-mobility 2DEG at 1.8 K leading to oscillatory rather than exponential spin evolution with frequency close to the calculated value. At higher temperatures the spin evolution becomes exponential and slows dramatically as expected for a transition to collision-dominated relaxation. Values of momentum scattering time extracted from the spin decays agree with the transport scattering time at 1.8 K but are about an order of magnitude shorter at higher temperatures. This additional scattering associated with randomization of spin precession appears consistent with electron-electron Coulomb scattering which has not previously been identified as a contributor to spin relaxation and implies the existence of an intrinsic limit to the spin-relaxation rate in a 2DEG.

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