Temperature and doping dependence of spin relaxation in *n*-InAs

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(Received 9 December 2004; revised manuscript received 21 March 2005; published 26 August 2005)

We have used time-resolved spectroscopy to measure the relaxation of spin polarizations in the narrow gap semiconductor material *n*-InAs as a function of temperature, doping, and pump wavelength. The results are consistent with the D'Yakonov-Perel mechanism for temperatures between 77 and 300 K. However, the data suggest that electron-electron scattering should be taken into account in determining the dependence of the spin lifetime on the carrier concentration in the range $5.2\times10^{16}-8.8\times10^{17}$ cm^{-3}. For a sample with doping of 1.22×10^{17} cm^{-3} the spin lifetime was 24 ps at room temperature. By applying a magnetic field in the sample plane we also observed coherent precession of the spins in the time domain, with a *g* factor $g^*=-13$, also at room temperature.

DOI: 10.1103/PhysRevB.72.085346 PACS number(s): 78.47.+p, 72.25.Rb, 72.25.Fe

I. INTRODUCTION AND THEORY

Utilization of the electron spin in semiconductors, or spintronics, has become a focus of interest in recent years (for a recent review see Ref. 1). Proposed spin transistor architectures[2] require that spins can be transported through the semiconductor with long lifetime and coherently manipulated in transit. The prime candidate for electrical spin manipulation expoits the Rashba effect in low-dimensional structures,[3–6] and there has been a great number of studies of the spin lifetime and manipulation in GaAs-based systems using optical orientation techniques with pulsed lasers.[6–11] On the other hand, relatively little attention has been paid to narrow gap semiconductors (NGSs), even though they may be important in future spintronics applications because of their high Rashba effect,[3–5] *g* factor (see below) and mobility,[12,13] etc. Indeed observation of electrical injection with subsequent long spin mean free path has been reported in NGS quantum wells.[14] Originally spin lifetimes in NGSs were studied only with spin-resonance techniques at low temperature[15,16] because of the need for long wavelength light sources.[17–20]

We report measurement of the spin relaxation between 300 and 77 K in bulk *n*-InAs (in agreement with earlier work[17,18] at 300 K) and over the range 5.2×10^{16} to 8.8×10^{18} cm^{-3}. Our results are consistent with the so-called D'yakonov-Perel (DP) (Refs. 21 and 22) mechanism which is important for spintronic devices because dominance of the DP mechanism, and a strong Rashba effect are necessary for gate modulation of spin populations.[6] The other main spin relaxation process due to Elliott and Yafet[23] (EY) predicts lifetimes much longer than observed. We show further that including the effect of electron-electron scattering greatly improves the agreement of the DP prediction with experiment. (The Bir-Aronov-Pikus[24,25] mechanism, by contrast with the other two processes, only exists in the presence of holes. It is thought to be particularly important in *p*-type wide gap materials and is based on the electron-hole exchange interaction,[10,22] and we ignore it here.)

The EY mechanism results from the fact that in real crystals Bloch states are not spin eigenstates because of the strong spin-orbit coupling induced by the lattice ions, which results in the valence band states having mixed spin character. In NGSs the conduction electron states, in turn, are strongly mixed with the valence states through the $\mathbf{k}\cdot\mathbf{p}$ interaction across a narrow energy gap. In this case spin-independent interactions with impurities, boundaries, phonons, etc., can connect spin up and down electrons, leading to spin flip transitions whose rate $1/\tau_s$ is proportional to $1/\tau_p$ where τ_p is the orbital momentum (mobility) scattering time. The EY spin relaxation rate for degenerate statistics is[22,23]

$$\frac{1}{\tau_s}\approx A\alpha^2\left(\frac{E_F}{E_G}\right)^2\frac{1}{\tau_p}, \tag{1}$$

where $\alpha=\gamma(1-\gamma/2)/(1-\gamma/3)$, $\gamma=\Delta/(E_G+\Delta)$, and Δ is the spin-orbit splitting of the valence band. E_G and E_F are the fundamental energy gap and Fermi energy, respectively. In the other limit of nondegenerate statistics the EY expression becomes

$$\frac{1}{\tau_s}\approx A\alpha^2\left(\frac{kT}{E_G}\right)^2\frac{1}{\tau_p}. \tag{2}$$

A is a dimensionless constant that varies depending on the orbital scattering interaction process between 2 (for lattice scattering) and 6 (for ionized impurity scattering).

The DP mechanism results from the lack of inversion symmetry so that spin-orbit interaction lifts the spin degeneracy even in the absence of a magnetic field. Because of the mixing mentioned above, injection of an electron with defi-

nite spin by circularly polarized light (or from a magnetic material) is not in a stationary state but a linear combination of the eigenstates. This superposition precesses coherently at a rate given by the spin splitting, which depends on **k**, and with a precession vector **Ω** that also depends on **k**. In an ensemble of electrons all having different **k** and hence precessing about different directions and at different rates, the macroscopic polarization dephases. At elevated temperature orbital momentum scattering causes **k** and hence also **Ω** for each electron to perform a random walk, slowing the DP unphasing, so that its rate $1/\tau_s$ is proportional to τ_p.

The DP relaxation rate for lattice scattering and nondegenerate statistics is given by[21,22]

$$\frac{1}{\tau_s} \approx Q_{\mathrm{nd}} \beta^2 \frac{(kT)^3}{\hbar^2 E_G} \tau_p \tag{3}$$

and for the case of degenerate statistics the DP expression becomes

$$\frac{1}{\tau_s} \approx Q_d \beta^2 \frac{{E_F}^3}{\hbar^2 E_G} \tau_p \tag{4}$$

where $\beta = (4\gamma/\sqrt{3-\gamma})(m_c/m_0), m_c/m_0$ is the electron effective mass in units of the free mass, and $Q_{n\mathrm{d},d}$ is a dimensionless constant that varies between about 0.1 and 3 depending on the dominant orbital scattering process and the electron statistics.

In NGSs both the EY and the DP mechanisms may be important, as shown by reports of spin relaxation in InSb at helium temperature[15,16] and InAs at room temperature[17,18]—the former being interpreted in terms of EY and the latter of DP. Typically spin lifetimes in the range 1–10 ns were obtained from spin resonance in degenerate *n*-type InSb at helium temperatures, whereas a spin lifetime of 20 ps was reported for lightly *n*-type InAs at 300 K. For larger gap materials the DP mechanism has been shown to dominate at 300 K,[1,6–10,22] but the subject remains somewhat controversial since for the smaller gap InGaAs quantum wells the EY process has been claimed to dominate.[26]

II. EXPERIMENT

The samples used here were grown by MBE, Si doped, on semi-insulating GaAs.[13] The low concentration sample IC313 was 3-μm thick (strain relaxed) and had a room-temperature electron concentration and mobility of $n = 5.2 \times 10^{16}$ cm^{-3} and $\mu = 16400$ cm^2 V^{-1} s^{-1}, respectively. IC311 (4-μm thick) had $n = 1.22 \times 10^{17}$ cm^{-3}, $\mu = 17\,200$ cm^2 V^{-1} s^{-1}, and IC301 had $n = 8.8 \times 10^{17}$ cm^{-3}, $\mu = 12\,000$ cm^2 V^{-1} s^{-1}. The temperature dependence of the concentration and hall mobility for IC313 and IC311 are shown in Fig. 1. As is usual, lattice scattering becomes a significant contribution to the mobility at high temperatures.

We have performed circularly polarized pump-probe experiments as described elsewhere.[9,17,18] The light source was a difference frequency generator, which mixes the signal and idler beams of an optical parametric amplifier, itself pumped by an amplified Ti:sapphire oscillator. The pulses had a duration of order 100 fs. The time resolution of the experiments

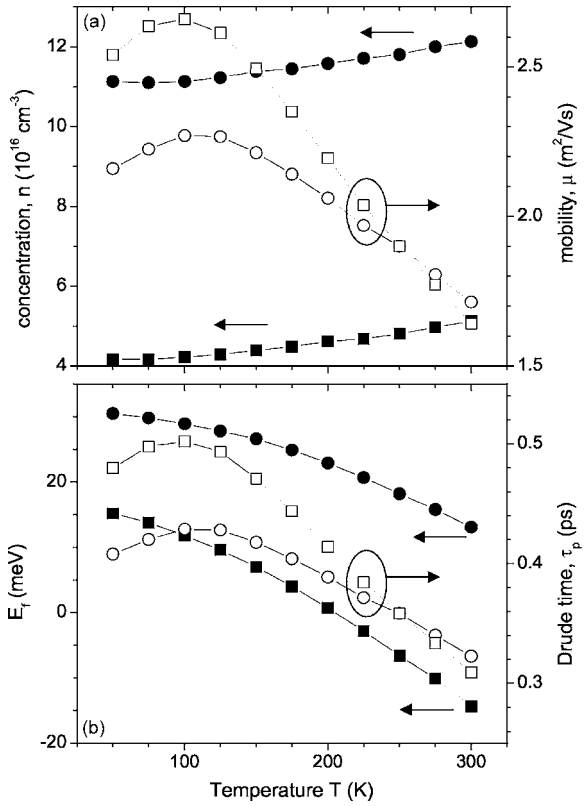

FIG. 1. (a) The carrier concentration *n* (filled symbols) and mobility μ (open symbols) as a function of temperature measured experimentally with the Hall effect and (b) the inferred chemical potential E_F (filled symbols) and orbital scattering time τ_p (open symbols). Data for IC313 is shown with squares and for IC311 with circles.

as measured from the leading edge of the pump-probe data was about 200 fs. In such measurements it is common to use quarter-wave plates to circularly polarize the pump and probe beams. The transient absorption bleaching of the probe is then measured for pump and probe beams having the same circular polarization (SCP) and then the opposite (OCP) by rotating one of the quarter-wave plates by 90°. The sum of these signals is a measure of the total population recombination and reproduces the linearly polarized result, whereas the difference is a measure of the population spin-polarization. The technique therefore measures which ever is the shorter of the spin flip scattering by EY (T_1) or spin dephasing by DP (T_2). The optical polarization defined by

$$P_{\mathrm{opt}} = \frac{\Delta T_{\mathrm{SCP}} - \Delta T_{\mathrm{OCP}}}{\Delta T_{\mathrm{SCP}} + \Delta T_{\mathrm{OCP}}} = \frac{\Delta T_{\mathrm{SCP}} - \Delta T_{\mathrm{OCP}}}{\Delta T_{\mathrm{LP}}} = P_0 \exp(-t/\tau_s), \tag{5}$$

where ΔT is the probe transmission change, which decays exponentially with a decay constant equal to the spin lifetime. P_0 is a constant that depends on the matrix elements and is 0.25 at best for bulk III-V semiconductors.[17]

We used a ZnSe photoelastic modulator (PEM) to modulate the polarization of the pump. When a single mirror was placed between the detector and the PEM a modulation in

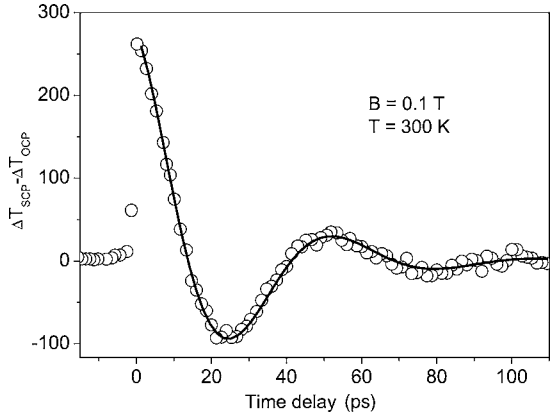

FIG. 2. Optical polarization as a function of time for sample IC311 at 300 K with an externally applied magnetic field of 0.1 T. The spin population precesses while decaying, and the solid line is a fit of a sinusoid with an exponentially decaying envelope (oscillation period 53.1±0.4 ps and decay constant 24.4±0.4 ps).

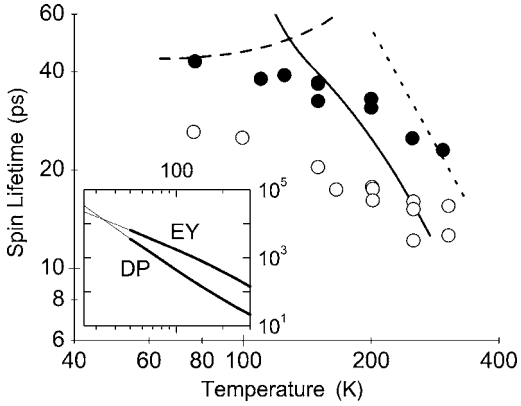

FIG. 3. The temperature dependence of the spin lifetime in n-InAs determined experimentally: sample IC313, $n=5.2\times10^{16}$ cm^{-3}, open circles; sample IC311, $n=1.22\times10^{17}$ cm^{-3}, filled circles. Also shown are the theoretical curves according to Ref. 22, $n=1\times10^{17}$ cm^{-3}, solid curve; Eq. (3) for nondegenerate DP, dotted curve; Eq. (4) for degenerate DP, dashed curve. For the latter two the carrier concentration and orbital momentum scattering time used was from the experimentally determined Hall measurement for IC311. Inset: comparison of the spin lifetime versus temperature for EY and DP mechanisms in the nondegenerate model [Eqs. (1) and (4), respectively]. The thick lines use measured mobility time and the thin lines are extrapolated, showing a crossover from DP to EY as the temperature is lowered.

intensity observed was of order 0.1%, which is due to imperfect circular polarization and unequal reflection coefficients of the mirror for the in-plane and out-of-plane components. For this reason the pump was placed at normal incidence to the sample with no optical components between it and the PEM and the sample except for a cryostat window, also at normal incidence. Under these circumstances, when a detector was placed at the sample position no modulation of intensity could be seen. The probe was circularly polarized with a variable quarter wave plate which was not altered during the experiment. The probe polarization was calibrated before the experiment with a polarizer immediately in front of the sample position, so that polarization changes on subsequent reflections etc. downstream from the probe waveplate were compensated.

We applied a small external magnetic field in the sample plane (the Voigt configuration). The spins injected by the normal incidence pump beam are polarized normal to the sample and undergo Larmor precession about the magnetic field axis. The measured polarization oscillates at angular frequency $g^*\mu_B B/\hbar$ while decaying with the spin lifetime. Here g^* is the Lande g factor, μ_B is the Bohr magneton, and B is the magnetic field. The results for $B=0.1$ T at $T=300$ K for sample IC311, shown in Fig. 2, are very well fitted by a simple exponentially decaying sinusoid with zero background. The magnitude of the g factor from the fit is $|g^*|=13.4\pm0.1$ (the measurement does not reveal the sign, which is known to be negative for InAs), and the lifetime is $\tau_s=24.4\pm0.4$ ps. Estimates of the error from the scatter in repeat measurements (below) were rather higher at 10%. The magnetic field was removed for all further experiments described here, and the field and temperature dependence of g^* will be described elsewhere.

The temperature dependences of the spin relaxation time for samples IC313 and IC311 are shown in Fig. 3. The data for each sample include two separate temperature runs, and the scatter in the data is an indication of the uncertainty. The wavelength was tuned at each temperature to the InAs band edge. We note that the laser pulses are very short and consequently have a large spectral width of about 4%, or about 0.015 eV, and the results are rather insensitive to fine tuning of the laser.

We have also measured the spin relaxation time at room temperature of sample IC301. The comparison of the three different samples is shown in Fig. 4 for temperatures 200 and 300 K.

III. DISCUSSION AND CONCLUSION

Figure 3 also shows theoretical predictions for sample IC311 from Ref. 22 and Eqs. (3) and (4). In Ref. 22 mobility values are used that do not correspond precisely with our measured mobility [Fig. 1(a)], the experimentally determined temperature dependence of which we used for calculation of τ_p [Fig. 1(b)]. The value of τ_s calculated from the degenerate model begins to rise rapidly with increasing temperature as the crossover with the nondegenerate regime is approached due to the fact that E_F starts to drop. As mentioned above the predicted constants of proportionality $Q_{\rm nd}$ and Q_d are between 0.1 and 3 depending on the electron statistics and the orbital scattering process, and we have therefore treated them as free parameters. We chose a value of $Q_{\rm nd}=1.5$ for the nondegenerate model curve shown on Fig. 3, to give a good match with the more sophisticated calculations of Ref. 27 (the data of which are for a value of carrier concentration well below degeneracy and much lower than for the samples considered here). Reference 22 gives predictions also for high concentrations (the data shown here are for 1×10^{17} cm^{-3}). However, their model ignores degeneracy effects and as can be seen from Fig. 3 their prediction runs

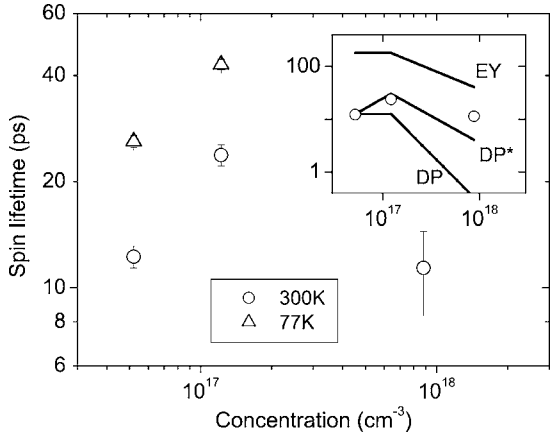

FIG. 4. Carrier concentration dependence of the spin lifetime in *n*-InAs for three samples at 300 and 77 K. Inset: DP and EY model predictions for carrier concentration dependence using the measured mobility time τ_p in Eqs. (1)–(4), taking the degenerate and nondegenerate models above and below 1.22×10^{17} cm^{-3}, respectively, and with the degenerate models scaled to meet the nondegenerate models (unscaled) at the crossover. DP* indicates the DP model with measured mobility time τ_p replaced by the electron-electron scattering time $\tau_p{}^*$ (i.e., $\tau_p{}^*\propto n^{-1}$ with the constant of proportionality chosen to give $\tau_s=24$ ps at $n=1.22\times10^{17}$ cm^{-3}).

parallel to the nondegenerate approximation of Eq. (3) (and may be matched approximately with a prefactor $Q_{\rm nd}=3.5$). For the degenerate model curve shown on Fig. 3, $Q_d=0.3$.

For sample IC311, at $n=1.22\times10^{17}$ cm^{-3}, the Fermi energy is approximately 30 meV above the band edge, but drops with rising temperature and is a few kT or less over the whole range of temperatures used. For sample IC313 the concentration is lower, at $n=5.2\times10^{16}$ cm^{-3}, but even here the chemical potential is always within kT of the band edge for the temperatures used. It is therefore to be expected that the transition from the degenerate to the nondegenerate models is within this range, and it may be that this is the reason for the experimentally determined temperature dependence of the spin lifetime being intermediate between the two regimes for both samples.

To identify the spin relaxation mechanism, we note that the EY model in both limits (degenerate and nondegenerate) gives a spin relaxation time substantially larger than the measured spin lifetime at all temperatures used, i.e., above 80 K. For example, for sample IC311 the lifetime predicted from the EY model [Eq. (1)] is 180 ps at 300 K; see the insets of Figs. 3 and 4. Hence the magnitude and the functional form of the temperature dependence of the EY mechanism suggest that it is not playing a dominant role here.

Now we turn to Fig. 4 which shows that the lifetime has a relatively weak dependence on carrier concentration. Both the remarkable result that we have an initial rise and also the small subsequent drop in lifetime with concentration are not possible to understand using the measured mobility time in the simple models of Eqs. (1)–(4). As shown in the inset of Fig. 4 the simple DP model predicts a nearly constant spin lifetime for nondegenerate concentrations because $\tau_p(300\ {\rm K})$ is nearly constant. This is followed by a predicted very strong decrease at degenerate concentrations due to the $E_F{}^3(\propto n^2)$ factor.

Electron-electron scattering modifies this picture strongly. This process does not contribute to the mobility because it conserves total momentum, but contributes to the DP process by causing $\mathbf{k}$ and hence $\mathbf{\Omega}$ to perform a random walk. Hence the mobility time τ_p in Eqs. (1)–(4) should really be replaced by $\tau=(\tau_p{}^{-1}+\tau_p{}^{*-1})^{-1}$, where $\tau_p{}^*$ is the electron-electron scattering time.[11,28,29] Although the case for the dominance of electron-electron scattering may be more obvious for remotely doped 2D gases at low temperatures[11] we also find justification for invoking this process in our case. The calculated lattice contribution to the mobility at 300 K is via polar optical phonons with a mobility of 3.6 m^2 V^{-1} s^{-1}, whereas the Brooks-Herring mobility for ionized impurity scattering at 5.2×10^{16} cm^{-3} is 3.8 m^2 V^{-1} s^{-1} (decreasing with concentration). Very similar values are obtained by an empirical fit of the two contributions to the experimental mobility (Fig. 1). The lattice and impurity contributions to τ_p and also to τ are therefore comparable at 300 K in our samples. Detailed calculations[29] have shown that the contribution to DP spin relaxation from electron-electron scattering is 2.5 times greater than that from ionized-impurity scattering in bulk materials. This implies that electron-electron scattering may indeed play a more significant role for DP than either lattice or ionized impurity scattering, even at room temperature, and especially for the intermediate and highly doped samples.

The mean free path between electron-electron scattering events, and hence also $\tau_p{}^*$, is proportional to the volume per electron $1/n$.[29] If we assume electron-electron scattering dominates ($\tau\sim\tau_p{}^*$) then the nondegenerate model [Eq. (3)] now becomes $\tau_s\propto n$, so going from IC313 to IC311 the carrier concentration increase by a factor of 2 causes a rise of a factor of 2 in the spin lifetime, in agreement with the experimental result (see Fig. 4). This initial increase of τ_s with concentration is also consistent with (though not as pronounced as) our earlier observations.[18] Using the degenerate model [Eq. (4)] and electron-electron scattering the net concentration dependence is now $\tau_s\propto n^{-1}$ (since $E_F{}^3\propto n^2$) so the expected change in spin lifetime from IC311 to IC301 is now much closer to the observation (also Fig. 4). The remaining discrepancy is likely to be due to the absence of full degeneracy for IC311.

In summary, we have measured the dependence of the spin relaxation time on temperature between 77 and 300 K, and for doping densities between 5.2×10^{16} and 8.8×10^{17} cm^{-3} for *n*-InAs. The maximum lifetime at room temperature was 24 ps for the intermediate doping, and all the lifetimes were within the range 5 to 50 ps. Within the approximations of Eqs. (1)–(4), the magnitude and temperature dependence of the results are in accord with the DP spin relaxation mechanism particularly if electron-electron scattering is taken into account.

ACKNOWLEDGMENTS

We are grateful for partial support from the EU FENIKS project (EC: Grant No. G5RD-CT-2001-00535), and from the UK EPSRC.

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