Suppression of Dyakonov-Perel Spin Relaxation in High-Mobility *n*-GaAs

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We report a large and unexpected suppression of the free electron spin-relaxation in lightly doped *n*-GaAs bulk crystals. The spin-relaxation rate shows a weak mobility dependence and saturates at a level 30 times less than that predicted by the Dyakonov-Perel theory. The dynamics of the spin-orbit field differs substantially from the usual scheme: although all the experimental data can be self-consistently interpreted as a precessional spin-relaxation induced by a random spin-orbit field, the correlation time of this random field, surprisingly, is much shorter than, and is independent of, the momentum relaxation time determined from transport measurements.

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Electrons in *n*-doped semiconductors demonstrate long spin-relaxation times (τ_s) at liquid-helium temperatures [1–4], because spin-orbit channels of spin relaxation (Dyakonov-Perel and Elliot-Yaffet mechanisms [5]) are frozen out. When temperature increases, the electrons bound to shallow donors dissociate with the conduction band, where τ_s is much shorter because of the spin-orbit interaction. This fact imposes serious limitations on high-temperature spintronics [6]. For example, spin-polarized charge carriers, optically or electrically injected into the semiconductor, considerably depolarize at elevated temperatures [5,7]. If the nonequilibrium spin of electrons is to be used for applications, the nature of spin relaxation at relatively high temperatures should first be understood. At $T > 50$ K, the Dyakonov-Perel (DP) [8] mechanism is believed to dominate spin relaxation in *p*-GaAs (Chap. 3, Ref. [5]) and *n*-GaAs [2,9]. This mechanism can be interpreted in terms of an effective magnetic field of spin-orbit interaction, which acts upon the electron spin in semiconductor crystals without inversion symmetry. The value and the direction of this "spin-orbit" field are determined by the value of the electron **k** vector and its direction with respect to the crystal axes. The standard DP approach [8] assumes that the frequent collisions with impurities or phonons, which change the electron wave vector, make the spin-orbit field fluctuate, resulting in a dynamical suppression of spin relaxation. As a result, the spin-relaxation rate is described by the motional-narrowing formula: $\tau_s^{-1} = \langle \Omega^2 \rangle \tau^*$, where τ^* is a correlation time proportional to the momentum relaxation time τ_p. Faster scattering leads to slower spin relaxation [8]. Therefore, the spin-relaxation rate increases with the electron mobility, and the DP mechanism should dominate in "clean" samples with low doping.

In this work we show that, against expectations, the standard DP mechanism is suppressed in clean, high-mobility GaAs samples. Measurements on a set of samples with the electron mobility μ_n ranging from 5000 to 200 000 $\mathrm{cm}^2/\mathrm{V\,s}$ show that the spin-relaxation rate first grows with mobility, but then, at $\mu_n > 20\,000\ \mathrm{cm}^2/\mathrm{V\,s}$, saturates at the level of $(1.5 \pm 0.5)\ \mathrm{ns}^{-1}$ at $T = 77$ K, where electrons obey nondegenerate (Boltzmann) statistics. This finding is in sharp contradiction to the DP theory, which predicts further increase of the relaxation rate. The fact that in high-mobility samples relaxation is 30 times *slower* than the DP prediction might be interpreted as a suppression of the DP mechanism. The suppression persists up to the room temperature. Further experiments in high-mobility pure samples in a longitudinal magnetic field show that the real dynamical picture differs from the usual DP scheme: although all the experimental data can be self-consistently interpreted as a precessional spin relaxation induced by a random spin-orbit (Dresselhaus) field, the correlation time of this random field, surprisingly, is much shorter than the momentum relaxation time determined from transport measurements. It weakly depends on mobility and is approximately equal to 300 fs for most pure samples. It means that electron spin relaxation in high-mobility *n*-type semiconductors decouples from the drift transport of charge.

We have measured a large number of samples grown with different technologies. Both mobility and concentration were measured using the Hall effect in the dark. In one set of samples, grown with molecular beam epitaxy (MBE), 2–7.5 μm thick GaAs layers were sandwiched between AlGaAs barriers. Concentrations of itinerant electrons were from 10^{14} to $6 \times 10^{16}\ \mathrm{cm}^{-3}$ (mobilities μ_n from 140 000 to 5000 $\mathrm{cm}^2/\mathrm{V\,s}$, respectively). The second group of samples, grown by liquid-phase epitaxy, had 20–30 μm thick GaAs layers with electron concentrations from 4.5×10^{14} to $3 \times 10^{16}\ \mathrm{cm}^{-3}$ and mobilities μ_n from 80 000 to 11 000 $\mathrm{cm}^2/\mathrm{V\,s}$. In the third group, grown by gas-phase epitaxy (GPE), GaAs layers were

30–40 μm thick with electron concentrations from 2×10^{13} to 3×10^{15} cm^{-3} and mobilities μ_n from 200 000 to 20 000 cm^2/V s. All the samples were grown in the [001] direction.

The optical orientation method, as applied to measuring spin-relaxation time in n-GaAs, is based on the following [5]: Circularly polarized light creates partially spin-polarized electrons. The simultaneously created holes rapidly lose their spin orientation. Their recombination with electrons of both spin directions accumulates the nonequilibrium mean spin of the electron ensemble. The experimentally measured circular polarization, ρ_c, is proportional to S_z, the projection of the electrons' average spin onto the excitation light beam direction. An external magnetic field, perpendicular to the pump and photoluminescence (PL) direction (Voigt geometry), makes the spin rotate with the Larmor precession frequency $\omega = \mu_B g_e B/\hbar$ (where g_e is the electron g factor, and μ_B is the Bohr magneton). As a result of the precession, S_z decreases with increasing magnetic field and is observed as PL depolarization (Hanle effect) with Lorentzian line shape and a half-width, $B_{1/2} = \hbar/\mu_B g_e T_s$. The inverse spin lifetime is $T_s^{-1} = \tau_s^{-1} + \tau_J^{-1}$. Specifically in n-type semiconductors, the lifetime τ_J has the meaning of the characteristic time during which the spin polarization of equilibrium electrons is accumulated by replacement with photoexcited electrons via the creation and recombination of electron-hole pairs. It is longer than the minority carrier lifetime and depends on their equilibrium concentration $n = N_d - N_a$ (where N_d and N_a are concentrations of donors and acceptors, respectively) and on the pump density, G, as $\tau_J = n/G$ (Chap. 2, Ref. [5]). Therefore, in the limit of low pump density, T_s is equal to the electron spin-relaxation time τ_s, whereas the degree $\rho_c \sim \tau_s/\tau_J$.

The spectra of the PL intensity (solid curve) and the polarization (circles) taken under quasiresonant excitation ($h\nu_{\text{exc}} = 1.55$ eV) at 77 K in zero magnetic field are shown in Fig. 1(a) for a 32 μm, GPE-grown GaAs layer ($N_d - N_a = 5 \times 10^{13}$ cm^{-3}, $\mu_n = 140\,000$ cm^2/V s). The polarization changes over the PL spectrum because of electron spin diffusion [1,5,10]. Nevertheless, the half-width of the Hanle curves vary only by 30%. The effect of spin diffusion is negligible in the low-energy ($h\nu_{\text{det}} = 1.505$ eV) tail [1,10] where the measurement was performed. An example of the Hanle effect at $W = 1$ W/cm^2 is shown in Fig. 1(b). The pump dependence of $B_{1/2}$ yields $1/\tau_s = (1.1 \pm 0.1)$ ns^{-1} [11].

The dependence (solid circles) of the spin-relaxation rate $1/\tau_s$ on the electron mobility is the main result at $T = 77$ K (Fig. 2). Within the range of mobility from $\mu_n = 5000$ to 20 000 cm^2/V s the rate $1/\tau_s$ increases with mobility, but then, at high mobility, it saturates at the level of (1.5 ± 0.5) ns^{-1}. This result suggests also a weak dependence of the rate $1/\tau_s$ on concentration $n = N_d - N_a$ because the variation of mobility at a fixed temperature is the result of variation of the doping level. It agrees well with experiment (inset of Fig. 2).

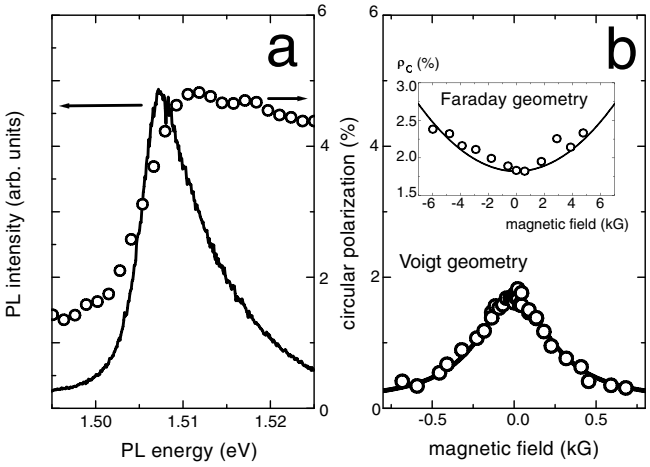

FIG. 1. (a) Spectra of PL intensity (solid line) and polarization (circles) at zero magnetic field in a GaAs layer ($N_d - N_a = 5 \times 10^{13}$ cm^{-3}, $\mu_n = 140\,000$ cm^2/V s), 32 μm thick, grown by GPE on a semi-insulating GaAs substrate; (b) Hanle effect (circles), excitation density 1.0 W/cm^2. Lorentzian fit (solid curve) gives $B_{1/2} = 280$ G, corresponding to $1/\tau_s = 1.1$ ns^{-1}. Inset: dependence of ρ_c on magnetic field in Faraday geometry. Solid curve: fit with Eq. (2) at $\tau_p = 270$ fs.

Our experimental results disagree with the standard theory of DP spin relaxation, which predicts proportionality between $1/\tau_s$ and the electron mobility. Indeed, the spin-relaxation rate for the nondegenerate statistics of electrons [12] is given by the expression (Chap. 3, Ref. [5])

$$\frac{1}{\tau_s} = Q_1 \alpha^2 \tau_p \frac{(kT)^3}{E_g \hbar^2}, \tag{1}$$

where the spin-orbit parameter $\alpha = 0.07$, E_g is the GaAs band gap. Q_1 is a number of the order of 1, depending on the scattering mechanism ($Q_1 = 3$ for scattering by polar optical phonons [13] and 1.5 for scattering by ionized impurities). Our results demonstrate a breakdown of Eq. (1) in high-mobility samples. The theoretical dependence of $1/\tau_s$, according to Eq. (1), is plotted in Fig. 2 for $Q_1 = 3$ (upper straight line) and 1.5 (lower straight line). In low-mobility samples the momentum scattering is determined by ionized impurities, while in high-mobility samples it is governed by phonons. Therefore the theory predicts that the dependence of $1/\tau_s$ on mobility should lie in between the two straight lines, starting from the lower one at low mobilities, and getting closer to the higher one at high mobilities (the theoretical limit is about 260 000 cm^2/V s at 77 K). One can see that the theory disagrees with the experiment both qualitatively and quantitatively: for high-mobility samples the experi-

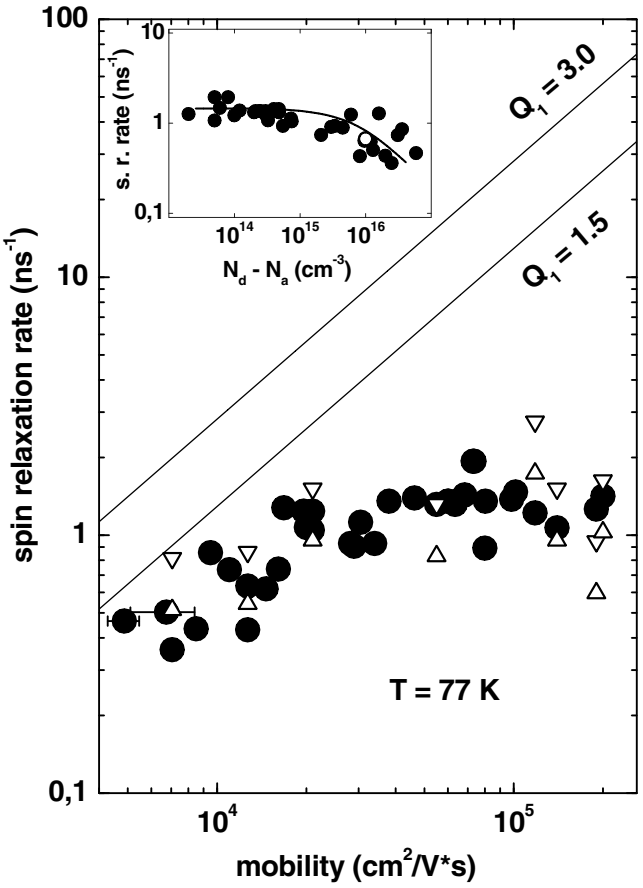

FIG. 2. Dependence of the spin-relaxation rate of electrons on their mobility. Solid circles: measurements with the Hanle effect. Solid lines: calculation with Eq. (1) at $Q_1 = 3$ (scattering by phonons, upper straight line) and $Q_1 = 1.5$ (scattering by ionized impurities, lower straight line). Triangles: calculation with Eq. (1) using τ_p measured from spin-relaxation suppression in Faraday geometry, assuming scattering by phonons ($\triangledown$), and ionized impurities ($\triangle$). Inset: Concentration dependence of the rate $1/\tau_s$. Solid curve is to guide for the eye. Data point ($\bigcirc$) is taken from Ref. [2] at $T = 100$ K.

mentally measured $1/\tau_s$ is more than an order of magnitude smaller than predicted by the DP theory and does not depend on mobility [14].

We are unlikely to understand this puzzling suppression result by searching for a *new* source of spin relaxation because an additional mechanism can only increase the $1/\tau_s$ rate in comparison with that given by the DP mechanism alone. Therefore, the main question to be understood here is the reason for the giant suppression of DP spin relaxation. The experiments on optical orientation in longitudinal magnetic field (Faraday geometry) provide an unambiguous answer: the spin-orbit (Dresselhaus) field in high-mobility GaAs is averaged out through a *new* mechanism that is much more efficient than and has nothing to do with the momentum scattering averaging. It is well known [5] that the optical orientation of electrons in a longitudinal magnetic field gives the correlation time of the random field responsible for the spin relaxation. In the case of free electrons, the effect of the longitudinal magnetic field is associated with the cyclotron rotation of the electron momentum around **B** with the frequency $\omega_c = eB/mc$. This rotation averages the momentum components perpendicular to **B**, thus diminishing the effective spin-orbit field. As a result, both τ_s and $\rho_c \sim \tau_s/\tau_J$ increase as a function of B. If **B** is directed along [100] and is not very strong ($\omega_c\tau_p < 1$), then the dependence $1/\tau_s(B)$ is parabolic (p. 96, Ref. [5]):

$$\frac{1}{\tau_s(B)} = \frac{1}{\tau_s(0)}[1 - \Theta(\omega_c\tau_p)^2]. \qquad (2)$$

Comparing Eq. (2) with experiment, one can determine the momentum relaxation time. The parameter $\Theta \approx 2$ [5] depends slightly on the specific scattering mechanism. The dependence of ρ_c on the longitudinal magnetic field is shown by circles in Fig. 1(b), inset. The solid curve is a fit using Eq. (2) resulting in $\tau_p \approx 270$ fs. This is 20 times *shorter* than the momentum scattering time taken from the Hall mobility, $\tau_p = 5.4$ ps ($\mu_n = 140\,000$ cm^2/V s). Substitution of the value of τ_p obtained from the longitudinal-field measurements into the expression for $1/\tau_s$ [Eq. (1)] gives the following results. For impurity scattering, we get $1/\tau_s = 1$ ns^{-1} (up-pointing triangle in Fig. 2), and for phonon scattering $1/\tau_s = 1.5$ ns^{-1} (down-pointing triangle in Fig. 2). These values are in good agreement with the value $1/\tau_s = 1.1$ ns^{-1}, measured independently in Voigt geometry (Hanle effect, solid circle in Fig. 2 for $\mu_n = 140\,000$ cm^2/V s). In the same manner, using the longitudinal-field measurements and Eq. (1), we have plotted spin relaxation for several other samples with different Hall mobilities (triangles in Fig. 2). The agreement with the spin-relaxation rates taken from the Hanle effect (circles in Fig. 2) is fairly good. We wish to stress that for all the samples for which the Faraday-geometry measurements were performed, the ''momentum relaxation time'' calculated using Eq. (2) (in fact, the *correlation time* of the spin-orbit field) was much shorter than τ_p taken from the Hall effect, and did not depend on mobility.

Temperature dependence of the spin-relaxation rate $1/\tau_s$ gives further evidence of the failure of the classical DP approach in high-mobility GaAs. The rate was measured (circles) on 7.5 μm thick MBE-grown GaAs ($n = 9 \times 10^{13}$ cm^{-3}, $\mu_n = 140\,000$ cm^2/V s at $T = 77$ K) at the center of the PL line (Fig. 3). One can see that the rate $1/\tau_s$ grows from 1 up to 20 ns^{-1} in the temperature range 50–250 K. The solid line in Fig. 3 is calculated according to Eq. (1) with the feature around 150 K being determined by the change of averaging mechanism from acoustic phonon and ionized impurity ($T < 150$ K) to polar optical phonon scattering ($T > 150$ K). Squares show the extension [15] of the DP scheme taking into

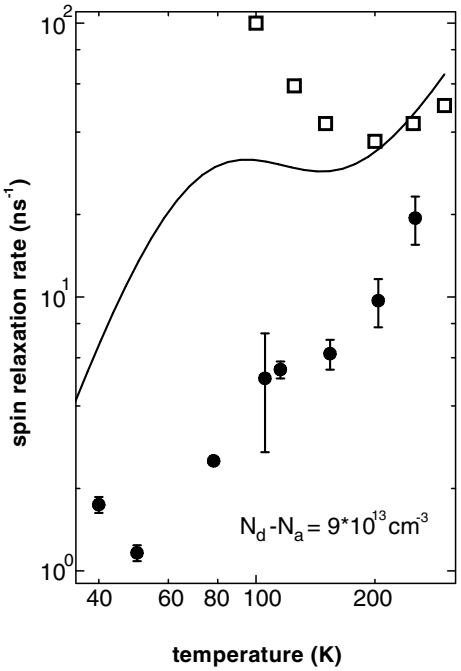

FIG. 3. Temperature dependence (solid circles) of the rate $1/\tau_s$ of MBE-grown 7.5 μ-thick GaAs with $N_d - N_a \approx 9 \times 10^{13}$ cm^{-3}. Solid curve is plotted according to Eq. (1) for ionized impurity, acoustic and polar optical phonon scattering elastic processes, whereas the open squares are calculated [15] for optical phonon scattering taking into account only its nonelastic character.

account the nonelastic character of the optical phonon scattering. In spite of the correct trend, there is quantitative disagreement between theory and experiment over the entire range of temperatures we have studied.

At first glance, electron-electron scattering can be invoked as a possible explanation since it suppresses spin relaxation but does not influence the mobility [16,17]. However, though effective in doped samples with low mobility, it normally plays no role in clean samples with high mobility. Another possibility is that, unlike the Hall effect, the Hanle effect measures the spin memory of *localized* near-band-gap electrons. Localization would suppress the DP mechanism. However, localization is highly unlikely in high quality samples at elevated temperatures. Therefore, the data lead us to conclude that ballistically flying electrons experience a rapidly fluctuating ($\tau_c \approx 300$ fs) spin-orbit field. However, the origin of the rapid fluctuations remains unknown [11].

To summarize, we have found a giant suppression of electron spin relaxation in high-mobility bulk *n*-GaAs that persists up to the room temperature.

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[11] An expanded version of this Letter can be found in R. I. Dzhioev *et al.*, cond-mat/0407133, which contains the details omitted in this version.
[12] All the samples studied are nondegenerated [Fermi energy $E_F(77\ \mathrm{K}) = kT$ at $n \approx 10^{17}$ cm^{-3}].
[13] The corrected value $Q_1 = 3$ is given in Russian version of Ref. [5].
[14] We rule out the photoinduced decrease in mobility. Otherwise we have to suppose that the mobility drops down from the dark values of 20 000 to 200 000 cm^2/V s to the same mysterious value of 10 000 cm^2/V s for samples with different doping, grown with different technologies. Also, illumination usually increases the mobility [C. Weisbuch and B. Vinter, *Quantum Semiconductor Structures* (Academic Press, New York, 1991), p. 102].
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