# Low-temperature spin relaxation in *n*-type GaAs

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Low-temperature electron-spin relaxation is studied by the optical orientation method in bulk *n*-GaAs with donor concentrations from $10^{14}$ cm$^{-3}$ to $5 \times 10^{17}$ cm$^{-3}$. A peculiarity related to the metal-to-insulator transition is observed in the dependence of the spin lifetime on doping near $n_D = 2 \times 10^{16}$ cm$^{-3}$. In the metallic phase, spin relaxation is governed by the Dyakonov-Perel mechanism, while in the insulator phase it is due to anisotropic exchange interaction and hyperfine interaction

## I. INTRODUCTION

The research on the physics of nonequilibrium spin in semiconductors has been conducted for more than 30 years, since the first experiments on optical orientation of electron and nuclear spins, performed by Lampel[1] on Si, Parsons[2] on GaSb, Ekimov and Safarov[3] on AlGaAs, and Zakharchenya *et al.*[4] on GaAs. Basic facts and a considerable body of experimental and theoretical results related to bulk Group III-V semiconductors are collected in Ref. 5. Later on, much new information concerning mainly low-dimensional structures has been obtained. Nevertheless, there remain gaps in this knowledge that have become visible with the emerging of an application-directed angle on spin-related phenomena (spintronics).[6] Though it was known to specialists that *n*-type semiconductors demonstrate, generally, extended spin lifetimes,[7,8] a recent finding of over-100-ns spin memory[9] in bulk gallium arsenide with the donor concentration of $10^{16}$ cm$^{-3}$ became a surprise, and generated an increased interest in *n*-type semiconductors as a possible base for spintronic devices. It was suggested that the spin lifetime as a function of donor concentration $n_D$ has a maximum at $n_D$ near $10^{16}$ cm$^{-3}$. Later on, an even longer spin lifetime of nearly 300 ns was reported in a GaAs/AlGaAs heterostructure.[10,11] However no detailed experimental or theoretical study of the dependence of the electron-spin relaxation on doping has been performed so far. This paper is aimed at filling this gap. The choice of GaAs for this study is justified not only by its prospective spintronic applications, but also by the fact that the physics of spin systems in this semiconductor is otherwise very well studied. Once an understanding of the spin-relaxation processes is reached for GaAs, it can be easily extended to other semiconductors. We use the optical orientation technique to measure the concentration dependence of the electron-spin-relaxation time in *n*-type epitaxial layers of GaAs at liquid-helium temperatures. Comparison of the experimental data with theory reveals the main mechanisms of spin relaxation relevant in this temperature range, and determines the limits to the spin lifetime in bulk *n*-type semiconductors.

## II. SAMPLES AND EXPERIMENTAL TECHNIQUE

We used 2-$\mu$m thick layers of GaAs between AlGaAs barriers, grown by the molecular-beam epitaxy (MBE)

($n_D = 5.5 \times 10^{14}, 2 \times 10^{16}, 4.6 \times 10^{16}, 5.6 \times 10^{16}, 9 \times 10^{16}$ cm$^{-3}$); 20-$\mu$m-thick layers grown by liquid-phase epitaxy ($n_D = 1 \times 10^{15}, 2 \times 10^{15}, 2.1 \times 10^{15}, 4 \times 10^{15}, 4.5 \times 10^{15}, 7 \times 10^{15}, 1 \times 10^{16}, 1.6 \times 10^{16}, 2.8 \times 10^{16}$ cm$^{-3}$); a bulk Chochralskii-grown crystal ($n_D = 5 \times 10^{17}$ cm$^{-3}$); a 0.1-$\mu$m-thick GaAs buffer layer of a multi-quantum-well structure ($n_D = 1 \times 10^{14}$ cm$^{-3}$).

The samples were placed in a liquid-helium cryostat and pumped by a tunable Ti-sapphire laser, with the circular polarization of light being alternated in sign at a frequency of 26.61 kHz with a photoelastic quartz modulator. This allowed us to eliminate the effect of the lattice nuclear polarization on the optical orientation of the electrons (Chaps. 5 and 9 of Ref. 5). The geomagnetic field was compensated for, and as a result, it did not exceed 0.1 G at the sample. The photoluminescence (PL) polarization was measured in the reflection geometry by a circular-polarization analyzer. The PL was dispersed by a double-grating spectrometer (5 Å/mm). A two-channel photon counting device synchronized with the quartz modulator provided the measurement of the effective degree of circular polarization, $\rho_c = (I_+ - I_-)/(I_+ + I_-)$, where $I_+$ and $I_-$ are the intensities of the $\sigma_+$ PL component under the $\sigma_+$ and $\sigma_-$ pumping, respectively. $\rho_c$ may be considered as a Stokes parameter characterizing the PL circular polarization. It is proportional to the amplitude value of the average electron spin induced by the alternately polarized pump light.

The method of determination of the spin relaxation time in *n*-type semiconductors by steady-state optical orientation is based on the following physical grounds.[5,10] After creation of an electron-hole pair by circularly polarized light, the hole rapidly loses the memory about its initial spin state. Then it recombines with an electron. Besides, under low pump intensity the probability of recombination with a photoexcited electron is negligible as compared with the probability to recombine with one of the unpolarized equilibrium electrons. Thus, spin- polarized photoexcited electrons eventually substitute unpolarized equilibrium electrons, and spin polarization accumulates in the crystal. If the density of photoexcited carriers is spatially uniform, then, under cw excitation, the spin lifetime is given by the expression

$$T_S = \left( \frac{1}{\tau_s} + \frac{1}{\tau_J} \right)^{-1}, \tag{1}$$

where $\tau_s$ is the spin-relaxation time, $\tau_J = n/G$, $n$ is the concentration of equilibrium electrons, and $G$ is the excitation density (the rate of creation of photocarriers per unit volume). $\tau_J$ is an analog of the electron lifetime. It has the meaning of the characteristic time during which the population of equilibrium electrons is renewed by replacement with photoexcited electrons via the creation and recombination of electron-hole pairs (see Chap. 2 of Ref. 5). The suppression of the electron-spin orientation in the transversal magnetic field (the Hanle effect) in this simplest case is described by the Lorentz curve:

$$s_z(B) = s_z(0) \frac{1}{1 + (\mu_B g B T_S / \hbar)^2}, \qquad (2)$$

where $B$ is the magnetic field, $\mu_B$ is the Bohr magneton, and $g$ is the electron $g$ factor.

If the concentration of photoexcited carriers significantly changes over the region where electron spins are polarized, it is not possible to describe the entire ensemble of electrons by the unique $\tau_J$. In this case, the Hanle curve is no longer Lorentzian. Also, spin diffusion may result in non-Lorentzian Hanle curves on the high-energy side of the PL spectrum.[8] However, in our experiments none of these effects have been observed: within the experimental accuracy, the Hanle curves were Lorentzian and identical within the width of the PL lines.

As in GaAs the $g$ factor is known, Eq. (2) allows to determine $T_S$ from the Hanle effect: the half-width of the curve, $B_{1/2} = (\hbar/\mu_B g) T_S^{-1}$, is proportional to the inverse spin lifetime. It follows from Eq. (1) that $T_S$ and, therefore, the width of the Hanle curve, depends on the excitation intensity. To obtain the value of $\tau_s$, one should take $T_S$ in the low-pump limit.

### III. EXPERIMENTAL RESULTS AND DISCUSSION

PL spectra [Fig. 1(a)] of samples with low doping level ($n_D < 10^{15}$ cm$^{-3}$) consist of up to three overlapping lines corresponding, to the best of our knowledge,[12] to the recombination of free excitons ($X$,1.5155 eV), excitons bound to neutral donors ($D^0 X$,1.5145 eV), and excitons bound to charged donors ($D^+ X$,1.5136 eV). The free-exciton recombination forms the high-energy wing of the spectrum. With the increase of donor concentration, these lines merge into one broad line. Under optical orientation conditions, PL is circularly polarized. Both the zero-field polarization and the width of the Hanle curve decrease with the decrease of excitation intensity, which is typical for the optical orientation of equilibrium electrons.[5] In samples with low donor concentration, the polarization degree is the highest at the high-energy wing corresponding to excitonic transitions, then it falls down to zero at the maximum of the $D^0 X$ line, and slightly increases with further decrease of the PL photon energy. The dip in the spectral dependence of $\rho_c$ results from the coupling of the spins of the two electrons in the $D^0 X$ complex into a singlet state.

In heavily doped samples, the circular polarization is only observed at the high-energy wing of the spectrum [Fig. 1(b)].
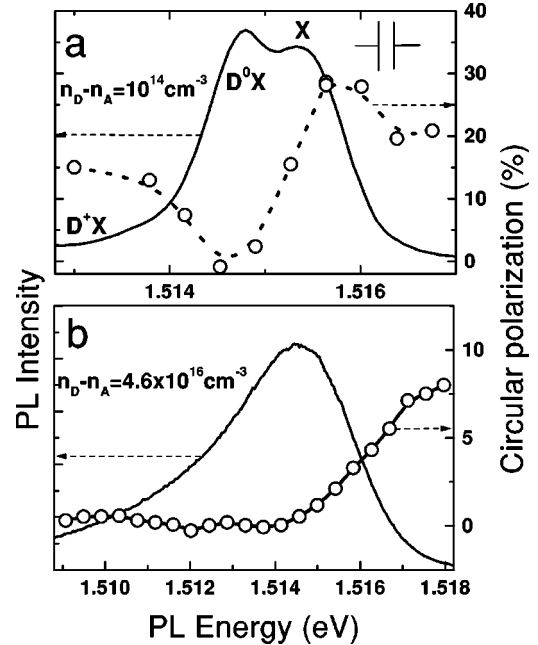


FIG. 1. Spectra of photoluminescence (PL) intensity (solid lines) and of the PL circular polarization (dash lines) in GaAs: (a) 0.1-$\mu$m-thick GaAs layer with electron concentration $n_D - n_A \approx 10^{14}$ cm$^{-3}$ (insulating). Spectra taken in zero magnetic field under excitation by light with the photon energy $h\nu = 1.519$ eV and intensity $W = 40$ mW/cm$^2$. (b) 2-$\mu$m-thick GaAs layer with $n_D - n_A \approx 4.6 \times 10^{16}$ cm$^{-3}$ (metallic), spectra measured at $h\nu = 1.520$ eV and $W = 2$ W/ cm$^2$.

This behavior reflects the Fermi statistics of delocalized electrons in degenerate semiconductor crystals: only Fermi-edge electrons may have a nonzero average spin. The dependence of the polarization degree on the transversal magnetic field (the Hanle effect) is the same for all the PL energies. This is an evidence that, under the sufficiently low excitation densities we used, the PL polarization at all the photon energies reflected the state of the same spin reservoir, namely, that of equilibrium electrons,[13] and the differences in the polarization degree were due to specific recombination conditions rather than spin dynamics. Consequently, measuring $T_S$ at the limit of low pump density yielded the value of $\tau_s$ characterizing the electron ensemble of the sample under study.

An example of the dependence of $T_S$ on pump intensity is shown in Fig. 2. The Hanle curve becomes steadily narrower with decreasing intensity. The half-width of the Hanle curve vs pump density is plotted in the inset. It is well fitted by a linear dependence, whose cutoff at zero pump gives the desirable spin-relaxation rate. This procedure was used to determine $\tau_s$ for each of our samples. The results are shown in Fig. 3. To fully represent the available experimental information, we plot here also data from Ref. 9 obtained using the time-resolved pump-probe technique. In spite of the considerable scattering of experimental points (this results, in our opinion, mainly from errors in the determination of the donor concentration, and from incontrollable impurities present in the samples), they give an unambiguous picture of spin relaxation over a wide range of doping. The most remarkable feature of the concentration dependence of $\tau_s$ is that it has
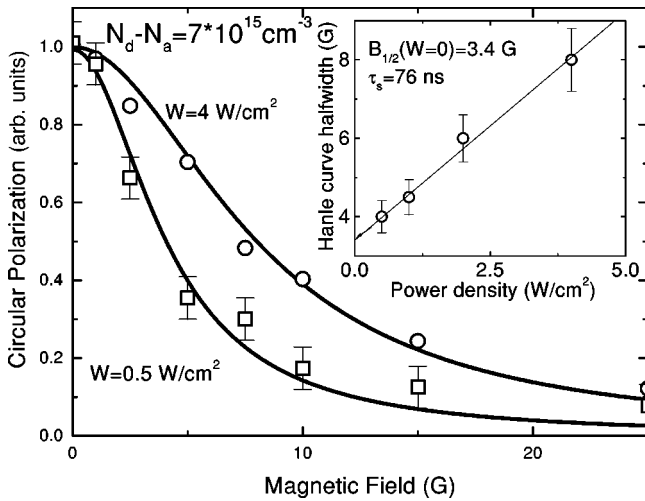
FIG. 2. Magnetic depolarization of photoluminescence (Hanle effect) at pump densities $W=4$ W/cm$^2$ (circles) and $W=0.5$ W/cm$^2$ (squares). Experimental values of the circular polarization degree $\rho_c$ are divided by $\rho_c(B=0)$. Solid lines: fit by Lorenzians with half-widths of 8 G and 4 G. Inset: the Hanle-effect half-width as a function of pump density. Extrapolation to zero pump density gives $B_{1/2}=3.4$ G, corresponding to the spin relaxation time $\tau_s=76$ ns.

*two* maxima. With the increase of doping from $10^{14}$ cm$^{-3}$ upwards $\tau_s$, being initially about 5 ns, becomes longer, reaching values around 180 ns at $n_D \approx 3 \times 10^{15}$ cm$^{-3}$, then decreases down to approximately 50 ns at $n_D \approx 1.5$
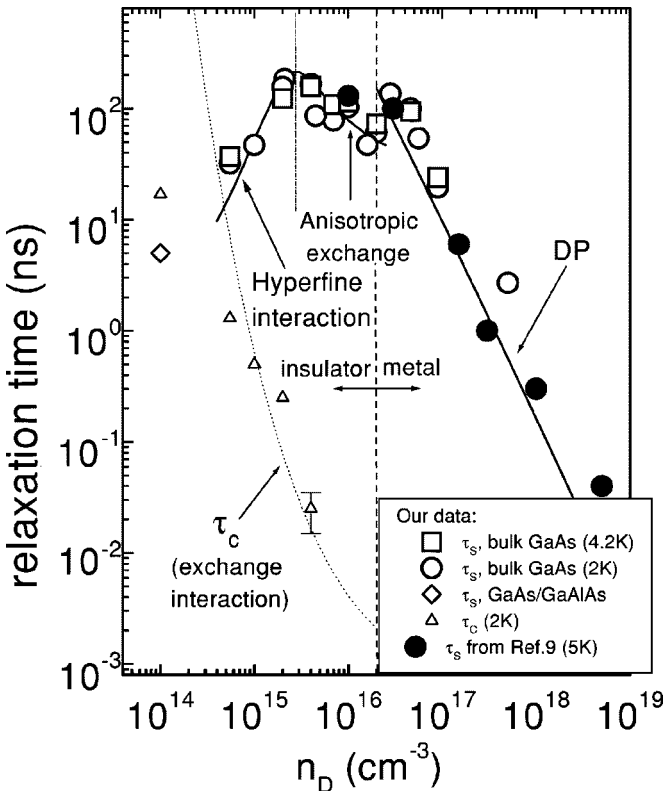


FIG. 3. Spin-relaxation time $\tau_s$ and spin-correlation time $\tau_c$ as functions of donor concentration in $n$-GaAs. Solid lines: theory.

$\times 10^{16}$ cm$^{-3}$. Further increase of the donor concentration results in an abrupt threefold rise of the spin-relaxation time, followed by its steady and steep decrease ($\tau_s$ becomes shorter by nearly four decimal orders over the next two orders in the donor concentration). The spin-relaxation time is virtually the same at 2 and 4.2 K, which suggests that in this temperature range, scattering by phonons has practically no impact on the electron spin, and that, in heavily doped samples, we observe the spin dynamics of electrons obeying a degenerate statistics.

We interpret this unusual concentration dependence as a manifestation of three mechanisms of spin relaxation relevant for equilibrium electrons at low temperature: hyperfine interaction with spins of lattice nuclei,[14,15] anisotropic exchange interaction of donor-bound electrons,[16] and the Dyakonov-Perel (DP) mechanism.[17] The maximum at $n_D = 3 \times 10^{15}$ cm$^{-3}$ is due to a crossover between relaxation mechanisms originating from the hyperfine interaction with lattice nuclei and from the spin-orbit interaction. The peculiarity at $n_D = 2 \times 10^{16}$ cm$^{-3}$ is associated with the metal-to-insulator transition (MIT).[18] It reflects the change of the specific mechanism through which the spin-orbit coupling affects the spin lifetime: in the metallic phase it is the DP mechanism, while in the insulator phase ($n_D < 2 \times 10^{16}$ cm$^{-3}$) it is the anisotropic exchange.

All the three mechanisms can be interpreted in terms of effective magnetic fields acting upon the electron spin. Spin-orbit interaction in crystals without inversion symmetry, such as GaAs, is known to produce effective fields determined by the direction and value of the electron wave vector **k**. Scattering by defects or phonons results in this field's rapid changing in time; the spin is therefore exposed to a stochastic field which causes its relaxation.[17] This is referred to as the Dyakonov-Perel mechanism. It has been shown that an analogous field affects the spin of an electron tunneling through a potential barrier.[16] As a result, the exchange interaction of donor-bound electrons in GaAs turns out to be anisotropic, and the flip-flop transition of spins of two electrons coupled by the exchange interaction goes along with the rotation of each of the spins through the same small angle $\gamma \approx 0.01$, but in opposite directions. The axis of the rotation, as well as the value of $\gamma$, depends on the orientation of the pair of donors in the crystal. In the ensemble of randomly distributed donors, this process leads to the relaxation of the total spin of the donor-bound electrons.[16] Another contribution into the spin-relaxation rate of localized electrons comes from their interaction with nuclear spins. As the donor-bound electron interacts with a great number of nuclei, $N \approx 10^5$, the effect of nuclei upon the electron spin **S** can be always presented as a Larmor precession of **S** in an effective "hyperfine" magnetic field with the contribution of all the nuclear spins within the electron orbit (Chap. 2 of Ref. 5; Ref. 14). The hyperfine field produced by the mean-squared fluctuation of the nuclear spin is equivalent to the combined action of $\sqrt{N} \approx 300$ spins, which amounts to $\approx 54$ Oe for GaAs.[11]

One can see that these three mechanisms give the qualitative picture of the concentration dependence of $\tau_s$, which is consistent with our experimental observations. Indeed, at low donor concentrations, electrons are effectively isolated,

and their spins precess independently in random static nuclear fields. This results in the disappearance of the most part of the electron-spin orientation within a few nanoseconds.[11,15] Then, with increasing donor concentration, electron wave functions begin to overlap, and the isotropic exchange interaction brings about flip-flop transitions, which results in the dynamical averaging of the hyperfine interaction: the electron spin ceases to be bound to a single donor and interacts with a greater number of nuclei, so that the effect of nuclear-spin fluctuations becomes smaller. As a result, $\tau_s$ increases. On the other hand, a stronger overlap of wave functions is accompanied by a greater probability to lose spin orientation due to the anisotropic exchange interaction. Eventually, the anisotropic exchange becomes stronger than the hyperfine interaction, and the rise of the spin lifetime is changed for the decrease. Finally, above MIT, the DP mechanism governs spin relaxation. The increase of the Fermi wave vector with the electron concentration makes the DP spin relaxation faster, and $\tau_s$ gets steadily shorter. The discontinuity in the concentration dependence of $\tau_s$, observed at MIT, suggests that at this concentration, spin relaxation in the insulator phase (via anisotropic exchange) is faster than in the metallic phase (DP). This conclusion agrees with the results of theoretical calculations for dielectric and metallic phases (see below); however, we cannot propose any quantitative theory of spin relaxation in the MIT region.

A common feature of all the spin-relaxation mechanisms based on spin precession in random magnetic fields is that they can be suppressed by applying a longitudinal magnetic field. Indeed, this is equivalent to placing the electrons in a rotating frame, where transverse components of random fields are reduced as a result of dynamical averaging. The characteristic magnetic field required to suppress spin relaxation can be found from the relation $\Omega_L \tau_c = 1$, where $\Omega_L$ is the Larmor frequency and $\tau_c$ is the correlation time of the random field. We performed experiments in longitudinal magnetic fields, placing our samples into a superconducting solenoid immersed in liquid helium under exhaust pumping (at 2 K). This setup did not allow to measure the Hanle effect; however we were able to detect changes in spin-relaxation time by measuring the dependence of $\rho_c$ on the magnetic field. Since we used excitation with light of alternating helicity, and detected the polarization signal at the modulation frequency (26.6 kHz), the field-induced circular polarization of PL (Ref. 19) did not contribute to the measured signal, which was, respectively, entirely due to the optical orientation of electron spins. The detected increase of $\rho_c$ with magnetic field was therefore associated with the suppression of spin relaxation, and the characteristic magnetic fields determined for each sample were used to calculate $\tau_c$. The results are shown by triangles in Fig. 3. We were unable to measure $\tau_c$ for samples with donor concentration higher than $4 \times 10^{15}$ cm$^{-3}$ because the strong magnetic fields required caused shifts of the PL spectral lines, which resulted in strong parasite signals due to the spectral dependence of $\rho_c$. Such measurements at higher donor concentrations can be possibly done using time-resolved techniques. The value of $\tau_c$ for the sample with a donor concentration of $10^{14}$ cm$^{-3}$, where $\tau_c > \tau_s$ and the regime of isolated donors

is supposed to be realized,[11] was calculated from experimental data by using a more complicated procedure, as described in detail in Ref. 11.

One can see that the measured values of $\tau_c$ fall into the nanosecond and subnanosecond range. Therefore, $\tau_c$ cannot be associated with the nuclear-spin system which has much longer relaxation times (Chap. 2 of Ref. 5), and must be attributed to electrons. This means that $\tau_c$ is in fact the local spin lifetime at a fixed donor; formally, this can be written as a decay time of the electron-spin correlation function:

$$\tau_c = \frac{1}{S(S+1)N_D} \sum_i \int_0^\infty \langle \mathbf{S}_i(0) \cdot \mathbf{S}_i(t) \rangle dt, \qquad (3)$$

where angular brackets denote quantum-mechanical averaging, $i$ numerates donors, and $N_D$ stands for the total number of donors in the crystal.

Due to various spin-conserving processes providing spin transfer within the impurity band, $\tau_c$ indeed can be much shorter than the spin lifetime of the entire electron ensemble. For donor-bound electrons at low temperature, the most relevant mechanism of spin transfer is the exchange interaction of electrons localized at adjacent donors. This conclusion is qualitatively consistent with the steep decrease of $\tau_c$ with donor concentration—this is a consequence of the increased overlap of electron wave functions. The estimation we performed using this model (see dotted line in Fig. 3; details of calculations are given in the following section) indeed shows a good agreement with all the available experimental data on bulk samples, i.e., at concentrations from $5.5 \times 10^{14}$ cm$^{-3}$ to $4 \times 10^{15}$ cm$^{-3}$. At lower $n_D$, the exponential concentration dependence, characteristic for the exchange mechanism, gives very long $\tau_c$, which becomes much longer than corresponding spin-relaxation times at concentrations of the order of and below $10^{14}$ cm$^{-3}$. This fact suggests that additional mechanisms of correlation decay may be significant at low donor concentrations, where the exchange interaction is less effective. This conclusion is backed by the data of Weisbuch,[7] who reported a spin-relaxation time as long as 20 ns in a bulk GaAs sample with $n_D = 10^{13}$ cm$^{-3}$. At such a low donor concentration, the regime of isolated donors must have been realized, which would have resulted in a shorter $\tau_s$, about 5 ns, due to spin precession in the fluctuation nuclear field.[11,15] A longer time observed indicates that, most likely, $\tau_c$ in that sample was rather short; however the specific reason for shortening the correlation time is not clear. One of the possible mechanisms, namely, exchange interaction with free conduction-band electrons, was studied in Ref. 11. It was shown that additional electrons present in space-charge layers of doped heterostructures can significantly reduce $\tau_c$. In the presence of additional electrons, the spin lifetime in a GaAs layers in a MBE-grown multilayer structure (with the nominal doping level of $10^{14}$ cm$^{-3}$) was as long as 290 ns, which corresponds to $\tau_c \approx 0.1$ ns. Recharging the GaAs layer under illumination allowed to reduce the spin lifetime nearly 100-fold,[11] down to 5 ns, while $\tau_c$ became as long as 17 ns (these data are shown in Fig. 3). The correlation time of 17 ns is still much shorter that what can be expected of exchange interaction at $n_D = 10^{14}$ cm$^{-3}$. Possi-

bly, some background concentration of free electrons remained in the layer even under illumination, which would have explained why $\tau_c$ was shorter than expected in this specific sample. However, it remains unclear whether or not delocalized electrons can be present in bulk samples at liquid-helium temperatures. Our data do not give an unambiguous answer to this question, and the issue of mechanisms of correlation decay in samples with low donor concentrations remains open for future research.

## IV. THEORY

### A. Isolating phase ($n_D < 2 \times 10^{16}$)

In order to estimate whether or not the exchange interaction can provide the observed values of $\tau_c$, it is worth noting that the exponential dependence of the exchange constant $J$ on the interdonor distance must result in an exponential decrease of $\tau_c$ with increasing donor concentration. In the limit of extremely low concentrations, only nearest neighbors contribute to the exchange interaction. The distribution function of the distance to the nearest neighbor has a maximum at $r_1 \approx 0.54 n_D^{-1/3}$. At higher concentrations, second-nearest neighbors having a peak of the distribution function at $r_2 \approx 0.74 n_D^{-1/3}$, and third-nearest neighbors ($r_3 \approx 0.8 n_D^{-1/3}$), also contribute to the interaction. It is easy to estimate that at $10^{15}$ cm$^{-3} \lesssim n_D \lesssim 10^{16}$ cm$^{-3}$ the interaction with the nearest neighbor dominates, though second and third neighbors also contribute. Therefore, the correlation time can be estimated as

$$\tau_c \approx \hbar / \xi J(r_c), \tag{4}$$

where $r_c = \beta n_D^{-1/3}$, $\beta$ and $\xi$ are numerical factors of the order of one, $J(R) = 0.82 E_B (R/a_B)^{5/2} \exp(-2R/a_B)$ (Ref. 20). The value $r_c = \beta n_D^{-1/3}$ has the meaning of the average characteristic distance between effectively interacting donors at a given concentration. Therefore, one should expect $\beta$ to be in between 0.54 and 0.8. Figure 3 shows that a good fit to the available experimental data for bulk samples by Eq. (4) is reached at $\beta = 0.65, \xi = 0.8$ (Fig. 3, dotted line). In spite of some scattering of experimental points, the agreement with the model at very reasonable values of parameters is remarkable. This is indeed evidence that $\tau_c$ in this concentration range is governed by the isotropic part of the exchange interaction. One cannot exclude, however, that there exist other physical processes dominating the decay of the single-donor spin correlation [Eq. (3)] at low donor concentration, where the exchange interaction is ineffective. Since experimental data in this concentration range are insufficient, we consider it premature to include in the theoretical treatment specific mechanisms of the correlation decay which may be relevant here (see discussion at the end of the preceding section). In the following, we will use the experimentally determined values of $\tau_c$ to calculate spin-relaxation times.

With the knowledge of the concentration dependence of $\tau_c$, it becomes possible to calculate the contributions to the spin-relaxation rate coming from hyperfine interaction and anisotropic exchange interaction, and therefore to find out $\tau_s$ in the insulating phase. The expression for the spin-

relaxation time of donor-bound electrons due to hyperfine interaction with lattice nuclei was derived by Dyakonov and Perel.[14] At zero external magnetic field it reads

$$\frac{1}{\tau_{SN}} = \frac{2}{3} \langle \omega_N^2 \rangle \tau_c, \tag{5}$$

where $\omega_N$ is the frequency of the electron-spin precession in an effective fluctuating magnetic field produced by the nuclear spins within the electron orbit. For shallow donors in GaAs, $\langle \omega_N^2 \rangle^{1/2} = 2 \times 10^8$ s$^{-1}$ (Ref. 11). The spin dynamics of isolated localized electrons interacting only with nuclei (this case is possibly realized at donor concentrations of the order of, or less than, $10^{14}$ cm$^{-3}$) has been considered theoretically in Refs. 15 and 21. Equation (5), valid when $\langle \omega_N^2 \rangle^{1/2} \tau_c \ll 1$, is a result of motional averaging of the random hyperfine fields, acting upon the electron spin. As discussed above, the motional narrowing at $n_D > 1 \times 10^{15}$ cm$^{-3}$ is most likely due to rapid flip-flop transitions induced by the exchange interaction. In the ensemble of randomly distributed donors, these flip-flop transitions can be interpreted as the jumping of a chosen spin over different donors. The spin, on the average, spends a time equal to $\tau_c$ at each of the donors it visits. Due to the anisotropy of the exchange interaction, each jump is accompanied by a rotation of the spin through a small angle $\gamma$. This results in spin relaxation with the characteristic time $\tau_{sa}$ given by the expression

$$\frac{1}{\tau_{sa}} = \frac{2}{3} \langle \gamma^2 \rangle \tau_c^{-1}, \tag{6}$$

The mean-squared value of $\gamma$ as a function of the interdonor distance $R$ can be calculated numerically using Eq. (16) of Ref. 16, which gives the following approximate expression for $\langle \gamma^2(R) \rangle^{1/2}$ valid within the range of interdonor distances from 1 to 20 Bohr radii:

$$\langle \gamma^2(R) \rangle^{1/2} = \frac{\alpha \hbar^3}{m \sqrt{2 m E_g} E_B a_B^3} \left[ 0.323 + 0.436 \left( \frac{R}{a_B} \right) + 0.014 \left( \frac{R}{a_B} \right)^2 \right], \tag{7}$$

where $m$ is the electron mass, $E_B$ and $a_B$ are the Bohr energy and the Bohr radius of the donor-bound electron, respectively, $\alpha$ is a dimensionless factor at the cubic in $k$ term in the conduction-band Hamiltonian (Chap. 2 of Ref. 5). For GaAs, $\alpha$ is known to be about 0.07 (Chap. 3 of Ref. 5); here we use the value 0.063, determined in Ref. 22 from spin-flip Raman scattering.

We took $R_{av} = 0.65(n_D)^{-1/3}$ for the average interdonor distance relevant for the exchange interaction, as the above considerations suggest. The solid line in Fig. 3 represents the theoretical concentration dependence of $\tau_s$, calculated as $\tau_s = (1/\tau_{sn} + 1/\tau_{sa})^{-1}$. The concentration dependence of the correlation time $\tau_c$ at $5 \times 10^{14}$ cm$^{-3} < n_D < 4 \times 10^{15}$ cm$^{-3}$ is taken from the experiment, while an extrapolation using Eq. (4) is used at $4 \times 10^{15}$ cm$^{-3} < n_D < 2 \times 10^{16}$ cm$^{-3}$.

### B. Metallic phase ($n_D > 2 \times 10^{16}$)

The spin-relaxation time at donor concentrations over $2 \times 10^{16}$ cm$^{-3}$, i.e., in the metallic phase, has been calculated assuming that the electron mean spin is accumulated near the Fermi level and that the Fermi energy $E_F \gg k_B T$. According to Chap. 3 of Ref. 5, if the electron momentum scattering is dominated by collisions with charged impurities, the spin-relaxation time of electrons with energy $E$ is

$$\tau_S = \frac{315}{16} \alpha^{-2} \frac{\hbar^2 E_g}{E^3 \tau_p(E)}, \tag{8}$$

where $\tau_p$ is the momentum relaxation time. In the degenerate case we deal with, $E$ stands for the Fermi energy $E_F = (3\pi^2)^{2/3} \hbar^2 n_D^{2/3}/2m$. To calculate $\tau_p$ as a function of $n_D$, we used the Brooks-Herring method,[23] i.e., evaluated, in the Born approximation, the scattering cross section of an electron off the Coulomb potential screened by the degenerate electron gas. This approach gives the following expression for $\tau_p$:

$$\frac{1}{\tau_p} = \frac{\pi n_D e^4}{\varepsilon^2 E_F^{3/2} \sqrt{2m}} \left[ \ln(1+x) - \frac{x}{1+x} \right], \tag{9}$$

where $x = 8mE_F r_0^2/\hbar^2 = 3^{1/3} \pi^{5/3} a_B n_D^{1/3}$ and the screening radius $r_0 = \frac{1}{2} (\pi/3)^{1/6} (a_B n_D^{-1/3})^{1/2}$. Substituting Eq. (9) into Eq. (8), and assuming that $E = E_F$, we obtain the formula for the spin-relaxation time

$$\tau_S = \frac{315}{16} \alpha^{-2} \frac{E_g}{\pi^5 \hbar^3 a_B^2 n_D^2} \left[ \ln(1+x) - \frac{x}{1+x} \right], \tag{10}$$

which was used to calculate the theoretical curve for $\tau_s(n_D)$ at $n_D > 2 \times 10^{16}$ cm$^{-3}$.

One can see that the theory demonstrates a fairly good agreement with the experimental data all over the studied concentration range, both in the dielectric and the metallic phase. A slight systematical shift of the calculated curve towards shorter $\tau_s$ in the metallic region may be due to the overestimation of the momentum relaxation time in our calculations. The measurement of the low-temperature electron mobility along with the experiments on spin orientation may be helpful in order to clarify this point. And, of course, the peculiarity observed near MIT demands detailed experimental and theoretical studies.

### V. CONCLUSION

Our results show that natural limits for the low-temperature spin lifetime in bulk GaAs and other cubic compound semiconductors are placed by stochastic precession of electron spins in random fields created by the hyperfine interaction and by the spin-orbit interaction. Against commonplace expectations, the crossover between these two main modes of spin decay in GaAs occurs not at the metal-to-insulator transition ($n_D = 2 \times 10^{16}$ cm$^{-3}$) but at lower donor concentrations [$n_D \approx (2-4) \times 10^{15}$ cm$^{-3}$], where electrons are bound to donors. A peculiarity related to the metal-to-insulator transition (MIT) is clearly seen in the dependence of the spin lifetime on doping near $n_D = 2 \times 10^{16}$ cm$^{-3}$. This peculiarity is due to changing the specific mechanism through which the spin-orbit coupling affects the spin lifetime: in the metallic phase it is the Dyakonov-Perel (DP) mechanism, while in the insulator phase it is the anisotropic exchange interaction. The maximal value of $\tau_s$ of free Fermi-edge electrons in heavily doped samples is reached just above the metal-to-insulator transition, where the Dyakonov-Perel relaxation is the weakest. Another maximum of $\tau_s$ is in the dielectric phase, at an optimal concentration determined by the interplay of the hyperfine interaction and the anisotropic exchange interaction. Specifically in bulk GaAs this is the absolute maximum of the spin lifetime, about 200 ns. However, this value is the lifetime of the mean spin of the entire electron ensemble. The spin lifetime at an individual donor, often discussed in relation to quantum information processing, is limited either by the period of precession in the fluctuation nuclear field ($\approx 5$ ns), or by the spin transfer to other donors, characterized by the correlation time $\tau_c$. In our experiments, $\tau_c$ never exceeded 20 ns; in samples with the longest spin relaxation times [$\tau_s \approx 180$ ns at $n_D \approx (2-4) \times 10^{15}$ cm$^{-3}$], $\tau_c$ was of the order of 0.2 ns. $\tau_c$ is a very important parameter that determines the relative contributions of hyperfine and spin-orbit interactions and, ultimately, the spin lifetime of localized electrons for a given semiconductor. In bulk GaAs samples at $n_D > 5 \times 10^{14}$ cm$^{-3}$, it is governed by the exchange hopping of the electron spin over the impurity band. At lower concentrations, it may be affected by other processes, for instance, by exchange interaction with delocalized electrons.[11] This fact opens a possibility to realize optical or electrical control over the spin lifetime of localized electrons in semiconductor structures.

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