



Extended pump-probe Faraday rotation spectroscopy of the submicrosecond electron spin dynamics in *n*-type GaAs

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(Received 28 October 2016; published 14 December 2016)

We develop an extended pump-probe Faraday rotation technique to study submicrosecond electron spin dynamics with picosecond time resolution in a wide range of magnetic fields. The electron spin dephasing time T_2^* and the longitudinal spin relaxation time T_1 , both approaching 250 ns in weak fields, are measured thereby in *n*-type bulk GaAs. By tailoring the pump pulse train through increasing the contained number of pulses, the buildup of resonant spin amplification is demonstrated for the electron spin polarization. The spin precession amplitude in high magnetic fields applied in the Voigt geometry shows a nonmonotonic dynamics deviating strongly from a monoexponential decay and revealing slow beatings. The beatings indicate a two spin component behavior with a g -factor difference of $\Delta g \sim 4 \times 10^{-4}$, much smaller than the Δg expected for free and donor-bound electrons. This g -factor variation indicates efficient, but incomplete spin exchange averaging.

DOI: [10.1103/PhysRevB.94.241202](https://doi.org/10.1103/PhysRevB.94.241202)

Initialized electron spins in semiconductors undergo a complex dynamics depending on external magnetic field, interaction with other charge carriers and nuclei, spin-orbit interaction, etc. Knowledge of the resulting spin dynamics provides information on these interactions and related spin properties such as g factors and relaxation times which are important for basic research and application in information technologies. Commonly, information on spin properties is mostly obtained from resonance techniques such as electron paramagnetic resonance, optically detected magnetic resonance, spin-flip Raman scattering, or polarized photoluminescence (Hanle effect). The development of pump-probe Faraday/Kerr rotation spectroscopy has facilitated exploration of the coherent spin dynamics, in particular the Larmor spin precession around a magnetic field, with picosecond temporal resolution and opened new ways for spin control and manipulation [1–5].

The main limitation imposed on the standard pump-probe technique is the restricted time range that can be monitored. This restriction comes from the finite length of mechanical delay lines for the pump-probe delay limiting this time range to a few nanoseconds, which can be too short to address the carrier spin dynamics in semiconductors. To evaluate longer spin dephasing times the resonant spin amplification (RSA) technique [6,7] can be used, which, however, does not provide detailed insight into complex spin dynamics such as a nonexponential decay of spin polarization. Also, the longitudinal spin relaxation characterized by the T_1 time typically exceeds the nanosecond range, so that indirect optical techniques like the spin inertia method [8] have to be used, again with limited access to nontrivial spin dynamics.

Here we extend the standard pump-probe Faraday rotation (PPFR) technique to address a much longer time range by employing a tailored pump and probe pulse sequence, while maintaining picosecond time resolution. The technique is applied to the submicrosecond electron spin dynamics in bulk *n*-type GaAs. The spin dephasing time T_2^* measured thereby

from the dynamics of spin precession at low magnetic fields is in agreement with data recorded from the Hanle effect [9,10], RSA [6], and spin noise spectroscopy [11,12]. However, at increased magnetic fields the spin precession decay becomes nonexponential, a behavior hardly accessible in detail by other methods. This peculiar dynamics is characterized by slow beatings between two electron subensembles shedding light on the spin exchange averaging in semiconductors [13]. Further, we demonstrate the buildup of electron spin polarization in the RSA regime with increasing number of pump pulses in a train, which provides an alternative approach for measuring T_2^* . In longitudinal magnetic fields (Faraday geometry) the longitudinal spin relaxation time T_1 is measured in a wide range of fields.

The sample under study is a 350- μm -thick GaAs epitaxial layer doped with Si to provide a donor concentration $n_D = 1.4 \times 10^{16} \text{ cm}^{-3}$. Some results are also presented for samples with $n_D = 3.7$ and $7.1 \times 10^{16} \text{ cm}^{-3}$ having thicknesses of 170 μm . The samples were placed in a split-coil magnetocryostat in contact with helium gas at a temperature $T = 6 \text{ K}$. Magnetic fields up to 6 T were applied either parallel (Faraday geometry) or perpendicular (Voigt geometry) to the light propagation vector (and sample growth axis).

The extended PPFR technique [Fig. 1(a)] is a modification of the common pump-probe Faraday rotation technique, where circularly polarized pump pulses generate carrier spin polarization, which is then probed by the Faraday rotation of linearly polarized probe pulses after transmission through the sample. The temporal evolution of the spin polarization is traced by varying the time delay between pump and probe pulses. In order to go for long time delays and to have flexibility with setting excitation protocols we implement pulse picking for both pump and probe laser beams.

We use a Ti:sapphire laser emitting a train of 2 ps pulses with a repetition rate of 76 MHz (repetition period $T_R = 13.1 \text{ ns}$). The laser output is split into pump and probe [Fig. 1(a)]. In the pump path an electro-optical modulator (EOM) is installed to select trains of N pulses (from 1 to about 100) separated by T_R with arbitrarily long delay between

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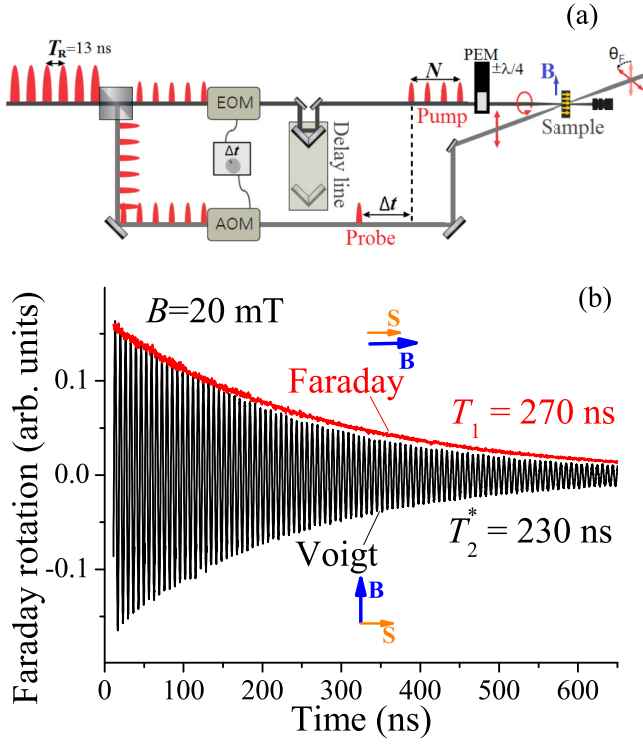


FIG. 1. (a) Scheme of extended PPR experiment. (b) Dynamics of Faraday rotation signal for $B_{\perp} = 20$ mT applied in Voigt (black line) and Faraday (red line) geometry. $n_D = 1.4 \times 10^{16} \text{ cm}^{-3}$.

the trains. An acousto-optical light modulator (AOM) in the probe path is used to select single pulses at the required delay after the pump train. Electronic variation of the delay between the synchronized AOM and EOM (also synchronized with the laser) allows for a coarse change of the delay between the pump pulse sequence and the probe pulse in steps of T_R , providing the desired time range. In addition, a mechanical delay line in the pump path allows for fine delay variation up to T_R . In this way the Faraday rotation dynamics could be measured over a microsecond time range with still 2 ps time resolution. Except for RSA experiments with variable N , we use here trains of $N = 8$ successive pump pulses, applied at a train repetition period of $80T_R = 1.05 \mu\text{s}$. To perform synchronous detection and to avoid nuclear polarization, the polarization of the pump was modulated between σ^+ and σ^- by a photoelastic modulator operated at a frequency of 84 kHz. The laser wavelength was set to 825 nm (827 and 829 nm for the samples with $n_D = 3.7$ and $7.1 \times 10^{16} \text{ cm}^{-3}$, respectively), below the GaAs band gap, to avoid complete absorption of the probe. The average pump power for the protocol “8 out of 80 pulses” was $P = 0.1$ mW. The diameter of the pump spot on the sample was about $100 \mu\text{m}$.

The black line in Fig. 1(b) shows the Faraday rotation dynamics measured with the extended PPR technique in a magnetic field $B_{\perp} = 20$ mT applied in the Voigt geometry for the sample with $n_D = 1.4 \times 10^{16} \text{ cm}^{-3}$. The signal shows oscillatory behavior caused by the electron spin precession at frequency $\omega = |g|\mu_B B_{\perp}/\hbar$, where $g = -0.44$ is the electron g factor in bulk GaAs [14] and μ_B is the Bohr magneton. The oscillation amplitude decays exponentially with the ensemble

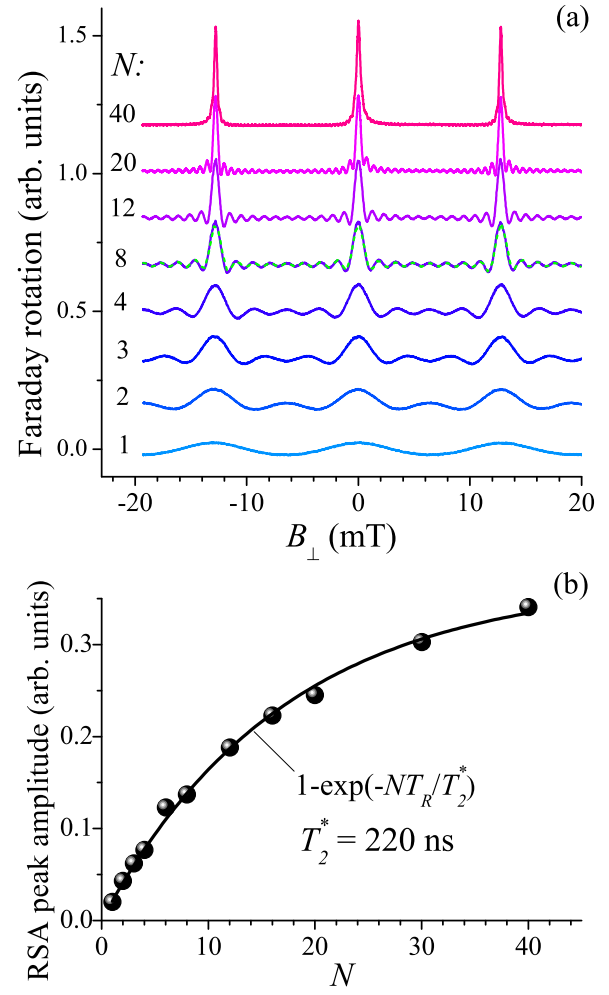


FIG. 2. (a) Resonant spin amplification curves for different numbers of pump pulses N before the probe pulse that is delayed by 12.9 ns relative to the last pump in a train. The green dashed line shows a fit to the experiment with Eq. (1) for $N = 8$. (b) Dependence of the RSA peak amplitude at $B_{\perp} = 0$ on the number of pump pulses. The solid line gives a fit to the experiment. $n_D = 1.4 \times 10^{16} \text{ cm}^{-3}$.

spin dephasing time $T_2^* = 230$ ns. Note that the separation between the pump pulse trains is $80T_R \approx 1.05 \mu\text{s} \gg T_2^*$. The measured T_2^* is close to the values obtained from RSA [6], Hanle [9,10], and spin noise [11,12] experiments at $B \approx 0$.

The extended PPR technique allows one to study the buildup of electron spin polarization in the RSA regime with increasing number of pump pulses N in the train that precedes the probe pulse. The delay between the probe and the last pump pulse in a train is set to $\Delta t = T_R - 0.2 \text{ ns} \approx 12.9$ ns [Fig. 1(a)]. The Faraday rotation signal is measured as a function of magnetic field applied in the Voigt geometry. Figure 2(a) shows RSA curves for different N . For $N = 1$, the RSA curve shows a sinusoidal oscillation with period $\Delta B_{\perp} = 2\pi\hbar/(\Delta t|g|\mu_B)$. With increasing N , resonances at magnetic fields $B_q = 2\pi\hbar q/(T_R|g|\mu_B) \approx q \times 13$ mT appear, where q is an integer. These resonances correspond to q complete spin revolutions between subsequent pump pulses. With increasing N the main RSA resonances increase in amplitude and narrow resulting in a curve with sharp peaks.

Between the RSA resonances $N - 1$ oscillations are seen corresponding to interference of spin precessions initiated by different pump pulses within a train. With increasing N the oscillations become faint and disappear for $N \rightarrow \infty$, so that they are not seen in standard RSA curves.

The RSA curve can be described by superposition of N damped oscillations [7]:

$$S = S_0 \sum_{q=0}^{N-1} \cos[\omega(\Delta t + qT_R)] \exp\left(-\frac{\Delta t + qT_R}{T_2^*}\right), \quad (1)$$

where S_0 is the spin polarization induced by a single pump pulse and the magnetic field dependence is represented by $\omega = |g|\mu_B B_\perp/\hbar$. The experimental dependencies are perfectly reproduced with Eq. (1); the corresponding fit is shown in Fig. 2(a) by the green dashed line for $N = 8$.

The half width at half maximum (HWHM) of the RSA peak for $N \gg T_2^*/T_R$ saturates at $\delta B_\perp = \hbar/(|g|\mu_B T_2^*)$, which gives the established way to evaluate T_2^* [6]. In our case the HWHM for $N \rightarrow \infty$ is 0.15 mT corresponding to $T_2^* \approx 170$ ns, underestimating somewhat the value from a direct measurement [see Fig. 1(b)], but being still in reasonable agreement.

The dependence of the RSA peak amplitude on N gives another way to determine T_2^* . Indeed, according to Eq. (1) $S(\omega = 0) \propto 1 - \exp(-NT_R/T_2^*)$, which well describes the experimental dependence in Fig. 2(b). The fit gives $T_2^* \approx 220$ ns, in good agreement with the extended PPRF measurement from Fig. 1(b).

Thus, the RSA technique gives the reasonable value of T_2^* for simple exponential dephasing of the spin polarization. At increased magnetic field, basic characteristics of the long-lasting spin dynamics can also be extracted from the RSA technique [6] as well as from the recently developed heterodyne detection of spin noise [15]. However, as we will show below, at increased B_\perp the dynamics reveals peculiarities hardly accessible by indirect methods. We turn now to measurements of the electron spin dynamics at long delays for pump trains containing eight pulses. The dynamics

are measured for different magnetic fields applied in the Voigt geometry. The precise magnetic field strength was adjusted in a range of a few mT around the given value to meet the RSA condition for maximal signal. Figure 3(a) shows the time dependence of the Faraday rotation oscillation amplitude in steps of 13.1 ns, i.e., of the envelope of the spin precession dynamics [upper inset in Fig. 3(a)], for the sample with $n_D = 1.4 \times 10^{16} \text{ cm}^{-3}$.

At $B_\perp = 50$ mT the spin precession amplitude shows an exponential decay [Fig. 3(a)]. At $B_\perp = 125$ mT the dynamics accelerates at longer times, thereby significantly deviating from an exponential decay. Further field increase reveals a dip in the dynamics, which shifts to shorter times with increasing field, while the overall dynamics accelerate.

The dip in the dynamics originates from beatings of the signals from spin subensembles with a small difference in precession frequency $\Delta\omega$. Indeed, the amplitude dynamics are well fitted with a precession amplitude (envelope) of the sum of two oscillating components $A_1 \exp(-t/\tau_1) \cos(\omega t) + A_2 \exp(-t/\tau_2) \cos[(\omega + \Delta\omega)t]$ as shown by the lines in Fig. 3(a). From the fits we determine the $\Delta\omega$, which scales linearly with magnetic field [see lower inset in Fig. 3(a)]. This suggests that $\Delta\omega$ arises from different g factors of two electron subensembles, so that $\Delta\omega = \Delta g \mu_B B/\hbar$ with $\Delta g \approx 4 \times 10^{-4}$.

The magnetic field dependencies of the decay times T_2^* for both subensembles are shown in Fig. 3(b) by the black squares. Their decrease with increasing field is related to the g -factor spread δg within each subensemble, described by the equation $1/T_2^*(B_\perp) = 1/T_2^*(0) + \delta g \mu_B B_\perp/\hbar$ [16]. Corresponding fits are shown by the solid lines in Fig. 3(b) and give δg of 2×10^{-4} and 1×10^{-4} for the two subensembles in the sample with $n_D = 1.4 \times 10^{16} \text{ cm}^{-3}$.

The spin precession dynamics for the samples with higher doping concentrations of $3.7 \times 10^{16} \text{ cm}^{-3}$ and $7.1 \times 10^{16} \text{ cm}^{-3}$ give spin dephasing times of 80 and 40 ns, respectively, at low B_\perp , while at higher magnetic fields they also reveal slow beatings, corresponding to $\Delta g = 1.9 \times 10^{-3}$

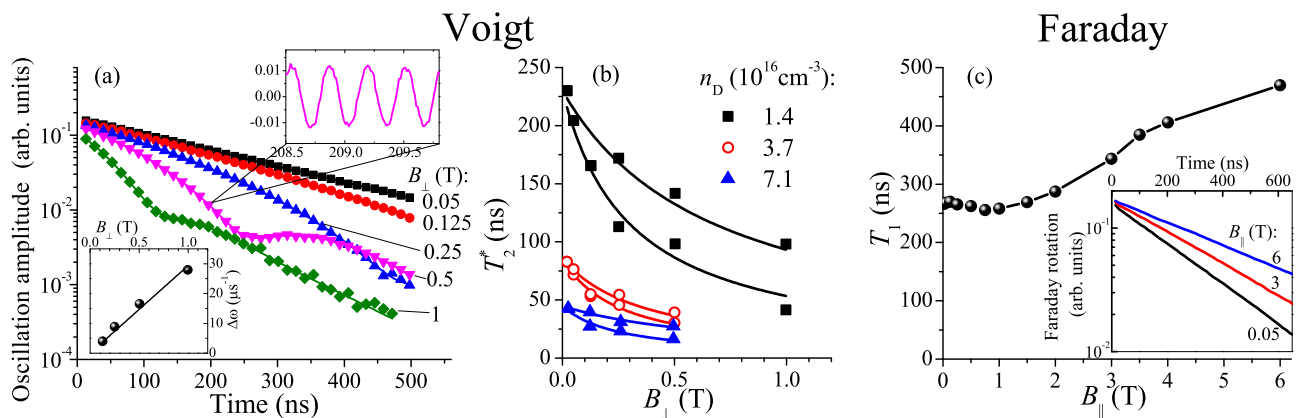


FIG. 3. (a) Dynamics of Faraday rotation oscillation amplitude for different magnetic fields applied in Voigt geometry. The upper inset shows a close-up of the spin precession. The lower inset shows magnetic field dependence of the beating frequency $\Delta\omega$; the line is a linear fit. (b) Magnetic field dependence of the transversal spin dephasing times T_2^* of the two beating components for three samples with different doping concentrations. (c) Dependence of the longitudinal spin relaxation time T_1 on the magnetic field applied in Faraday geometry. The inset shows the corresponding dynamics of the Faraday rotation. The lines in panels (a) and (b) show fits to the experimental data (see text); the lines in panel (c) are guides to the eye. The data in panels (a) and (c) correspond to the sample with $n_D = 1.4 \times 10^{16} \text{ cm}^{-3}$.

and 1.6×10^{-3} , respectively. The magnetic field dependencies of the decay times of the two components for these samples are included in Fig. 3(b).

The electron concentrations in the studied samples are close to the metal-insulator transition ($\sim 2 \times 10^{16} \text{ cm}^{-3}$) [9]. Therefore, it seems reasonable to attribute the two electron subensembles to free and donor-bound electrons. One can estimate the g -factor difference Δg_0 for free and bound electrons from the difference in their transition energies of $\sim 6 \text{ meV}$ [17] using the Roth-Lax-Zwerdling equation [18]. The result is $\Delta g_0 \sim 10^{-2}$, considerably larger than the measured $\Delta g \approx 4 \times 10^{-4}$. On the other hand, the exchange interaction between free and bound electrons provides an efficient averaging mechanism [13]. This mechanism can be qualitatively understood as frequent spin exchange between free and bound electrons by scattering, and it is analogous to the motional narrowing described in Ref. [19]. As a result, spin precession occurs on a *single* average frequency. However, spatial inhomogeneity in the donor distribution may result in a broadening of the frequency distribution. In particular, one may consider different spatial domains of free and bound electron concentrations.

The dispersion of the free electron g factor near the GaAs band gap that is approximately given by $g(E) = -0.44 + \beta E$, where $\beta = 6.3 \text{ eV}^{-1}$ [20] provides an additional mechanism of g -factor broadening. In particular, at $T = 6 \text{ K}$ the temperature broadening is expected to be $\delta g_T \approx 3 \times 10^{-3}$, which is an order of magnitude larger than the measured $\Delta g \approx 4 \times 10^{-4}$. Obviously, the spin exchange averaging mechanism is responsible for that.

It is straightforward to apply the extended PPRF technique to measuring the longitudinal spin relaxation time T_1 in a magnetic field B_{\parallel} applied in Faraday geometry. The red line in Fig. 1(b) shows the dynamics of the Faraday rotation at $B_{\parallel} = 20 \text{ mT}$ for the sample with $n_D = 1.4 \times 10^{16} \text{ cm}^{-3}$. The signal shows a monoexponential decay without oscillations with $T_1 = 270 \text{ ns}$, close to the measured $T_2^* = 230 \text{ ns}$ at low

magnetic fields. Note that for $B \rightarrow 0$ we expect $T_2^* = T_2 = T_1$. The decay is monoexponential in the whole range of magnetic fields $B_{\parallel} \leq 6 \text{ T}$ [inset in Fig. 3(c)], and the corresponding decay time T_1 increases with B_{\parallel} above $\sim 1.5 \text{ T}$ [Fig. 3(c)]. The suppression of spin relaxation by a longitudinal magnetic field is much weaker than reported for bulk GaAs with lower donor concentrations (well below the metal-insulator transition) [21–23]. Indeed, for the studied donor concentration close to the metal-insulator transition, the electron spin relaxation is dominated by the Dyakonov-Perel and anisotropic exchange mechanisms [9], which are less suppressed by the external field compared to the spin relaxation due to the electron hyperfine interaction with nuclei that is the dominating factor at low doping concentrations [22]. We are not aware of reports on the magnetic field dependence of T_1 near the metal-insulator transition and at higher electron densities.

In conclusion, we have developed an extended pump-probe Faraday rotation technique and demonstrated its potential in studying electron spin dynamics with picosecond resolution in a wide temporal range up to $1 \mu\text{s}$ and potentially longer. This enables direct measurement of the spin dephasing time T_2^* and longitudinal spin relaxation time T_1 for carriers in arbitrary magnetic fields. The technique can be used for high sensitivity spectroscopy of g factors barely accessible by other methods, e.g., RSA. The possibility of varying the pump pulse train composition from single to multiple pulses provides access to the detailed process of electron spin synchronization under periodic laser excitation.

We are grateful to M. M. Glazov, V. L. Korenev, and A. V. Poshakinskiy for valuable advice and discussions and to S. A. Crooker for providing the samples and discussions. We acknowledge the financial support of the Deutsche Forschungsgemeinschaft in the frame of the ICRC TRR 160 and the Russian Science Foundation (Grant No. 14-42-00015). Also, the support by the BMBF project Q.com-H is appreciated.

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