

Room-temperature electron spin relaxation in bulk InAs

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Polarization-resolved, subpicosecond pump-probe measurements at a wavelength of 3.43 μm are used to determine the electron spin relaxation time T_1 in bulk InAs at room temperature. The measured T_1 of 19 ± 4 ps is in excellent agreement with the theoretical value of 21 ps, which is obtained from a nonperturbative calculation based on the D'yakonov-Perel' mechanism of precessional spin relaxation [M. I. D'yakonov and V. I. Perel', *Sov. Phys. JETP* **38**, 177 (1974)].
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Modern electronics relies upon the principle of controlling electronic charge in semiconductors for information processing. Recently, however, there has been intense interest in using another fundamental electronic property, specifically spin, to design novel electronic devices and circuits ("spintronics").¹⁻⁵ The successful controlled electrical injection of spin packets into semiconductors could lead to a variety of important new devices, including semiconductor-based magnetic memories and spin-sensitive transistors and diodes. Spin dynamics have also drawn much attention recently due to the broad proposal that physical realizations of quantum computer bits (or qubits) might be achieved in semiconductors using coherent superpositions of carrier spin. Such potential applications have generated considerable research activity focused on the injection of spin-polarized currents from ferromagnetic metals or spin filters into semiconductors.^{3,6-9}

Spin dynamics in semiconductors have been studied extensively, with the majority of work focusing on either bulk or, more recently, heterostructures of GaAs. Early research relied on the continuous-wave, polarization-resolved measurement of photoluminescence from these structures.¹⁰⁻¹⁶ More recent time-resolved studies have shown, however, that the continuous-wave measurements are unreliable due to multicomponent decay processes that may be present and sample-to-sample dependence of the initial degree of spin polarization.¹⁷ *Time-resolved* photoluminescence, based on either the application of streak cameras¹⁸⁻²⁰ or upconversion techniques,^{17,21} has been used to determine the degree of initial spin polarization and the time dependence of the polarized component of emission from an optically generated and spin-polarized carrier distribution. Alternatively, time-resolved differential transmission²²⁻²⁷ or reflection^{28,5,29} have been used to probe the spin-polarized electronic distributions. In both cases, these techniques have been widely applied in the near infrared, with the focus in most instances being on GaAs-based heterostructures. In contrast, little attention has been directed toward spin dynamics in InAs, even though it may well play a critical role in the future of

spintronics. This may be in part due to the narrow band gap of InAs, which dictates the need for mid-infrared ultrafast spectroscopy of the spin dynamics.

At this stage it is not at all apparent what device designs will emerge for the various applications of semiconductor spin control or what specific material systems will form the basis of these devices. However, the InAs material system has several positive properties that may prove important for future spintronic devices. The presence of a surface state on InAs allows for contact with metallic materials without a significant Schottky barrier. This makes InAs a strong potential candidate for applications utilizing spin injection from ferromagnetic metals.^{30,31} InAs is also well suited for high-speed electronics, due to its large electron mobility.³² It is roughly lattice matched to GaSb and AlSb, which have very large band offsets with InAs, allowing a variety of interesting device structures to be designed from these three materials and their alloys. For example, the InAs/AlSb heterojunction offers an extremely large electron barrier, which can be used to produce large peak-to-valley current ratios in resonant tunneling and resonant interband tunneling diodes.^{33,34} The controlled injection and transport of spin packets in these structures could lead to high-speed, spin-sensitive electronic devices.

Whereas the electronic injection of spin-polarized populations in semiconductors remains a challenging and important research problem, the optical injection of such populations in quantum confined and/or strained III-V semiconductors can be achieved with sources in the appropriate spectral region. In such semiconductors, the light- and heavy-hole bands are split, and selection rules dictate that optical excitation with circularly polarized light tuned near the band edge will produce highly spin-polarized populations of electrons and holes. On the other hand, the optical injection of fully spin-polarized carriers in *bulk* III-V semiconductors is not possible due to the degeneracy of the light and heavy-hole states at $\mathbf{k}=0$. Nevertheless, partially spin-polarized distributions can still be created and probed as a consequence of the different transition strengths associated with heavy-hole and light-hole to conduction band transitions. We note that the spin relaxation observed in undoped bulk III-V semiconductors at room temperature is expected to be associated with electrons, as the fourfold degeneracy of

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the valence band at $\mathbf{k}=0$ and the large spin splitting that occurs away from the zone center are expected to result in extremely rapid decay of the hole spin polarization.

In a time- and polarization-resolved differential transmission measurement, a spin polarized carrier population is created with, e.g., a left circularly polarized (σ_-) pump pulse. The relative density of the optically generated spin-up (n_\uparrow) to spin-down (n_\downarrow) electrons is 3:1, owing to the form of the matrix elements for heavy-hole and light-hole interband transitions in III-V semiconductors. Thus, the resulting generated conduction spin polarization is $P=(n_\uparrow - n_\downarrow)/(n_\uparrow + n_\downarrow)=0.50$. If there is a background density n_B of unpolarized electrons in the sample, the spin polarization would be less than 50%, $P=0.5/(1+n_B/n_0)$ for an optically generated electron density n_0 . The change in transmission of the probe is dictated by both the densities (n_\uparrow, n_\downarrow) and the matrix elements. For small signals, the measured differential probe transmission is proportional to the change in absorption coefficient $\Delta\alpha$, which for low densities is proportional to the optically generated carrier density. A σ_- probe will therefore experience $\Delta\alpha^- \propto 3n_\uparrow + n_\downarrow$, while a right circularly polarized (σ_+) probe will experience $\Delta\alpha^+ \propto 3n_\downarrow + n_\uparrow$. Hence, one would anticipate a maximum spin-polarized signal of $(\Delta\alpha^- - \Delta\alpha^+)/(\Delta\alpha^- + \Delta\alpha^+)=0.25$. For a background carrier density, the maximum signal is reduced to $0.25/(1+n_B/n_0)$.

We describe measurements of the spin relaxation time T_1 associated with optically injected, spin-polarized electrons in *bulk* InAs at 300 K and the comparison of these results with calculations of T_1 based on the D'yakonov-Perel' mechanism.³⁵ Polarization-resolved, subpicosecond-resolution, differential transmission measurements were performed using pulses from a mid-infrared, ultrafast, optical parametric oscillator (OPO).³⁶ The system produces pulses of less than 200 fs duration tunable in the range of 2.7–4.5 μm (idler) and 1.0–1.18 μm (signal). For these measurements, the OPO idler was tuned to 3.43 μm , near the 300 K bulk InAs band edge. The sample was excited using either a linearly or circularly polarized pump pulse to produce either an unpolarized or partially spin-polarized carrier distribution. The optical excitation is estimated to produce a carrier density of $2 \times 10^{16} \text{ cm}^{-3}$. The photogenerated population was interrogated by measuring, as a function of time delay, the transmission of a degenerate and much weaker probe pulse, which was either of the same circular polarization (SCP), opposite circular polarization (OCP), or opposite linear polarization (OLP) relative to the pump. The InAs sample is not intentionally doped but *n* type with a measured electron mobility of $2.6 \times 10^4 \text{ cm}^2/\text{Vs}$ and an estimated background electron density of $2 \times 10^{16} \text{ cm}^{-3}$. The sample was bonded to a *c*-cut sapphire crystal and mechanically lapped and polished to a thickness of approximately 3 μm . This, and the use of a probe that was more tightly focused than the pump, ensured interrogation of a nearly uniform distribution of photogenerated carriers.

The results of these measurements are shown in Fig. 1 for SCP, OCP, and OLP configurations. The data are characterized by three distinct features. In all cases there is a transient increase in transmission that is initiated and decays in less than 1 ps. We attribute this to thermalization of the

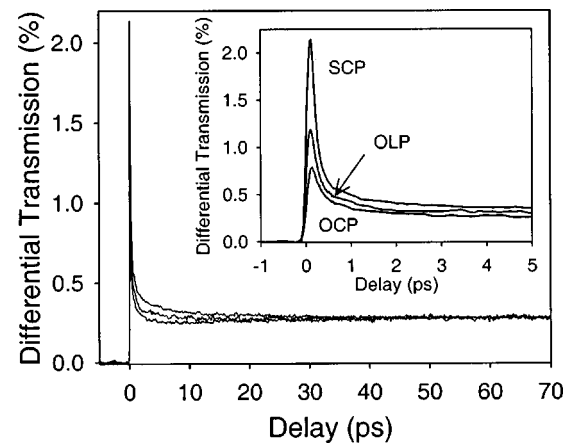


FIG. 1. Time-resolved differential transmission as a function of delay between pump and probe pulses for bulk InAs at 300 K. The three curves are, in order of increasing amplitude at early delay times, the OCP, OLP, and SCP signals. The inset shows an expanded view near zero delay.

optically injected carriers, hole spin relaxation, and to coherent artifacts associated with gratings written by the pump and probe for delays near zero. This feature is shown clearly in the inset. A second feature is a quasisteady-state increase in transmission for delays greater than 40 ps that is identical for all three configurations. This we attribute to band filling associated with a randomly spin-polarized carrier distribution. The final feature is a clear distinction among the SCP, OCP, and OLP data that occurs during the first 40 ps after excitation. In this delay region, the OCP data initially fall below and then slowly rises to the steady-state value. Both the SCP and OLP data decay to the steady-state value, with the OLP data decaying more rapidly.

The SCP and OCP data are characteristic of the evolution of a partially spin-polarized carrier distribution into a randomly polarized distribution,^{24–27} while the OLP data are consistent with expectations for an initially unpolarized population. The distinction between the SCP and OCP data is not nearly as dramatic as measured typically for quantum well structures.^{24–27} This is a consequence of both the light-hole/heavy-hole degeneracy and of the background electron density. The measured initial spin-polarized signal, $(\text{SCP}-\text{OCP})/(\text{SCP}+\text{OCP})$, is approximately 15%. This is consistent with the value of 12.5% expected for equal densities of background and optically generated electrons.

The difference in the SCP and OCP signals $(\text{SCP}-\text{OCP})$ is shown in Fig. 2 for delays in excess of 1 ps, a range for which the measurements are unaffected by coherent artifacts. The $(\text{SCP}-\text{OCP})$ data, which are not sensitive to the unpolarized component of the carrier population, are characterized by a single exponential decay with a time constant of 19 ps, as illustrated by the solid curve in Fig. 2. An uncertainty in this result of ± 4 ps is obtained by considering the error associated with fits to multiple similar data sets.

As a result of fast hole spin relaxation, the observed signal $(\text{SCP}-\text{OCP})$ is proportional to the spin polarization of the electrons and decays with a time constant T_1 . If a simple incoherent two-level model is used, the time T_1 is half the spin flip time. At room temperature, however, T_1 is expected to be dominated by the D'yakonov-Perel' mechanism,³⁵ which is a collective precessional mechanism.

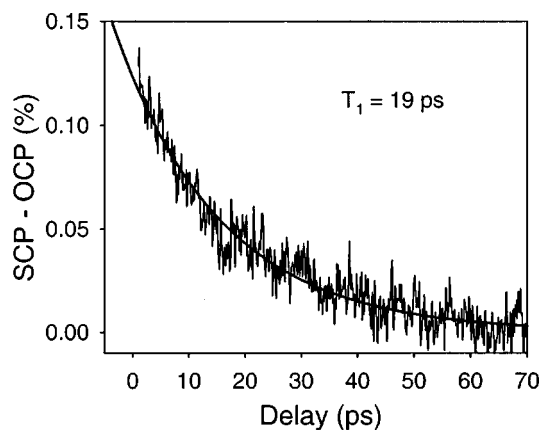


FIG. 2. Difference between the SCP and OCP signals as a function of delay between the pump and probe. The smooth curve is a decaying monoexponential fit that yields a spin relaxation time of 19 ps.

The origin of the precession is a splitting of the conduction band (due to spin-orbit coupling and inversion asymmetry) that results in an effective, \mathbf{k} -dependent magnetic field. Between collisions, spin precession around this pseudofield results in decay of the macroscopic spin polarization. Since the effective magnetic field is proportional to the cube of the electron momentum in zincblende semiconductors, this mechanism is particularly effective at room temperature.

We have performed calculations of T_1 in InAs at 300 K using both perturbative^{35,37} and nonperturbative values for the spin splitting. Both calculations, assuming a charged impurity scattering mechanism and a momentum relaxation time of 0.34 ps, result in a T_1 of 21 ps, which is in excellent agreement with the measured results. The momentum relaxation time is determined from the known 300 K mobility and the electronic effective mass $m^* = 0.023 m_0$.³⁸ The nonperturbative calculation is based on a 14-band $\mathbf{k}\cdot\mathbf{p}$ band structure that includes two conduction (antibonding) s states, six valence (bonding) p states, and six antibonding p states.³⁹ This restricted basis is the minimum required to generate nonperturbatively both the cubic anisotropy and the inversion asymmetry of the zincblende semiconductors. The small spin splitting in the conduction band, which leads to spin relaxation, emerges nonperturbatively from this multiband calculation. The lowest-order term (cubic) in the spin splitting (ΔE) of the conduction band calculated near zone center is the perturbative expression. The magnitude of the spin splitting energy along the (110) direction is $\Delta E = \gamma k^3$, where $\gamma = 103 \text{ eV \AA}^3$.³⁸

In summary, we have used a mid-infrared, subpicosecond, OPO and polarization-resolved pump-probe measurements to determine a room-temperature spin relaxation time T_1 of 19 ± 4 ps for bulk InAs. The measured results are in excellent agreement with both perturbative and nonperturbative calculations based on the D'yakonov-Perel' mechanism. The results are expected to be important in the consideration of future spin-sensitive electronics based on either bulk InAs or InAs-related heterostructures.

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