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Anomalous magnetic field dependence of the $T_1$ spin lifetime in a lightly doped GaAs sample

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The $T_1$ spin lifetime of a lightly doped $n$-type GaAs sample has been measured via time-resolved polarization spectroscopy under a number of temperature and magnetic field conditions. Lifetimes up to 19 $\mu$s have been measured. The magnetic field dependence of $T_1$ shows a nonmonotonic behavior, where the spin lifetime first increases, then decreases, then increases again with field. The initial increase in $T_1$ is understood to be due to correlation between electrons localized on donors. The decrease in $T_1$ is likely due to phonon-related spin-orbit relaxation. The final increase in $T_1$ with $B$ indicates a suppression of the spin-orbit relaxation that may involve a level-crossing related cusp in the Rashba or Dresselhaus contributions to relaxation, or may arise from an unknown source.

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Due to the twin emerging fields of spintronics and quantum computing, there is much current interest in the study of the spin of electrons in semiconductors.1–3 GaAs and related materials are especially interesting for reasons which include (a) the optical ability to orient and detect electron spins;4 (b) recent observations of long spin lifetimes in $n$-GaAs by many groups, beginning with Dzhioev et al.5 and Kikkawa and Awschalom,6 and (c) technology which makes possible the construction of single dot GaAs devices.7–9

Since spintronic and quantum computing applications require long spin lifetimes, the measurement and prediction of spin lifetimes is important in this field of study. Inhomogeneous transverse lifetimes, $T_2^*$, have been measured through a variety of techniques, and consistently yield lifetimes from $\sim 5$ ns to hundreds of nanoseconds for lightly doped GaAs material (depending on material parameters and experimental conditions). These techniques include the Hanle effect,5,10–12 time-resolved Faraday rotation,6 magnetic resonance,13–15 spin noise spectroscopy,16 coherent population trapping,17 and Raman spectroscopy.18 In these and other works, lightly doped GaAs has been studied due in part to the similarity between spin properties of electrons localized on isolated donors and electrons localized in quantum dots, the latter being potential building blocks for scalable solid-state quantum computers.19 The measured values for $T_2^*$ match up well against theoretical predictions of dephasing dominated by the electron-nuclear hyperfine interaction in this regime20 and are well-understood.

$T_2$ sets a lower bound for $T_2$, the homogeneous spin dephasing lifetime. $T_2$ has been theoretically predicted to be in the microsecond regime,20–22 and the recent experimental measurement of a 1 $\mu$s two-electron $T_2$ in a gated double quantum dot bears this out.23 Although $T_2$ is the quantity of most interest for quantum computing, the longitudinal spin flip time, $T_1$, is typically easier to measure. Recent theoretical work predicts that $T_2$ should be about the same as $T_1$ under realistic conditions, and the dephasing time can be as large as $2T_1$.24 Experimental measurements have confirmed that the $T_1$ lifetimes for GaAs are in the microsecond to millisecond regime, through time-resolved photoluminescence25,26 and through electronic measurement of spin relaxation in gated quantum dots.27 Theories which concentrate on the hyperfine interaction in GaAs quantum dots have predicted $T_1$ and $T_2$ times of microseconds, or possibly much longer;28–30 theories which focus on spin-orbit interaction agree that lifetimes should be in the microsecond regime, but predict that at high fields the spin-orbit interaction will cause substantial spin relaxation, as the spin relaxation rate varies as $B^2$.31–33 A power-law dependence of the spin relaxation rate on field was experimentally verified for donors in low-doped bulk GaAs by Fu et al. at high fields26 ($B \approx$ about 4 T), albeit with a dependence closer to $B^4$ rather than the quantum-dot prediction of $B^6$.

In the description of spin properties of GaAs, there can be effects from both localized and delocalized electrons. For example, in the lightly doped case the electrons are often considered to be localized, but correlation between electrons on nearby donor sites does occur. This is characterized by a “correlation time,” which can be pictured as the time it takes for spin information to hop from electron to electron. Although some theories account for this correlation time,34 much of the recent theoretical work assumes fully localized electrons as would be the case in quantum dots. There has in fact been some experimental evidence that in the lightly doped regime, both localized and delocalized electrons play a role in the spin characteristics of samples.25,35,36 The spin lifetimes of the two types of electrons can be very different; for example, for fully delocalized conduction electrons, spin dephasing and spin-flip times are predicted to be only in the tens of nanoseconds,37 much shorter than the above-quoted numbers for localized electrons. Substantial quantitative and qualitative experimental differences in spin lifetimes have been seen between electrons in $5 \times 10^{13}$ cm$^{-3}$ doped $n$-GaAs (Ref. 26) and $3 \times 10^{15}$ cm$^{-3}$ doped $n$-GaAs (Ref. 25), the degree of localization being the major culprit.

This paper uses the microsecond-regime time-resolved photoluminescence (PL) polarization technique developed in Ref. 25 (with minor modifications) to measure the $T_1$ of an $n$-GaAs sample in Faraday geometry. A long optical pump pulse orients the electronic spin; a significantly shorter optical probe pulse detects the polarization state some time later. The decay of the polarization is mapped out by varying the pump-pulse delay time. The polarization was shown to fol-
low an exponential decay, with characteristic time equal to $T_1$. The sample studied in detail in this paper was doped at $n = 1 \times 10^{15} \text{ cm}^{-3}$, higher than the sample investigated in Ref. 26 ($n = 5 \times 10^{13} \text{ cm}^{-3}$), and lower than the sample investigated in Ref. 25 ($n = 3 \times 10^{15} \text{ cm}^{-3}$). The $T_1$ times in this $1 \times 10^{15} \text{ cm}^{-3}$ sample fall between those two studies, but have a nonmonotonic dependence on magnetic field which resembles neither—the lifetimes first increase with $B$, then decrease, then increase again. This is markedly different from the $T_1$ measurements of localized electrons in Ref. 26 and the theories for localized electrons cited above, which all predict/show a strict decrease in $T_1$ with $B$ once spin-orbit interaction becomes the dominant relaxation mechanism.

The sample studied was a 1 $\mu$m thick GaAs layer in an AlGaAs heterostructure; its specific characteristics are described in detail in Ref. 12. The sample was placed in an optically accessible Oxford Instruments 7 T superconducting magnet and cooled to low temperatures with liquid helium. A Melles-Griot 785 nm 57ICS010 diode laser with modulation input was used for the optical excitation—typically at 10 mW—and focused onto the sample with a cylindrical lens providing a power density of approximately 2 W/cm$^2$. To obtain pulses, the laser was switched on and off in a controlled fashion on a nanosecond time scale with an Agilent 81110A pulse sequence generator. Photoluminescence was detected with a single grating Jobin-Yvon spectrometer with an integrated photomultiplier tube. A two-channel Stanford Research Systems photon counter was used to synchronously detect the effects of right and left circular polarized light excitation, and a 42 kHz Hinds Instruments photoelastic modulator was used in conjunction with a linear polarizer to vary the polarization state of the incident light. The experimental pulse sequence followed that of Ref. 25, and both right and left circular polarizations were detected so that the effects of thermal and optical polarizations could be separated. (The thermal polarization was not used; henceforth “polarization” will refer to the optical polarization only.) Figure 1 shows a representative PL spectrum and the optically induced polarization.

In the pump-probe experiment, the pump pulse initially aligned electron spins and the probe pulse detected the spin state of electrons at some time later. Rather than having the pump pulse differ from the probe pulse in intensity, as is often the case in pump-probe experiments, the two differed in length: the pump pulse was long enough to inject enough polarized photoelectrons to substantially affect the polarization of the doped electrons (through rapid spin exchange between the photoelectrons and the doped electrons). The probe pulse did not inject enough polarized electrons to substantially affect the overall electron polarization, but was able to probe the existing state of the polarization. For the bulk of the experiments, an 80 ns pump pulse and an 8 ns probe pulse were used (see Fig. 1 inset). Varying the delay between pump and probe pulses caused the polarization to decay exponentially after the initial pump pulse (see Fig. 2); the exponential decay constant is the $T_1$ spin flip time. Data was collected for a variety of temperatures and magnetic fields; see Fig. 3(a) for 1.5 and 5 K data. The longest lifetimes, up to 19 $\mu$s, were measured at high field and 1.5 K. This is an order of magnitude longer than the $T_1$ lifetime measured by the same technique in Ref. 25 for the $n = 3 \times 10^{15} \text{ cm}^{-3}$ sample.

The very longest spin lifetimes proved challenging to measure. The 42 kHz PEM triggered the overall pulse sequence and set a limit on how long the pump-probe delay could be made—delays greater than 5 $\mu$s proved problematic. Thus for the very longest spin lifetimes (more than ~8 $\mu$s), only the initial, linear, part of the decay could be observed. To obtain the decay time from fits we had to make assumptions about the decay baseline, which translated to a much larger uncertainty in the longest lifetimes. Additionally, it was not clear if the dark period between the pump and probe pulses was completely dark—if a small amount of light continued to shine from the laser or from stray room light,
the small continual addition of oriented photoelectrons to the system would create an artificial pump-pulse over long time scales, and would set an artificial limit on the measured lifetime. Our estimates on the stray light place this limit at around 40–400 $\mu$s, but we could potentially be underestimating the stray light. These two factors combine to make us reluctant to give too much weight to the apparent leveling out of the extremely long spin lifetimes at high fields in the 1.5 K data.

On the other hand, the dip in the spin lifetime at 2.5 T in the 1.5 K data is likely real. Figure 3(b) shows $T_1$ plotted vs $B$ for many more temperatures. As the temperature increases, the dip becomes more and more pronounced—the depth is a higher fraction of the peak. It also shifts to higher fields, to about 4 T. Notice that at the higher temperatures the lifetimes are smaller, and the large uncertainty that was present around 4 T. Notice that at the higher temperatures the lifetimes are smaller, and the large uncertainty that was present around 4 T, the spin lifetime first increases, then decreases, then increases again. The intermediate decrease is enhanced for higher temperatures.

To analyze the $T_1$ data, we discuss the three regimes of magnetic field dependence individually. First, in the low field data, $B$ less than 2–2.5 T, the electron spin relaxation is due to hyperfine interaction with the nuclei. Correlation between electrons leads to motional averaging, increasing the spin lifetime from the fully localized case. The initial increase in $T_1$ with $B$ is due to a reduction in this mechanism, and is well-understood.\textsuperscript{34} This was also seen in the earlier work on the $3 \times 10^{15}$ cm$^{-3}$ sample.\textsuperscript{25} As with the previous sample, $1/T_1$ followed a Lorentzian dependence with $B$, and the width yielded the correlation time. The correlation times obtained by fitting the 0.1–2.5 T data\textsuperscript{38} for various temperatures were 65±9, 39±4, 32±5, 28±2, and 20±2 ps for 1.5, 5, 6.5, 8, and 12 K, respectively. These are fairly consistent with the correlation times measured in Ref. 25, but are much shorter than the, e.g., 500 ps deduced by Dzhioev et al. for this doping level at 2 K.\textsuperscript{11} The $T_1$ data for this low-field regime all display a similar temperature dependence. Although theoretical predictions of this dependence are not known to the authors, an empirical fit is given here to aid future theoretical work: all curves seem to share the same power-law dependence of $T_1 \sim T^{-1.57}$ (see Fig. 4). By way of contrast, the other two regimes did not yield any easily recognized temperature dependence (although the potentially problematic nature of the long-lived 1.5 K points at the highest fields might make any existing dependence challenging to recognize).

In the second regime, starting at about 2–2.5 T, a decrease in $T_1$ with $B$ is exactly the expected behavior when phonon-related relaxation from spin-orbit interaction comes into play: as mentioned above, for the completely localized case of electrons in quantum dots, $1/T_1 \sim B^5$. While the electrons in this sample are not completely localized like those in the quantum dots for whom the $B^5$ dependence has been predicted, it seems likely that in this sample phonon-related relaxation will increase substantially with $B$ (as was seen in the sample studied in Ref. 26). Thus while a power-law behavior is not necessarily seen here, the decrease in $T_1$ in this regime seems very likely to be from this source, which would then be expected to dominate for large enough fields.

Adding support to this identification are the following observations: (a) The beginnings of this type of behavior may have been seen in the $3 \times 10^{15}$ cm$^{-3}$ sample, as reported in Ref. 25. Since the electrons in the current sample are more localized than in that sample, they would be expected to be even closer to the case of the $5 \times 10^{13}$ cm$^{-3}$ electrons of Ref. 38.
26, which show spin relaxation from this source very clearly. (b) As mentioned above and shown in Fig. 3(b), there is a more pronounced decrease with field for elevated temperatures—this is consistent with the temperature dependence one would expect for phonon-related effects.

In the third regime, $T_1$ increases again with $B$, for $B$ larger than $\sim 4$ T. This is unexpected. Most theories have consistently predicted a continual increase in phonon-related spin-orbit relaxation for large fields, and not a decrease. If such is the case, additional spin relaxation mechanisms cannot explain the data since they would only continue to decrease $T_1$. Instead, a suppression of the spin relaxation from spin-orbit effects must be taking place—this may be similar to the 77 K suppression above the ground state, abruptly increases several orders due to an accidental degeneracy of the two lowest levels with magnetic field. However, the Rashba-related relaxation, Dresselhaus-related spin relaxation increases monotonically with magnetic field. In two-dimensional quantum dots. Dresselhaus-related spin-orbit relaxation for large fields, and not a...

As an alternate possible explanation, we reference the theory of Bulaev and Loss, who studied the Dresselhaus and Rashba components of spin-orbit-related spin relaxation in two-dimensional quantum dots. Dresselhaus-related spin relaxation is produced by bulk inversion asymmetry and is found in all GaAs; Rashba-related relaxation is produced by structural inversion asymmetry and is typically only found in quantum-confined structures. Bulaev and Loss found that the Dresselhaus-related spin relaxation increases monotonically with magnetic field. However, the Rashba-related relaxation, due to an accidental degeneracy of the two lowest levels above the ground state, abruptly increases several orders of magnitude and has a cusplike peak at $\sim 5$ T, with a width of $\sim 1$ T. This is nearly the same field and width as the cusplike dip in $T_1$ we measured in our sample, shown in Fig. 3(b).

Although Bulaev and Loss’s specific theory involved wave functions of 2D quantum dots, if a similar level crossing exists in the energy levels of our electrons localized on donors, it could give rise to a similar cusp in spin relaxation—this seems an intriguing possible explanation of the decrease and subsequent increase in $T_1$ that we observed. The cusp could potentially arise from either Dresselhaus- or Rashba-related relaxation; Dresselhaus would be the natural candidate for bulk GaAs, but Rashba could be a possibility if the main source of relaxation is occurring near the boundary of our 1 μm thick layer. An experimental confirmation of Bulaev and Loss’s cusplike spin relaxation would obviously be highly important.

In conclusion, $T_1$ spin flip lifetimes up to at least 19 μs have been measured in an $n=1\times10^{15}$ cm$^{-3}$ GaAs sample. These long lifetimes bode well for measuring microsecond or longer $T_2$ spin coherence lifetimes via a spin echo technique under similar temperatures and magnetic fields, and experiments are progressing towards that end. The spin flip lifetimes reported in this work increase with magnetic field, as expected from correlation effects, then decrease with magnetic field, as expected from spin-orbit relaxation. However, there is an anomalous increase with field again, for $B \geq 4$ T, indicating a suppression of the spin-orbit relaxation. This suppression may involve a cusp in the Rashba- or Dresselhaus-related contributions to relaxation, or may arise from an unknown source; additional theory seems to be needed.

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27 Reference 7 set a lower bound of a few microseconds for $T_1$ at 150 mK and $\sim$2 T. Reference 8 set a lower bound of 50 $\mu$s for $T_1$ at $\sim$20 mK and 7.5 T. That lower bound was extended in a more precise measurement in Ref. 9: $T_1$ spin lifetimes up to 850 $\mu$s were measured at $\sim$20 mK (lattice temperature) and 8 T.


38 The lifetimes at 0 T are not discussed here; as with Ref. 25 they were considerably shorter than at B=0.1 T, presumably due to a component of hyperfine relaxation of localized spins which gets suppressed when the external field is larger than the hyperfine fluctuation field (see Ref. 25). Decreased spin lifetimes for fields below $\sim$0.1 T have also been seen by the following group and attributed to a similar mechanism: P.-F. Braun, X. Marie, L. Lombez, B. Urbaszek, T. Amand, P. Renucci, V. K. Kalevich, K. V. Kovokin, O. Krebs, P. Voisin, and Y. Masumoto, Phys. Rev. Lett. 94, 116601 (2005).
