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 μs 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 ~5 ns to hundreds of nanoseconds for lightly *n*-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 regime[11] 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 μs 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 in some situations 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 photoluminescence[25,26] 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^5.[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 fields[26] ($B \geqslant$ about 4 T), albeit with a dependence closer to B^4 rather than the quantum-dot prediction of B^5.

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 5×10^{13} cm^{-3} doped *n*-GaAs (Ref. 26) and 3×10^{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 $n=1\times10^{15}$ cm^{-3}, higher than the sample investigated in Ref. 26 ($n=5\times10^{13}$ cm^{-3}), and lower than the sample investigated in Ref. 25 ($n=3\times10^{15}$ cm^{-3}). The T_1 times in this 1×10^{15} 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 μm 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 μs, 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 $n=3\times10^{15}$ cm^{-3} sample.

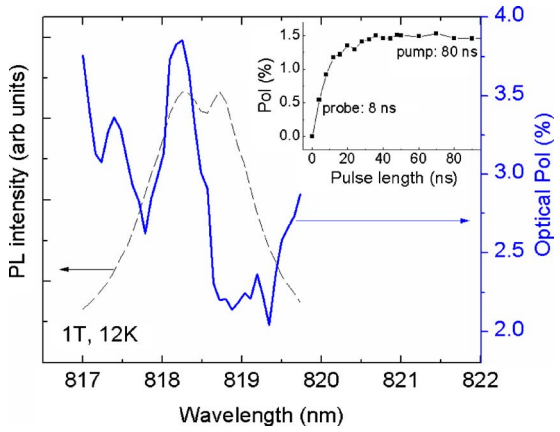

FIG. 1. (Color online) Representative cw photoluminescence is shown, under 1 T and 12 K conditions. The PL polarization is plotted, with thermal and optical effects separated. The optical polarization displays a characteristic peak at the free exciton luminescence which was used to set the spectrometer wavelength for the pulsed light experiments. Inset: representative optical PL polarization vs pulse width, which allowed pump and probe pulse widths to be set. This particular data was at 4 T and 1.5 K.

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 μs proved problematic. Thus for the very longest spin lifetimes (more than $\sim$8 μs), 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,

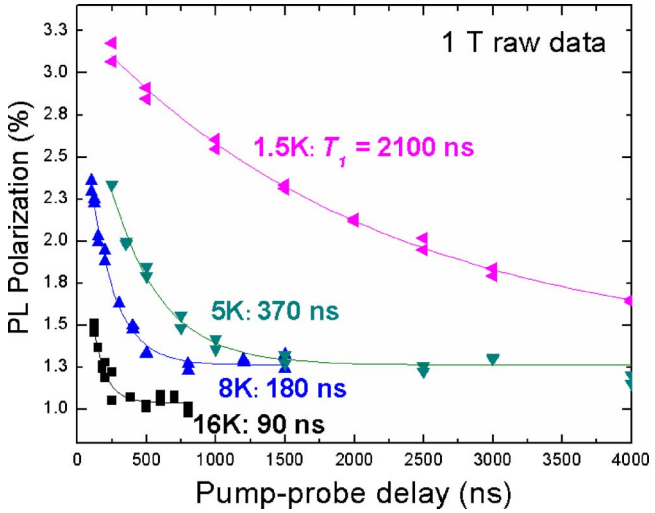

FIG. 2. (Color online) Representative raw data, at 1 T and various temperatures. The polarization decrease vs pump-probe delay is plotted. Data has been fit to exponential decays, from which the T_1 spin flip time is deduced.

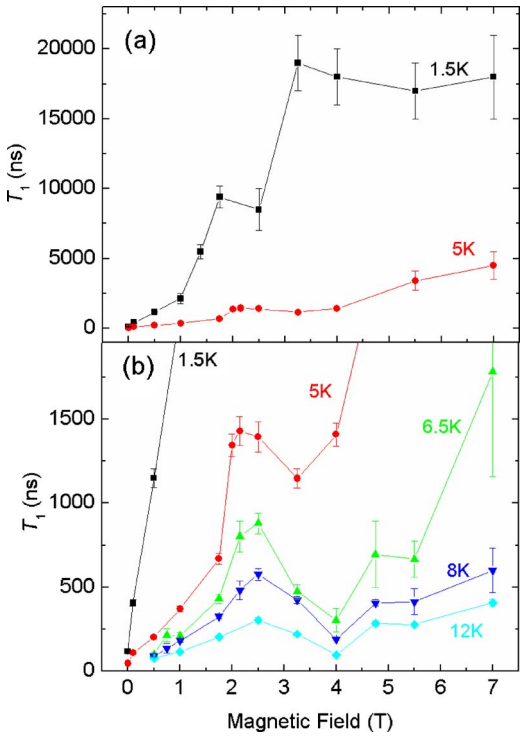

FIG. 3. (Color online) (a) Summary of the T_1 spin-flip measurements for 1.5 and 5 K, plotted vs magnetic field. The maximum spin lifetimes were about 19 μs and occurred at low temperature and high field. (b) T_1 vs B for additional temperatures. The dip at 3 to 4 T becomes more and more pronounced for higher temperatures.

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 μs, 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 due to the very long lifetimes, disappears—so the dip is well within the experimental measurement capability. Thus as the magnetic field is increased from 0 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

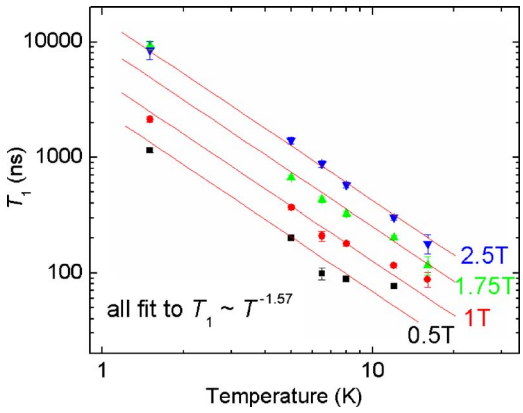

FIG. 4. (Color online) T_1 lifetimes plotted as a function of temperature, for $B \leqslant 2.5$ T. The lifetimes display a power-law dependence, as is evidenced by a straight line on a log-log scale graph. Best fits of the data with fully adjustable parameters all yield similar power exponents; the shown lines are best fits for a fixed exponent of −1.57: $T_1 \sim T^{-1.57}$.

well-understood.[34] This was also seen in the earlier work on the 3×10^{15} cm^{-3} sample.[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[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.[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 3×10^{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 5×10^{13} cm^{-3} electrons of Ref.

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 ~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 work in lightly doped GaAs by Dzhioev *et al.*, who found that the Dyakonov-Perel spin relaxation was suppressed under certain conditions for an unknown reason.[39]

As an alternate possible explanation, we reference the theory of Bulaev and Loss,[40] 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 ~5 T, with a width of ~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 $B \geqslant 4$ 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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