



Universal scheme for measuring the electron T_1 in semiconductors and application to a lightly-doped n -GaAs sample

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ABSTRACT

A new technique has been developed for measuring the T_1 spin lifetime of electrons, and should have near universal applicability among III–V semiconductors. The technique has been applied to a lightly doped GaAs sample with $n = 3 \times 10^{14} \text{ cm}^{-3}$. Spin decays were measured for fields from 0.5 to 7 T, at temperatures of 1.5 and 5 K. The spin lifetimes were shorter than expected, possibly due to compensation in the sample.

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1. Introduction

The possibility of spin-based quantum computing relies on the existence of materials in which there are long spin lifetimes, measured relative to the time of a potential computing operation [1]. Therefore it is critical to measure and understand spin lifetimes in potential quantum computing materials. In this paper we focus on measuring the T_1 , or longitudinal, spin lifetime. This is the time that describes how long it takes for a non-equilibrium population of electrons in Zeeman-split spin states to relax into their thermal equilibrium population. In general T_1 provides an upper bound on T_2 , the transverse spin lifetime and the most relevant lifetime for quantum computing, although in specific cases T_2 may exceed T_1 by up to a factor of two [2].

Various techniques have been used in the past to measure T_1 . In GaAs, a very important semiconductor for potential spintronic and quantum computing applications, these have included time-resolved optical measurements of the polarization of photoluminescence in n -type GaAs [3,4] and in p -type GaAs [5–7]; time-resolved optical measurements of the population of a spin-dependent state in n -type GaAs [8]; and time-resolved charge measurements (via charge-sensing quantum point contacts and a spin-to-charge conversion) of lithographically defined GaAs quantum dots [9].

In this paper we present a pump–probe technique for measuring T_1 which should be applicable to a large class of materials. The

requirements are two-fold. First, the electron spin states (called “spins” hereafter for simplicity) need to be able to be aligned through the standard optical orientation technique [10], whereby a circularly polarized laser causes one spin state to be preferentially populated. Second, the state of the spins needs to be measurable through the Kerr rotation of a probe laser [11]. The term “Kerr rotation” refers to the rotation of the plane of polarization of a linearly-polarized probe beam, in response to a spin polarization of electrons in the material. These requirements are not very limiting, and this technique should work on (for example) the vast majority of III–V semiconductors.

2. Material and methods

As a specific application of this technique, we have used it to study a lightly-doped n -type GaAs sample. The sample was chosen because its doping level of $n = 3 \times 10^{14} \text{ cm}^{-3}$ lies close to the geometric mean of two previously-studied samples [4,8], and we felt that this sample might shed light on the differences observed between the measured T_1 lifetimes in those previous samples. To review the findings of those papers, in Ref. [4] Colton et al. studied an $n = 1 \times 10^{15} \text{ cm}^{-3}$ sample and found a non-monotonic dependence of spin lifetime on field where the spin lifetime first increases, then decreases, then increases again as magnetic field is increased. The maximum lifetimes were around 19 μs , obtained at 1.5 K and $B > 3$ T. It was postulated that Dresselhaus- or Rashba-related effects could be causing an abrupt increase in spin relaxation at a particular field. In Ref. [8], Fu et al. studied an $n = 5 \times 10^{13} \text{ cm}^{-3}$ sample and found T_1 spin lifetimes to be in excess of 3 ms at 1.5 K and fields between 2 and 4 T. For higher fields the spin lifetime decreased steadily with field, in a $B^{-3.8}$ dependence. This is

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close to the expected B^{-5} dependence for spin relaxation in localized electrons in quantum dots [12,13], indicating that as expected electrons in lightly doped GaAs provide a good testing ground for the spin properties of electrons in quantum dots. No low field data were reported in those experiments.

Each of those two methods had severe limitations. The method employed in Ref. [4] could not be applied to samples with doping less than $1 \times 10^{15} \text{ cm}^{-3}$ because for very low doping levels the probe pulse used to stimulate the photoluminescence polarization would re-align the doped electrons' spin. The method employed in Ref. [8] had a maximum measurable lifetime of 3.5 ms, could not be applied to fields below 2 T, and required exceptionally small light levels so as to not shift the absorption line off-resonant with the excitation laser (due to nuclear polarization and the Overhauser effect). Therefore, a new, more broadly applicable method for measuring T_1 is desired: one which would apply to as many doping levels as possible, and one which would apply to as many magnetic fields as possible.

As mentioned, the sample chosen for this study was doped at $n = 3 \times 10^{14} \text{ cm}^{-3}$ in order to be compared to the 1×10^{15} and $5 \times 10^{13} \text{ cm}^{-3}$ samples in Refs. [4,8]. This specific sample has been the subject of other spin-related research [14,15], and has proven to be of high quality. The results obtained in the present study are somewhat surprising, however: the T_1 lifetimes increased steadily with field unlike both of the previous two samples, and the lifetimes themselves are much shorter than those seen in the previous two samples. These results are discussed below.

As with most previous T_1 -measuring techniques, our new technique is a two step pump–probe technique. The spins are first aligned with a circularly polarized pump pulse. The spins are later detected with a linearly polarized probe pulse. The Kerr rotation of the probe beam is measured. The degree of rotation depends on how long the probe pulse arrives after the pump pulse, and allows one to trace out the decay of the electron spin polarization with time.

For the data presented here, pulses in the probe beam were produced with an acousto-optic modulator whereas the pump beam (a fast diode laser) was modulated on/off via a direct input. The widths and relative delay of the pump and probe pulses were controlled by a two channel pulse generator. The pump beam (at 781 nm) was above the GaAs bandgap in energy; the probe beam (typically at 821 nm) was below. The probe beam was controlled with an external power stabilizer. The Kerr rotation was measured using a polarizing beam splitter and a balanced photodiode detector whose output is proportional to the difference in intensities of the split beams (to eliminate common mode noise). For the data presented below, the probe beam was set to 1.4 mW (unpulsed) and focused down on the sample to a spot size of 0.16 mm in diameter. The pump beam was also focused, to 0.19 mm diameter; care was taken to maintain a larger spot size than the probe beam so that all of the spins being probed were inside the pump beam spot. Fig. 1 displays an experimental schematic.

This technique bears many similarities with the well-known time resolved Kerr (or Faraday) rotation (TRKR) used by Awschalom and others to measure the inhomogeneous dephasing lifetime, T_2^* [11]. The major differences are the orientation of the pulses relative to the magnetic field (longitudinal in our case), and the electronic gating of the pump and probe pulses (as opposed to mechanical delay lines) which makes it possible to probe arbitrarily long time scales. This seems like a logical extension of the TRKR technique, but we are unaware of any groups to measure T_1 of III–V semiconductors in this fashion.

To separate out the effects of the pump pulse from sources of noise and to avoid dynamic nuclear polarization, we modulated the helicity of the pump laser. In the results presented below,

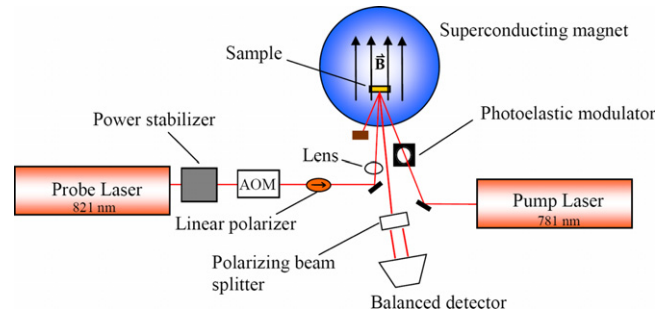


Fig. 1. (Color online) Simplified schematic of the experimental setup.

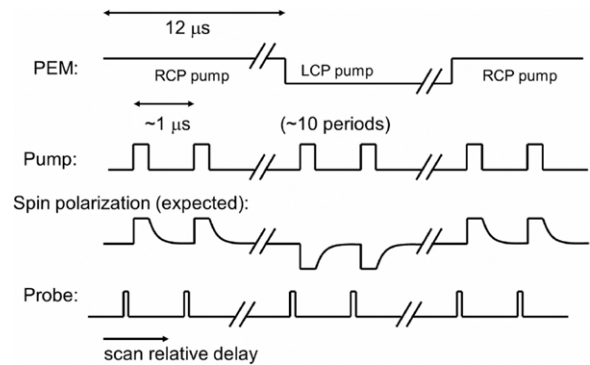


Fig. 2. Timing diagram of the experiment. The overall repetition period of pump and probe pulses (shown here are $1 \mu\text{s}$) was intentionally unrelated to the overall repetition period of the photo-elastic modulator ($24 \mu\text{s}$), but typically kept within 2% and 20% of it.

we did that with a 42 kHz photo-elastic modulator (PEM). In other experiments, not shown here, we have alternatively used an electro-optic modulator at 1 kHz (or slower) to achieve the same result. The overall pulse repetition cycle controlled by the pulse generator was typically *not* synchronized with the PEM frequency, although in some experiments we did so. Rather, the frequency of the overall pulse sequence was set independently such that the time of the pulse sequence was roughly two to twenty percent of the time of the PEM period. That is, we typically had five to fifty pulse sequences in each PEM period. Fig. 2 illustrates a typical timing sequence. Signal was detected with a lockin amplifier referenced to the PEM frequency.¹ As the delay between pump and probe pulses is varied, the detected signal traces out the spin polarization as a function of time. Fig. 3 gives a representative measured spin decay, for a field of 2 T and a temperature of 1.5 K. The spin polarization rises as the pump and probe pulses begin to overlap in time, then decays exponentially after the probe pulse starts to arrive after the pump pulse has completed. The points are raw data collected from the lockin amplifier; the curve is an exponential fit, whose decay time (equal to 359 ns for this specific case) is the T_1 spin lifetime.

3. Results and discussion

Data were taken at 1.5 and 5 K, for a number of different magnetic fields. The measured T_1 lifetimes are summarized in Fig. 4. At lower fields, the uncertainty in each data point was

¹ Other lockin schemes were also investigated. For example, we tried adding a second (or even third) lockin referenced either to the Agilent pulse generator or to a chopper in the pump or probe beam. However, our experience was that a single lockin amplifier referenced to the PEM, as described in the text, worked at least as well as any of the more complicated dual or triple lockin schemes.

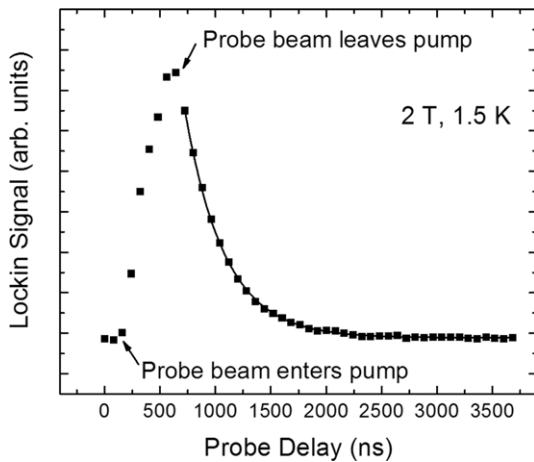


Fig. 3. Representative spin decay, at 2 T and 1.5 K. In this case the pump pulse was 320 ns and the probe pulse was 160 ns. The line is an exponential fit to the data, with a decay time of 359 ns.

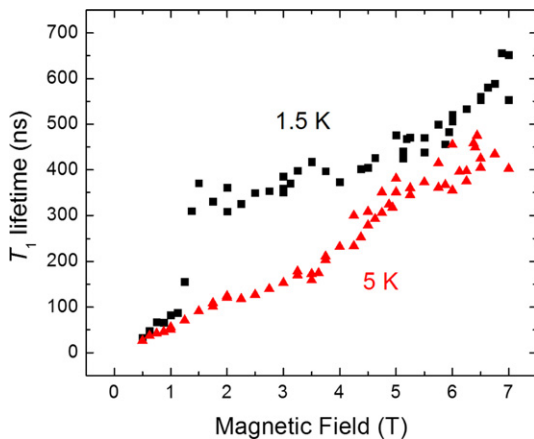


Fig. 4. (Color online) Summary of measured spin lifetimes, for 1.5 K (squares) and 5 K (triangles).

quite low—typically less than 10 ns. At the higher fields (larger than about 4 T), the decays started to have a linear baseline; incorporating this into the fit led to larger uncertainties as can be seen by the scatter. At the highest fields (~ 5.5 T and above), in addition to this linear baseline there was substantially increased noise. The linear term and increased noise could potentially be resulting from nuclear polarization, although care was taken to minimize this by modulating the pump beam (as mentioned above) and by blocking the light from the sample when not actively taking data. At the lowest fields (less than about 1 T), when the shortest pulses were employed, there was actually a phase reversal in our lockin signal as the probe pulse temporally moved through the pump pulse. This is unexplained.

As the data in Fig. 4 are examined, one of the most striking things is that the lifetimes are substantially shorter than those reported in both Refs. [4,8], instead of being between the two sets of measured lifetimes as would be expected from our doping level. It seems very unlikely that the doping level alone can explain our measured lifetimes.

We turn to another observation: the 1.5 K data shows a clear change in behavior around 1.5 T. Below that field, the lifetimes increase rapidly, and likely fall into the hyperfine-governed motional averaging situation described by D'yakonov and Perel' [16]. When $1/T_1$ is analyzed according to that theory, it follows a rough Lorentzian with a width of about 0.40–0.65 T, corresponding to a correlation time of approximately 41–66 ps. Similarly, the 5 K data

below about 2 T can be roughly fit with a Lorentzian to obtain a correlation time of 33–50 ps. These times are exactly in line with the correlation times measured in Ref. [4] for the $n = 1 \times 10^{15} \text{ cm}^{-3}$ sample for those two temperatures. That is somewhat surprising. Correlation between electrons arises through electron “hopping” between donor sites, and a similar correlation time implies a similar distance between donor impurities—as opposed to the factor of 1.5 farther apart that is implied through the doping level alone. That in turn implies this sample may have a substantial degree of compensation. Compensation is certainly possible; two other samples grown in the same growth chamber around the same time (one of them the sample studied in Ref. [4]) had compensation ratios of $N_A/N_D \approx 40\%–50\%$ [17]. With the smaller overall doping level of this sample the compensation ratio is likely to be much higher; given similar background impurities, we estimate around 80%.²

We postulate that both of these unexpected items (the short T_1 lifetimes and the fast correlation times) are related. That would imply that the random electric fields of charged donors and acceptors are the dominant source of spin relaxation in the regime from ~ 2 to 7 T. That type of spin relaxation has been seen in Mn-doped GaAs, where for example the presence or absence of compensation-related ionized impurities caused the T_2^* spin lifetimes to change by more than a factor of 10 [18]. Compensation-related spin relaxation has not been observed in non-magnetic GaAs, however, to the authors' knowledge. We additionally note that the lifetimes measured in this regime increase very monotonically with field, almost linearly. This may give theorists some clues as to how to model this type of spin relaxation. (Presumably if the field were increased further, the regime where spin lifetimes decrease with field would eventually be reached, as it was in Ref. [8].)

4. Conclusions

In summary, a new technique has been developed for measuring the T_1 spin lifetime of electrons, and has been applied to a lightly doped GaAs sample with $n = 3 \times 10^{14} \text{ cm}^{-3}$. Decays as fast as 26 ns and as long as 650 ns were measured, and in principle the technique can be extended to arbitrarily long lifetimes. The technique should be universally applicable to all semiconductors which allow optical orientation to pump the spins and Kerr rotation to probe the spins, which includes (at least) most of the III–V semiconductors.

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² One mystery that remains, is that given that the donors in this sample might be separated by similar distances compared to the donors in Ref. [4], why the T_2^* spin lifetime of this sample as measured through spin resonance would be shorter than that measured for the $n = 1 \times 10^{15} \text{ cm}^{-3}$ sample [14].

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