Millisecond spin-flip times of donor-bound electrons in GaAs

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We observe millisecond spin-flip relaxation times of donor-bound electrons in high-purity n-GaAs. This is three orders of magnitude larger than previously reported lifetimes in n-GaAs. Spin-flip times are measured as a function of magnetic field and exhibit a strong power-law dependence for fields greater than 4 T. This result is in qualitative agreement with previously reported theory and measurements of electrons in quantum dots.

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Since recent proposals for semiconductor spin-based quantum information processing (QIP), there has been a renewal of interest in the study of spin processes in semiconductors. The optical accessibility and the possibilities of integrated device fabrication in III-V semiconductors make electron spins in GaAs particularly promising candidates for QIP. Theoretical research predicts long (> ms) spin-flip lifetimes and long (µs - ms) decoherence times for electrons in GaAs quantum dots. In this work we measure millisecond spin-flip lifetimes (T1) of donor electrons in high-purity GaAs. This long relaxation is comparable to the longest reported value in quantum dots and is three orders of magnitude longer than lifetimes reported in n-GaAs samples with higher donor dopant concentrations.

Neutral donors (D0) in semiconductors are in many ways natural quantum dots. At low doping concentrations n < 1015 cm-3 and liquid helium temperatures, donor impurities are non-interacting and bind a single electron. This single electron is always present in contrast to the quantum dot case where the number of electron spins can be difficult to control. Just as with quantum dots, it is possible to excite a bound exciton, or electron-hole pair, at this site. The total bound exciton (D0X) complex is composed of two electrons in a spin-singlet state, one hole, and the donor impurity. Since each donor electron is in the same environment and has the same wavefunction (effective mass Bohr radius aB ≈ 100 Å), there is little inhomogeneity and bulk optical transition linewidths are as narrow as several GHz. This is more than three orders of magnitude narrower than quantum dots. The homogeneity in D0X systems makes them attractive candidates for QIP applications where identical or nearly identical emitters are necessary.

Extensive study has been made of D0-D0X transitions and we used these transitions to study T1 of the bound electron.

The sample consisted of a 10 μm GaAs layer on a 4 μm Al0.3Ga0.7As layer grown by molecular-beam epitaxy on a GaAs substrate. The sample had a donor concentration of ∼5×1013 cm-3. We mounted the sample strain-free in a magnetic cryostat in the Voigt (k ⊥ B) geometry.

An energy diagram of the D0 and D0X complexes in a magnetic field are shown in Fig. 1a. To measure T1 we optically pump and probe the D0 state using the A (σ-polarized) transition. We detect the photoluminescence from the A* (σ-polarized) transition. A complete energy diagram of the GaAs D0X states in a magnetic field can be found in Ref. 12.

![Energy level diagram of the D0X and D0 states at 0 T. In PLE experiments we probe the excited state populations via the two-electron satellite transitions.](image)

FIG. 1: (a) Energy level diagram of the D0X and D0 states at 0 T. In PLE experiments we probe the excited state populations via the two-electron satellite transitions. (b) Energy level diagram of the D0 1s state and the lowest D0X state in an applied magnetic field. To measure T1 we optically pump and probe the D0 state using the A (σ-polarized) transition. We detect the photoluminescence from the A* (σ-polarized) transition. A complete energy diagram of the GaAs D0X states in a magnetic field can be found in Ref. 12.
then probes the population in the $A^*$ transition as electrons were pumped into the other electron Zeeman state and not to a state outside the system. We perform single-laser and two-laser photoluminescence excitation (PLE) scans described in previous work\textsuperscript{13}. In the single laser PLE scan a continuous wave Ti:Sapphire laser is scanned over the $D^0X$ transitions as emission from the $|m_h = -\frac{1}{2}\rangle$ state to its two-electron satellites (TES) is detected (Fig. 1). In this case as the laser scans over each transition the photoluminescence is weak due to optical pumping. In a second scan a diode laser resonantly excites the $A^*$ transition during the Ti:Sapphire scan. The diode laser repumps the trapped electrons back into the $| - \frac{1}{2}\rangle$ state allowing efficient population of the $|m_h = -\frac{1}{2}\rangle$ state (Fig. 2). An eight-fold increase in the PLE intensity of the $A$-transition is observed. This result indicates that electrons are being pumped between the two Zeeman states and not to a state outside the system.

In the $T_1$ pump-probe measurement we resonantly excited the $A$ transition for a time $\tau_1 > 100 \mu s$ with a Ti:Sapphire laser. The intensities used in this experiment needed to be extremely low (1-10 mW/cm$^2$) to minimize the dynamic polarization of the nuclear spins within the donor electron wavefunction\textsuperscript{14}. If larger powers were used a diamagnetic shift in the energy of the $A$ line was observed of up to 10 GHz, shifting the line off of resonance with the excitation laser. It is possible to estimate the nuclear field using a phenomenological relation describing the diamagnetic shift of the GaAs $D^0X$ in a magnetic field\textsuperscript{12}. At 9 T this shift corresponds to a modest nuclear field of $B_{nuc} = 0.06$ T and 1% nuclear polarization.

During the pump down phase we monitored the excited state population by spectrally filtering the $A^*$-transition. Additionally, we reduced the collection of the scattered pump light by collecting the opposite polarization. We observed the time-resolved decay in the PL from the $A^*$ transition as electrons were pumped into the $| + \frac{1}{2}\rangle$ state (Fig. 2). The polarization of the $A$ ($\pi$-polarized) and $A^*$ ($\sigma$-polarized) transitions indicate that the resonant $D^0X$ state corresponds to a hole-spin of $m_h = -\frac{1}{2}$. This assignment is consistent with previous spectroscopic work with crystal axis (110) parallel to the magnetic field\textsuperscript{12}. In the spherical approximation the corresponding branching ratio is 1:2 to the electron Zeeman states $| + \frac{1}{2}\rangle; | - \frac{1}{2}\rangle$. Although spherical symmetry is reduced to tetrahedral symmetry by the crystal, we still observe strong PL from both of these transitions (Fig. 3). With the fast (ns) radiative relaxation of the $D^0X$ state and the long ($> 100 \mu s$) measured $T_1$ times, the intensity ratio of the optically pumped to the recovered populations should be close to 0. However the observed PL ratio never exceeded 1/8. This ratio increases at lower field and indicates that the effective $T_1$ is shorter when the optical pumping field is on compared to $T_1$ in the dark.

After optical pumping, the excitation laser was then deflected with an acoustic optic modulator (AOM) for a time $\tau_{\text{wait}}$. Next, a short pulse ($0.5 - 2 \mu s$) resonant on the $A$ transition probed the sample and light was de-
tected from the $A^*$ transition. We were able to perform the experiment on several different $D^0$-$D^0X$ transitions at high fields ($>5$ T). However at low field it was only possible to cleanly isolate the lowest energy $A$ transition and all data presented are from this transition.

The magnetic field dependence of the electron spin relaxation is shown in Fig. 4. At fields less than 4 T there appears to be a levelling off of $T_1$ at several ms. This is related to the finite extinction ratio of the AOM ($\approx 1000$) and a small leakage field even when the beam is deflected away from the sample. Thus, for fields less than 4 T our data only gives a lower bound on $T_1$. As the magnetic field is increased, $T_1$ decreases rapidly exhibiting a strong power-law dependence. The $T_1$ dependence on B-field observed is very different from previously reported $T_1$ measurements in $n$-GaAs by Colton et al.\(^\text{[15]}\) Previously $T_1$ was measured in higher doped samples ($n = 3 \times 10^{15}$ cm$^{-3}$) by time-resolved polarization photoluminescence measurements on free excitons. $T_1$ was shown to increase with magnetic field reaching a maximum of 1.4 $\mu$s at the maximum attainable field of 5 T. This different dependence may be due to the larger doping density resulting in a greater interaction between neighboring donor-bound electron spins. A further study on the effect of doping density on $T_1$ is necessary to understand the discrepancy between the two experimental results.

Theoretical calculations of the B-field dependence of $T_1$ due to the modulation of electron spin-orbit coupling by phonons predict at $B^{-4}$ dependence of $T_1$ for neutral donors.\(^\text{[5]}\) A fit to data for $B > 5$ T (Fig. 4) shows a strong power law dependence with an exponent of $m = -3.8 \pm 0.2$. This indicates that the one-phonon spin-orbit relaxation process is the dominant process for GaAs neutral donors at high magnetic fields, $\mu_B B > k_B T$. In our system the temperature $T$ is 1.5 K, the electron g-factor is 0.41\(^\text{[13]}\) and the low-field to high-field transition occurs at $B = 5.6$ T. Our results are similar to previously reported quantum dot electron-spin relaxation times. In GaInAs quantum dots, Kroutvar et al.\(^\text{[14]}\) found an inverse power-law dependence of $T_1$ on magnetic field. The observed power law exponent, $m = -5$, was theoretically predicted for disk-like quantum dots\(^\text{[15]}\) for single-phonon spin-orbit relaxation processes. The similarity between the donor electron and quantum dot systems indicates that in our sample electrons are extremely well-isolated and non-interacting.

In conclusion, we have observed long, millisecond spin-flip times of electrons bound to donors in bulk GaAs. This direct pump-probe measurement is possible due to the homogeneity of the $D^0$ and $D^0X$ systems in high-purity GaAs. Our result represents a three-order of magnitude increase over previously measured $D^0$ electron $T_1$ times. Both the long $T_1$'s observed and the strong power law dependence of $T_1$ on magnetic field indicate that donor-bound electrons are non-interacting in sufficiently pure GaAs. Thus these systems are an attractive testing ground to study electron spin dynamics for quantum information processes.

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