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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),[1–3] 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[4,5] and long (μs-ms) decoherence times[6,7] for electrons in GaAs quantum dots. In this work we measure millisecond spin-flip lifetimes (T_1) of donor electrons in high-purity GaAs. This long relaxation is comparable to the longest reported value in quantum dots[8] and is three orders of magnitude longer than lifetimes reported in *n*-GaAs samples with higher donor dopant concentrations.[9]

Neutral donors (D^0) in semiconductors are in many ways natural quantum dots. At low doping concentrations $n \leq 10^{15}$ cm^3 and liquid helium temperatures, donor impurities are noninteracting 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 (D^0X) 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 wave function (effective mass Bohr radius $a_B \approx 100$ 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 D^0X systems makes them attractive candidates for QIP applications where identical or nearly identical emitters are necessary.[10,11] Extensive study has been made of D^0-D^0X transitions[12] and we used these transitions to study T_1 of the bound electron.

The sample consisted of a 10 μm GaAs layer on a 4 μm $Al_{0.3}Ga_{0.7}As$ layer grown by molecular-beam epitaxy on a GaAs substrate. The sample had a donor concentration of $\sim 5\times 10^{13}$ cm^{-3}. We mounted the sample strain-free in a magnetic cryostat in the Voigt ($\vec{k}\perp\vec{B}$) geometry.

An energy diagram of the D^0 and D^0X complexes in a magnetic field are shown in Fig. 1(b). To measure T_1 of the donor electrons a long pulse is applied to the *A* transition. The system is excited to the $|m_h=-\frac{1}{2}\rangle$ bound exciton state where it then decays to both electron spin states. After many cycles, this pumps electrons from the $|-\frac{1}{2}\rangle$ state to the $|+\frac{1}{2}\rangle$ state. After this pulse, the system evolves in the dark for a variable time τ_{wait}. During this time the electron population returns to thermal equilibrium and the $|-\frac{1}{2}\rangle$ state is repopulated with a characteristic time T_1. A short pulse resonant on the *A* transition then probes the population in the $|-\frac{1}{2}\rangle$ state and the intensity of the A^* transition is measured. The experiment is then repeated with different time delays τ_{wait}. An

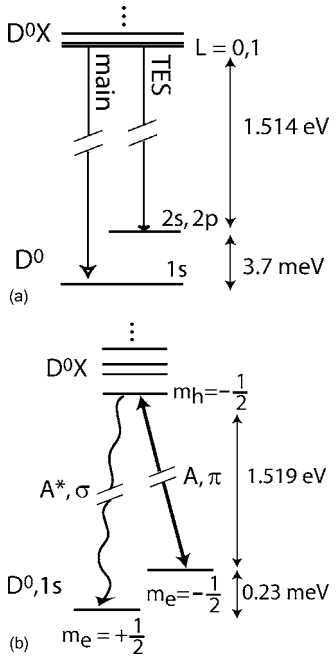

FIG. 1. (a) Energy level diagram of the D^0X and D^0 states at 0 T. In PLE experiments we probe the excited state populations via the two-electron satellite transitions. (b) Energy level diagram of the D^0 1*s* state and the lowest D^0X state in an applied magnetic field. To measure T_1 we optically pump and probe the D^0 state using the *A* (π-polarized) transition. We detect the photoluminescence from the A^* (σ-polarized) transition. A complete energy diagram of the GaAs D^0X states in a magnetic field can be found in Ref. 12.

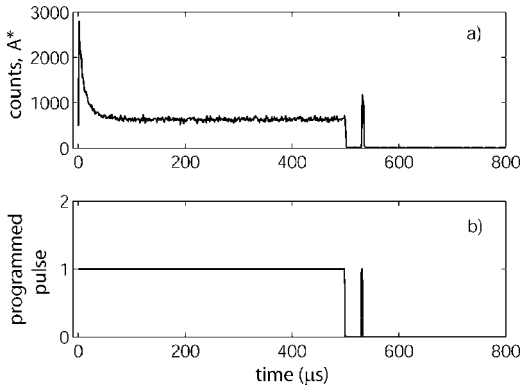

FIG. 2. (a) Typical data obtained during the pump-probe experiment. Time-resolved photoluminescence from the A^* line is collected at 9.9 T. Optical pumping can be observed during the first 20 μs of the pump pulse. Partial recovery is observed after the 30 μs delay. (b) Pump-probe programmed intensities.

example pulse sequence and photoluminescence (PL) data are shown in Fig. 2.

In order to ensure that population is indeed being pumped into the other electron Zeeman state and not to some state outside our system, we perform single-laser and two-laser photoluminescence excitation (PLE) scans described in previous work.[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(a)]. 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. 3). An eightfold 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 $\tau_1 > 100$ μs 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 wave function.[14] If larger powers were used a diagmagnetic 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.[12] At 9 T this shift corresponds to a modest nuclear field of $B_{\text{nuc}}=0.06$ 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 (π-polarized) and A^* (σ-polarized) transitions indicate that the resonant D^0X state corresponds to a hole-

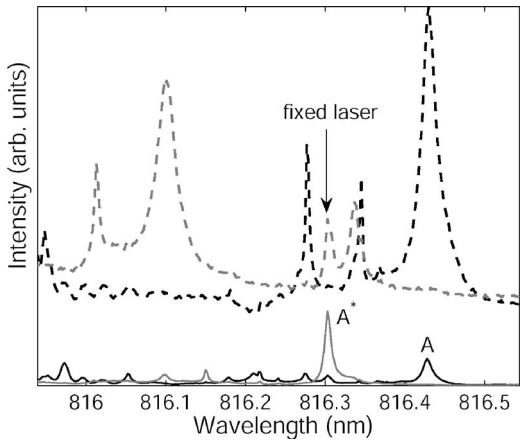

FIG. 3. Single laser PLE scans (solid lines) and double laser PLE scans (dashed lines) at 9.9 T. In the single laser PLE scans, a π-polarized (black) and σ-polarized (gray) laser is scanned over the D^0X transitions. Photoluminescence (PL) is collected from an $|m_h=-\frac{1}{2}\rangle$ transition and is proportional to population in the D^0X A state. Overall intensity is weak due to optical pumping. In the two laser PLE scans, a second laser is fixed resonantly on the A^* transition as the first laser scans over the D^0X transitions. A large enhancement in PL intensity is observed for transitions originating from the $|-\frac{1}{2}\rangle$ state.

spin of $m_h=-\frac{1}{2}$. This assignment is consistent with previous spectroscopic work with crystal axis $\langle 110\rangle$ parallel to the magnetic field.[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 symmetry, 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 μs) 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 τ_{wait}. Next, a short pulse ($0.5-2$ μs) resonant on the A transition probed the sample and light was detected 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 (≈ 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 give 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 the B field observed is very different from previously reported T_1 measurements in n-GaAs by

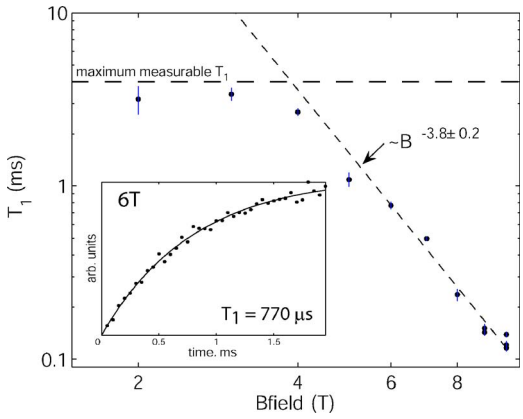

FIG. 4. (Color online) Log-log plot of the bound electron spin lifetime T_1 dependence on B field. A strong power-law dependence on B-field is observed for $B>5$ T. Recovery times measured at $B \leq 4$ T are limited by the experimental apparatus and are only a lower bound on the electron T_1. Inset: Example exponential fit of the pump-probe data at 6 T.

Colton *et al.*[9] Previously T_1 was measured in higher doped samples ($n=3\times10^{15}$ 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 μs 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.[15] A fit to data for $B>5$ T (Fig. 4) shows a strong power-law dependence with an exponent of $m=-3.8\pm0.2$. This indicates that the one-phonon spin-orbit relaxation process is the dominant process for GaAs neutral donors at high magnetic fields, $g\mu_B B > k_B T$. In our system the temperature T is 1.5 K, electron g-factor is 0.41 (Ref. 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.*[8] found an inverse power-law dependence of T_1 on magnetic field. The observed power-law exponent, $m=-5$, was theoretically predicted for disklike quantum dots[4,5] for single-phonon spin-orbit relaxation processes. The similarity between the donor electron and quantum dot systems indicate that in our sample electrons are extremely well-isolated and noninteracting.

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 noninteracting 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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