Coherent population trapping of electron spins in a high-purity n-type GaAs Semiconductor

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In high-purity n-type GaAs under strong magnetic field, we are able to isolate a lambda system composed of two Zeeman states of neutral-donor bound electrons and the lowest Zeeman state of bound excitons. When the two-photon detuning of this system is zero, we observe a pronounced dip in the excited-state photoluminescence indicating the creation of the coherent population-trapped state. Our data are consistent with a steady-state three-level density-matrix model. The observation of coherent population trapping in GaAs indicates that this and similar semiconductor systems could be used for various EIT-type experiments.

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In the past decade great steps have been made toward the coherent control of light using techniques based on electromagnetically induced transparency (EIT) [1]. Light has been slowed by seven orders of magnitude [2], stored and released on command [3, 4, 5], and coherently manipulated while stored in atomic gases [16, 17]. In a three-level Λ-system, a probe field couples states |1⟩ and |3⟩, and the creation of large optical non-linearities [12] for photonic gates in non-linear quantum computation [11], and the creation of large optical non-linearities [12] for photonic gates in non-linear quantum computation [11].

EIT is based on the effect of coherent population trapping which was first observed in the 1970’s in atomic gases [16, 17]. In a three-level Λ-system, a probe field with Rabi frequency Ωp couples states |1⟩ and |3⟩, and a coupling field with Rabi frequency Ωc couples states |2⟩ and |3⟩ (Fig. 1b). Optical pumping leads to a coherent superposition of states |1⟩ and |2⟩ that is decoupled from |3⟩ due to a quantum interference between the two transitions. The crucial condition for coherent population trapping is that the decoherence rate γ12 between states |1⟩ and |2⟩ is slow compared to the radiative decay rate of |3⟩. Furthermore, for photon-storage applications, γ12 determines how long quantum information can be stored [2].

Long decoherence times, which naturally arise in atomic systems [3], are also possible in solids [18, 19, 20]. EIT has been observed in rare-earth doped insulators [3], N-V centers in diamond [18], and in the transient optical response of GaAs quantum wells [21]. Here, we consider electron spins bound to neutral donors (D0) in a semiconductor, a system that could offer some unique advantages. For example, the optical transitions to the donor-bound exciton states feature a small inhomogeneous broadening (2 GHz) combined with a large oscillator strength (1 ns radiative lifetime 22). Furthermore, the ground state is long-lived, unlike the exciton states used in previous semiconductor EIT experiments [21]. Finally, donor impurities can easily be integrated into monolithic microcavities. In this letter, we report the observation of coherent population trapping in an ensemble of D0 spins, demonstrating that a lambda system can be optically addressed and manipulated. While the degree of ground-state coherence currently obtainable is small, it is thought to be limited mainly by inhomogeneous broadening of the electron-Zeeman splitting, which can hopefully be remedied in pulsed experiments with spin-echo techniques.

The energy level structure of a neutral donor is shown in Fig. 1b. Due to the small electron effective mass and high dielectric constant of GaAs, the wavefunction of a neutral-donor bound electron (D0) extends over many lattice sites and is well described by the hydrogenic wavefunction with a 100 Å Bohr radius [22]. With an applied magnetic field, the 1s state splits into the two electron-Zeeman spin states which are labelled |1⟩ and |2⟩ in Fig. 1b. The excited states consist of an electron-hole pair, or exciton, bound to the D0 center. This donor-bound-exciton complex (D0X), consisting of two electrons in a spin-singlet state, a hole with quasi-spin-3/2, and the donor impurity, can be resonantly excited from the D0 state. At zero magnetic field, the D0X is composed of closely spaced orbital angular momentum states (Fig. 1b). In a magnetic field each of the D0X states splits into the four hole-Zeeman spin states. In Fig. 1b we identify the lowest-energy D0X states as A and A1 following Ref. [15]. We denote transitions to the D0 state |me = −1/2⟩ with a label only (e.g. A) and transitions to the state |me = 1/2⟩ with an asterisk (e.g. A∗). Although the D0X predominately relaxes to the D0 1s state, there
Detector the excited state population using the TES fluorescence. In photoluminescence excitation (PLE) scans, a decrease in the excited-state population when two-laser excitation detuned from resonance, and two-laser excitation on two-photon resonance. With the probe laser only, the PLE spectrum gives a linewidth of only 2 GHz. The data fit a Lorentzian lineshape extremely well and indicate that there is little inhomogeneous broadening. The emission intensity is weak due to optical pumping of most of the electron population into state \( \text{2s, 2p} \), which for the probe laser only, is a dark state. With both lasers exciting the sample but detuned from two-photon resonance, the emission becomes much stronger since in this case there is no dark state. When the probe and coupling lasers are brought into two-photon resonance, a pronounced and narrow reduction of the emission intensity is observed as a new dark state is formed which is a coherent superposition of states \( |1\rangle, |2\rangle \).

The decrease in the excited-state population observed on two-photon resonance is incomplete because of decoherence and population relaxation between levels \( |1\rangle \) and \( |2\rangle \). The results can be understood in terms of a 3-level system interacting with a reservoir, described by the density-matrix master equation:

\[
\frac{\partial}{\partial t} \rho = \frac{1}{\hbar} [H, \rho] - \mathcal{L}(\rho) = 0
\]

in which \( H \) is the Hamiltonian of the system and \( \mathcal{L}(\rho) \) is the Liouville operator describing the decoherence processes. In the interaction picture and rotating wave ap-
The only parameter that must be changed to fit simultaneously both the single laser (a) and two laser (b) scans is the level $|3\rangle$ dephasing rate. The fit indicates slow ($\mu$s) electron population relaxation rates and fast (1-2 ns) electron decoherence rates in our system. Thus, the system exhibits a lower level dephasing rate on the same order as the excited state radiative lifetime (1 ns). From this fit we can also obtain the ratio of the two-state coherence, $\rho_{12}$, to the ideal case, $\rho_{12,\text{ideal}} = \frac{\Omega_p}{\Omega_c} \frac{\Omega_c}{\Omega_p} + \frac{\Omega_c}{\Omega_p}$, and find that $\rho_{12}/\rho_{12,\text{ideal}} = 0.23$. In the weak probe limit ($\Omega_p << \Omega_c, \Gamma_{31}, \Gamma_{32}$) $\rho_{12}$ reduces to

$$\rho_{12} \approx \frac{\Omega_p}{4\gamma_{13}\gamma_{12}} + \frac{\Omega_c}{\gamma_{12}}$$

in which $\gamma_{13}$ ($\gamma_{12}$) is the total decay rate for $\rho_{13}$ ($\rho_{12}$) given in Eq. (1). From this relation it is evident that the coherence of this system is currently limited by the short lower-level decoherence time as well as additional dephasing of state $|3\rangle$.

Additional measurements to verify the theoretically expected behavior are shown in Fig. 4. In Fig. 4b, PLE scans were performed at several coupling intensities. As the coupling intensity increases, the population-trapped window at zero two-photon detuning becomes relatively wider and deeper as expected. The data fit our theoretical model if the lower-level population-relaxation rate is allowed to increase with increased coupling field intensity. This increase could be due to sample heating at large coupling-laser powers. In our sample, the GaAs substrate was not removed and absorbs all of the incident radiation. If we assume a one-phonon spin-orbit relaxation process $[21, 22]$, we can be simultaneously fit the coupling power dependence series by varying only the sample temperature from 1.5 K to 6 K. In a second experiment (Fig. 4b), the two lasers are tuned to different excited states and the PLE dip is not observed. In this case, the probe laser is tuned to the $A_1$ transition and the coupling laser is tuned to the A transition (see Fig. 4b). As in the previous case, if only the probe laser is applied, population becomes depleted from state $|1\rangle$ and the PLE intensity is weak. The coupling laser repopulates this state and the PLE intensity is enhanced. The absence of the dip in this experiment as well as the narrow dip in the lambda system (FWHM $\ll$ homogeneous broad-
been proposed that further improvements on storage time relaxation time, on the order of microseconds. It has also not been measured but could be close to the population coherence time, $T_2^*$. This indicates that $T_2$ of our sample up to 6 K, although the overall PLE intensity in the two laser case is observed without a dip at zero detuning.

We only observe a modest suppression of the excited-state population. This is due to the 1-2 ns inhomogeneous decoherence time, $T_2$. At the extremely low densities ($\sim 5 \times 10^{13}$ cm$^{-3}$) in our sample, the nuclear-electron hyperfine interaction becomes very efficient. At these densities, the donor electrons are well localized and do not interact with each other. At 2 K the nuclei are essentially unpolarized and theoretical calculations predict nanosecond $T_2^*$ due to the random nuclear states. Experimentally, a 5 ns $T_2^*$ has been measured in n-type GaAs with $n \sim 3 \times 10^{14}$ cm$^{-3}$ via optically detected electron-spin resonance. This result is consistent with our value given our sample’s lower donor density. Additionally, we find that if we increase the temperature of our sample up to 6 K, although the overall PLE intensity decreases dramatically, the width of the dip does not change significantly. This indicates that $T_2^*$ in our system is not temperature dependent and is consistent with the nuclear-electron hyperfine decoherence model.

Although the inhomogeneous $T_2$ limits the depth of the population-trapped dip, in an EIT-type experiment with pulsed lasers and electron spin-echo techniques, the storage time should be limited by the homogeneous decoherence time, $T_2$. $T_2$ of electron spins in GaAs has not been measured but could be close to the population relaxation time, on the order of microseconds. It has also been proposed that further improvements on storage time could be made by transferring the electron spin coherence to the nuclear spins. If this is achieved, a storage time on the order of seconds may be feasible.

In summary, we have observed coherent population trapping of donor-bound electrons in GaAs. To our knowledge, this is the first demonstration of a lambda system in a semiconductor that utilizes the true electron ground states. In addition, due to the substitutional nature of donor impurities and high crystal quality, this system has little inhomogeneous broadening in the optical transitions. Although current population trapping is limited by a short $T_2^*$, there exist several possible ways to engineer this system for long $T_2$ and storage times. Spin echo techniques and electron to nuclear information transfer should be able to extend possible storage times by orders of magnitude in GaAs. Additionally, the $D^0$X system exists in every semiconductor. Thus, a crystal composed of nuclear spin-0 elements would significantly extend the storage lifetime. We also note that larger bandgap semiconductors have larger effective masses, larger $D^0$ binding energies, and thus larger $D^0$X binding energies, which allow higher temperature operation.

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FIG. 4: (a) PLE scans with varying coupling field intensities. $I \sim 1$ W/cm$^2$. The dip becomes wider and deeper relative to the wings of the curve as the coupling field is increased. The only fitting parameter varied between the two laser scans is $T$ ($\Gamma_{12} \propto (e^{-A_1} - 1)^{-1}$). In order of increasing coupling intensity, $T = 1.5, 1.5, 2, 3.1, 4.6, 6.0$ K. (b) Incoherent optical pumping experiment with (i) coupling laser off, (ii) coupling laser on. The coupling laser is tuned to transition $A$. The probe laser is scanned over $\Delta E$ (see Fig. 1c). An enhancement of the PLE intensity in the two laser case is observed without a dip at zero detuning.

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