Coherent spin dynamics of donor bound electrons in GaAs

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(Received 2 June 2011; revised manuscript received 29 July 2011; published 24 August 2011)

We report experimental studies of coherent spin dynamics of donor-bound electrons in high-purity GaAs by using transient differential transmission. The donor-bound exciton transitions, which are not visible in the linear absorption spectrum, are spectrally resolved in the nonlinear differential transmission spectra. The spin beats in the transient differential transmission response, arising from electron spin precession in an external magnetic field, are investigated with the pump and probe coupling to various donor-bound exciton transitions. The spectral dependence of the spin beats provides important information on the polarization selection rule for the underlying donor-bound exciton transitions. The polarization selection rules deduced from these experiments indicate that contributions from higher-energy donor-bound exciton transitions can severely limit the effectiveness of optical spin control using mechanisms such as polarization-dependent optical Stark shifts.

DOI: 10.1103/PhysRevB.84.085205 PACS number(s): 71.35.−y, 78.55.Cr, 78.67.−n, 42.50.Md

I. INTRODUCTION

Electrons bound to neutral donors in bulk semiconductors are an interesting and promising spin system due to their long spin coherence times and atomic-like optical transition linewidths. Electron spin decoherence times as long as several microseconds have been observed in a recent spin echo study in GaAs.[1] Donor-bound electrons are in similar local environments, leading to relatively small inhomogeneous linewidths. Emission linewidths less than 0.1 meV can be routinely observed for donor-bound exciton (D^0X) transitions in high-purity GaAs.[2–4] In many respects, neutral donors in semiconductors resemble quantum dots (QDs), but without the complication of large inhomogeneous broadening common to epitaxially grown QD systems. The robust spin coherence and the atomic-like D^0X transitions in GaAs have enabled the experimental realization of coherent optical phenomena, such as coherent population trapping and electromagnetically induced transparency associated with donor-bound electrons.[4,5]

Optical spin control of donor-bound electrons has also been realized with the use of single off-resonant ultrafast laser pulses,[6] although only a limited degree of spin rotation ($\pi/3$ or less) has been achieved in spite of the spectrally sharp D^0X transitions. In comparison, ultrafast optical spin rotations exceeding multiples of π have been successfully demonstrated for single as well as ensemble electron spins in QDs[7–9] and for ensemble electron spins in modulation-doped quantum wells (QWs).[10] The ultrafast optical spin control is based on a polarization-dependent optical Stark shift, for which an off-resonant laser pulse induces an electron spin splitting via the optical Stark shift.[7] Quantum control of electron spins is essential for applications such as spin-based quantum information processing. It is thus important to understand the physical mechanisms that limit the optical spin control of donor-bound electrons.

In this paper, we report experimental studies of coherent spin dynamics of donor-bound electrons in high-purity GaAs using transient differential transmission (DT). We show that transient DT responses arising from nearby D^0X transitions can be spectrally resolved. Spin beats in the transient DT response due to different D^0X transitions feature distinct oscillatory behaviors, providing information on the polarization selection rules for the underlying optical transitions. Our studies indicate that contributions from higher-energy D^0X transitions can limit the effectiveness of coherent optical spin control using mechanisms such as polarization-dependent optical Stark shifts. In this regard, the energy level structure of D^0X transitions in GaAs poses a unique challenge for realizing effective optical spin control.

II. SAMPLES AND EXPERIMENTAL SETUP

Our experimental studies were carried out in high-purity bulk GaAs of 10 μm thickness, grown by molecular beam epitaxy on a semi-insulating GaAs substrate. The substrate was removed by wet chemical etching for transmission measurements. Two special sample holders were constructed, in which the thin GaAs layer remains freestanding. The experimental results presented here were obtained with a sample holder machined from brass. A second sample holder was constructed from pieces of a GaAs wafer, such that the holder and the sample have the same thermal expansion coefficient. Similar experimental results were obtained from both holders. The sample holder was mounted in an exchange gas cryostat with an external magnetic field ($B = 0.45$ T) applied in the sample plane (Voigt configuration). All experiments were carried out at 1.5 K, unless otherwise specified.

DT measurements were performed in a pump-probe setup, in which we measured the change in the probe transmission induced by a pump beam with a lock-in amplifier, as shown schematically in Fig. 1. Both the pump and probe laser pulses were derived from a picosecond mode-locked Ti:Sapphire laser with a repetition rate of 80 MHz. A spectral pulse shaper was used to narrow the spectral linewidth of the pump pulse to 0.04 nm (with a duration of 25 ps), such that the pump pulse can couple to a specific D^0X transition (note that the spectral linewidth is still large compared with the electron Zeeman spin splitting). An average pump power of 0.1 mW corresponds approximately to a peak pulse intensity of 800 W/cm^2. A weak

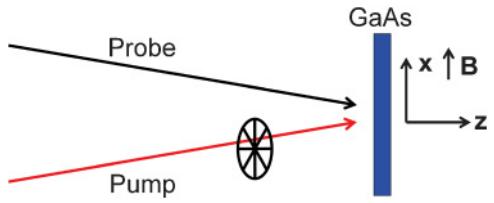

FIG. 1. (Color online) The configuration and geometry for the differential transmission experiment.

and spectrally broad probe pulse was used. The probe pulse was spectrally resolved in a spectrometer after its propagation through the sample. The transient DT response was obtained at a fixed probe wavelength as a function of the delay between the pump and probe pulses. The spectral DT response was obtained at a fixed pump-probe delay as a function of the wavelength of the probe. Both the pump and probe had the same circular polarization.

For the DT experiment, a circularly polarized pump pulse excites a spin-polarized electron and hole pair, which can form a D^0X by binding to a donor-bound electron. Since the two electrons involved have the opposite spin, the ensemble of the donor electrons acquires a net spin polarization after the optical excitation. The rapid spin relaxation of the holes ensures that the recombination of the D^0X does not significantly affect the electron spin polarization.[11,12] The pump-induced electron spin polarization precesses around the external magnetic field, leading to periodic oscillations or spin beats in the DT response.

Figure 2(a) shows the photoluminescence (PL) spectrum of the GaAs sample excited by a 532-nm continuous wave laser. The PL spectrum features a series of sharp emission resonances corresponding to D^0X transitions near 819.5 nm with a spectral linewidth as narrow as 0.06 nm (which is in part limited by the spectrometer resolution). The D^0X resonances, as well as those associated with the ionized donor-bound exciton (D^+X), acceptor-bound exciton (A^0X), and free excitons (X), are readily identified by comparison with previous experimental results.[2,3] Although only three D^0X emission resonances are individually resolved in the data shown, a fourth is known to overlap with the lowest energy peak. We label these peaks from lowest to highest energy with

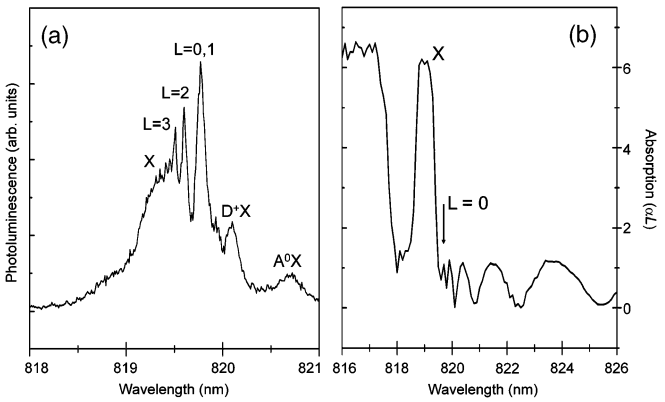

FIG. 2. (a) Photoluminescence spectrum of the bulk GaAs sample at 4 K. (b) Absorption spectrum with $L = 0$ D^0X spectral position indicated by arrow.

the notation $L = 0$, 1, 2, and 3, in accordance with Ref. 3. The $L = 0$ and $L = 1$ resonances, which play a major role in optical spin control, are the lowest-energy transitions and are furthest from the free exciton peak. As will be discussed further below, the $L = 3$ resonance also plays a significant, but detrimental, role in the optical spin control. Note that $L = 0$ and $L = 1$ transitions are nearly degenerate. For brevity, we refer to both transitions as the $L = 0$ transition.

Figure 2(b) shows the absorption spectrum of the GaAs sample. The D^0X absorption resonances are not visible in the absorption spectrum due to the high purity of the sample and also due to Fabry–Perot interference fringes arising from multiple reflections from the sample surface. However, the $L = 0$ D^0X resonance, which is indicated by the arrow in Fig. 2(b), can be identified in DT spectra, as will be described shortly. As shown in Fig. 2(b), the low-energy tail of the strong free exciton absorption overlaps with the D^0X resonances. For this reason, care was taken to minimize the spectral broadening due to inhomogeneous sample strain. Nevertheless, it is difficult to excite the D^0X transitions without significant free exciton excitation. Note that while the sample is freestanding, it is not strain free. Comparison of PL spectra between etched and unetched samples shows that the residual strain red-shifts the D^0X transitions by approximately 1 nm.

III. EXPERIMENTAL RESULTS

The D^0X transitions, while not visible in the linear absorption spectrum, can be clearly identified in DT spectra. Figure 3(a) shows the DT spectra obtained at two fixed delays between the pump and probe. For this experiment, the pump pulse is resonant with the $L = 0$ D^0X transition. Upon arrival of the pump pulse, two pronounced and spectrally sharp resonances emerge in the DT spectra at 819.85 nm and 819.6 nm, which we attribute to the $L = 0$ and $L = 3$ transitions, respectively. The resonances arise from bleaching as well as a small spectral broadening of the underlying D^0X transitions. The $L = 0$ resonance is 0.65 nm (1.1 meV) below

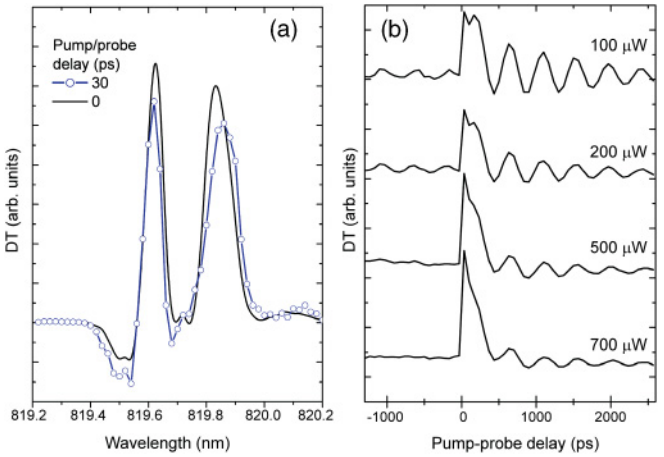

FIG. 3. (Color online) (a) DT spectra taken at two different pump-probe delays as indicated in the figure. Pump pulse with an average power of 100 μW is resonant with $L = 0$ D^0X transition. (b) Transient DT responses with both the pump and probe at $L = 0$ transition.

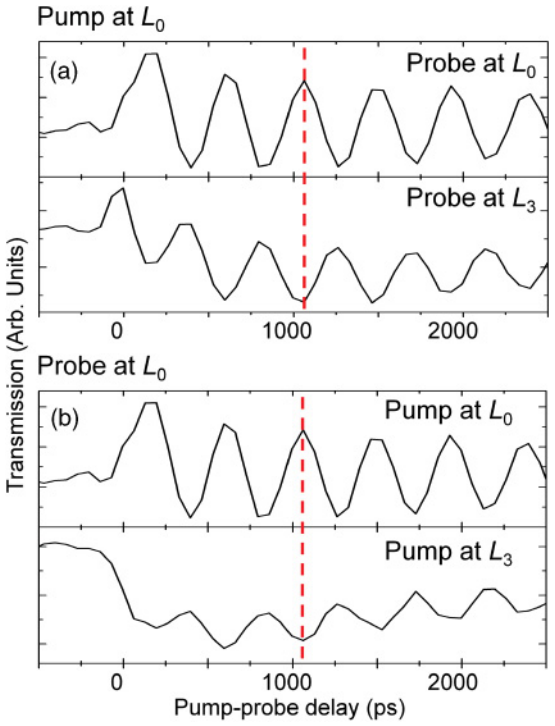

FIG. 4. (Color online) Comparison of spin beats observed at $L = 0$ and $L = 3$ D^0X transitions, with an average pump power of 100 μW. (a) Pump at the $L = 0$, and probe at $L = 0$ (top) and $L = 3$ (bottom) transitions. (b) Probe at $L = 0$ and pump at $L = 0$ (top) and $L = 3$ (bottom) transitions. Dashed lines are a guide to the eye.

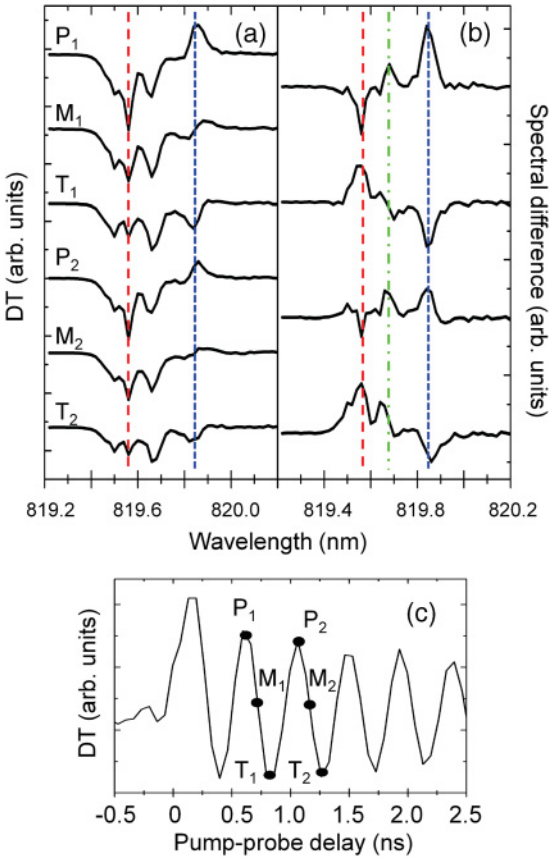

FIG. 5. (Color online) (a) DT spectra obtained at delay times defined in (c). (b) Difference of the DT spectra. From top to bottom, S(P_1)-S(M_1), S(T_1)-S(M_1), S(P_2)-S(M_2), and S(T_2)-S(M_2) are plotted, with S(t) denoting the DT spectrum obtained at delay time t. For both (a) and (b), blue short dashed and red long dashed lines indicate the spectral positions for the $L = 0$ and $L = 3$ transitions, respectively. Spectral position of the $L = 2$ D^0X transition is labeled by the green dashed-dotted line in (b). (c) Transient DT response with dots labeling delay times, at which DT spectra were measured in (a). The delay times are $P_1 = 600$ ps, $M_1 = 710$ ps, $T_1 = 830$ ps, $P_2 = 1040$ ps, $M_2 = 1150$ ps, and $T_2 = 1270$ ps.

the free exciton absorption resonance. The assignment of the resonance at higher energy as the $L = 3$ transition, rather than the $L = 2$ transition, is based on its spectral separation (0.25 nm) from the $L = 0$ transition. Note that the $L = 2$ resonance does not seem to appear in this measurement. As will be discussed later, the $L = 2$ resonance makes an appearance in a different DT spectral response, where the $L = 2$ resonance is situated between the $L = 0$ and $L = 3$ resonances, serving as an additional confirmation for our assignment.

Coherent spin dynamics of donor-bound electrons were investigated with transient DT responses. Figure 3(b) displays transient DT responses obtained with both the pump and probe at the $L = 0$ D^0X transition. The spin beats result from the precession of the pump-induced electron spin polarization about the external magnetic field. The precession period observed, 440 ps, is in excellent agreement with the well-known g factor ($|g_e| = 0.42$) for donor-bound electrons in GaAs. The spin decoherence time, $T_2{}^*$, decreases with increasing pump power, which is likely due to spin decoherence processes associated with the excitation of excitons.[4,6] At sufficiently weak excitation powers, the spin decoherence time observed is of the order of several ns, and spin beats occur prominently at negative delays as the spin polarization outlasts the 12.5 ns repetition period of the laser. The spin decoherence time is limited by the hyperfine interaction with the surrounding nuclei. The intrinsic decoherence time, T_2, of electron spins in GaAs, with the hyperfine-induced dephasing suppressed, is of the order of μs, as shown in earlier spin-echo studies.[1,13]

It should be pointed out that a spike in Fig. 3(b) occurs at zero pump-probe delay. This spike is due to a coherent-wave mixing process that occurs only when the pump and probe overlap in time and is not related to electron spin precession. The spike can be avoided if the measurement skips over the zero delay region, which is the case for the experiment shown in Figs. 4 and 5. Note also that the pump laser spot size at the sample used for Fig. 3(b) is slightly greater than that used for Figs. 4 and 5.

Spin beats were also observed with the probe at the $L = 3$ resonance and the pump remaining at the $L = 0$ resonance. As shown in Fig. 4(a), these beats are π out of phase with the beats observed when both the pump and probe are at the $L = 0$ resonance. Figure 4(b) also shows the transient DT responses with the probe at the $L = 3$ resonance and with the pump at either the $L = 0$ or the $L = 3$ resonance. Again, the resulting two sets of spin beats are π out of phase. As will be discussed in detail later, these results indicate the $L = 0$ and the $L = 3$ transitions have the opposite polarization selection rules. Note that free excitons excited by the pump pulse can lead to a broadening as well as bleaching of the exciton resonance,[14]

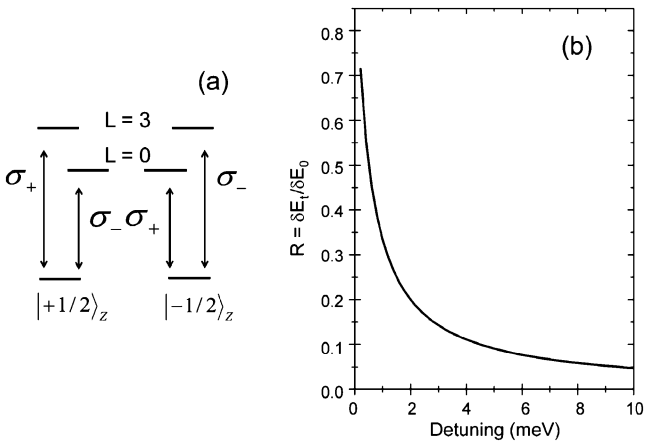

FIG. 6. (a) Optical selection rules with circularly polarized light for the $L = 0$ and $L = 3$ D^0X transitions as discussed in the text. (b) Calculated Stark shift efficiency as a function of detuning from the $L = 0$ transition.

along with a decrease in the electron spin decoherence time. The free exciton transition can thus contribute significantly to the DT responses shown in Figs. 3 and 4. Since the $L = 3$ transition is spectrally closer to the exciton transition than the $L = 0$ transition, effects of free excitons are more prominent when the pump and probe are centered at the $L = 3$ transition.

The full spectral dependence of the spin beats can be observed by comparing the DT response at the peak of an oscillation with that at the trough of an oscillation, providing information on how the absorption spectrum is changing as the electron spins precess. Figure 5(a) shows the DT spectra obtained at various fixed pump-probe delays and with the pump at the $L = 0$ resonance. The dots on the corresponding transient DT response shown in Fig. 5(c) mark the delay times at which the DT spectra were measured. The delay times are labeled P_1, P_2, T_1, T_2, M_1, and M_2 for peak, trough, and midpoint. To further single out the effects of precessing electrons, we subtract the DT spectrum taken at an oscillation midpoint from the DT spectra taken at a peak or a trough. The subtracted DT spectra are shown in Fig. 5(b).

Figure 5(b) illustrates transient behaviors of the DT response probed at individual D^0X transitions. The DT responses obtained with the probe at the $L = 0$ and $L = 3$ resonances are π out of phase, as discussed earlier and also as indicated by the opposite sign of the resonances in these graphs. Furthermore, additional contributions situated between the $L = 0$ and $L = 3$ resonances now become visible. We attribute this relatively weak contribution to the $L = 2$ D^0X transition. As shown in Fig. 5(b), the DT response with the probe at the $L = 2$ transition does not exhibit clear oscillatory behaviors or spin beats.

IV. DISCUSSIONS

While a considerable amount of information on the energy level structures of D^0X transitions in GaAs is available, the polarization selection rules, which are essential to optical spin control, are still not well established or are a subject of debate.[2] No theoretical model has yet been able to account fully for the energy level structure and the polarization selection rules of the D^0X transitions, although numerous models have been presented.[15–17] While detailed experimental studies on polarization selection rules of D^0X have been reported for the lowest lying ($L = 0$ and 1) transitions, there have been few experimental studies on the higher energy transitions ($L = 2$, 3, or higher).

The experimental results presented in the previous section can be understood with an energy level diagram shown in Fig. 6(a), where the $L = 0$ and $L = 3$ transitions have the opposite circular polarization selection rules (in the absence of a magnetic field). For the transient DT experiments, the pump pulse initializes an electron spin polarization, which then precesses around the external magnetic field (along the x axis). With the opposite circular polarization selection rule, the precession of the electron spins leads to a π-phase difference between the spin beats observed at the $L = 0$ and $L = 3$ transitions. The absence of spin precession for the DT response observed at the $L = 2$ transition suggests that the $L = 2$ transition does not have a well-defined selection rule for circularly polarized light.

The energy level structure shown in Fig. 6(a) can seriously limit the effectiveness of optical spin control using polarization-dependent optical Stark shifts. In this case, circularly polarized light (either $\sigma+$ or $\sigma-$ polarized) can couple to both electron spin states via the $L = 0$ and $L = 3$ transitions. This is in sharp contrast to other semiconductor spin systems, in which $\sigma+$ or $\sigma-$ polarized light interacts with only one of the electron spin states (in the absence of an external magnetic field).[7–10] For optical spin control in these systems, a circularly polarized, off-resonant laser pulse induces an optical Stark shift for only one of the electron spin states, leading to an optically induced electron spin splitting and serving effectively as a DC magnetic field along the optical axis.

For donor-bound electrons in GaAs, the optically-induced spin splitting due to the $L = 0$ and $L = 3$ transitions have the opposite sign and can thus cancel each other when the laser pulse is far detuned from the D^0X transition. For an estimate on the degree of cancellation, we define $R = \delta E_t/\delta E_0$ as the effective efficiency for the polarization-dependent optical Stark process, where $\delta E_0 = \Omega^2/\Delta_0$ is the Stark shift induced by a $\sigma+$ polarized field coupling to the $L = 0$ transition and $\delta E_t = \Omega^2(1/\Delta_0 - 1/\Delta_3)$ is the total or net Stark shift, including contributions from both the $L = 0$ and $L = 3$ transitions, and Δ_0 and Δ_3 are the detunings between the optical field and the $L = 0$ and $L = 3$ transitions, respectively. For simplicity, we assumed that Ω, the relevant Rabi frequency, is the same for both transitions. In this limit, we have simply $R = (\Delta_3 - \Delta_0)/\Delta_3$. Figure 6(b) plots R as a function of Δ_0 with $\Delta_3 - \Delta_0 = 0.5$ meV, the energy separation between the $L = 0$ and $L = 3$ resonances. At a modest detuning of 5 meV, R is reduced to about 10%.

The low values of R at modest detunings suggest that for optical spin control using polarization-dependent optical Stark shifts, the rotation pulse needs to be spectrally close to the $L = 0$ transition. However, the excitation of the large nearby free exciton resonance can lead to excessive decoherence, preventing effective optical spin control. In this regard, there are two possible directions for improving optical spin control of donor-bound electrons. One is to explore donor systems

that feature greater binding energy and hence greater energy separation between the relevant D^0X transitions and that between the D^0X transition and the free exciton transition. Another is to find and work with donor systems that feature the desirable polarization selection rules.

V. SUMMARY

Our experimental studies on coherent spin dynamics of donor-bound electrons in GaAs show that D^0X transitions can be spectrally resolved in the nonlinear optical response of the electron spins. The spectral dependence of the phase of the spin beats further reveal that the $L = 0$ and $L = 3$ D^0X transitions have the opposite circular polarization selection rule. As a result, contributions from higher-energy D^0X transitions can seriously limit the effectiveness of optical spin control using mechanisms such as polarization-dependent optical Stark shifts. Donor-bound electrons are unique electron spin systems. We hope that the insights gained from these studies can stimulate further efforts to search for suitable donor systems for optical spin control as well as for other spin-based applications.

ACKNOWLEDGMENT

This work has been supported by the National Science Foundation under grant No. 0804559.

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