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Detection of magnetic resonance of donor-bound electrons in GaAs by Kerr rotation

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Magnetic resonance of electrons in lightly doped GaAs layers has been detected at 5.8 T by magneto-optical Kerr rotation. A study over a wide range of microwave powers shows (1) resonance without dynamic nuclear polarization (DNP), (2) resonance enhanced by DNP, and (3) DNP pinning of the resonance to the external magnetic field. The dependences of the resonance on donor concentration and on the wavelength and intensity of the probing optical beam were also studied. For optimal conditions, the electron *g* factor is −0.428 and the inhomogeneous dephasing time is 5.4 ns. This time is limited by the fluctuation in the local fields produced by the host nuclei.

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There is high current interest in using electron spin as the useful property for a number of new applications. These include spintronics and quantum information where the quantum mechanical properties of spin are exploited.[1] Another set of applications follows from the phenomena of electromagnetically induced transparency (EIT) and slow light.[2,3] This also requires long coherence times in some medium and leads to applications in both quantum information and for optical devices such as delay lines. A solid-state medium of choice for these phenomena is a semiconductor. Long lifetimes have been shown to occur for electrons in GaAs over a very wide range of doping.[4,5] In our own previous work on spins in GaAs, we have focused on the lowest doping levels where the spins become localized and noninteracting. We have observed spin-lattice relaxation times as long as a few μs in this limit.[6] The long spin-lattice and dephasing times possible in this system have allowed a demonstration of coherent population trapping[7]—an important precursor to EIT.

While dephasing times at zero magnetic field for localized spins in GaAs have reached the limit set by the nuclear spins,[8] it has been difficult to realize long dephasing times in the presence of a strong magnetic field. There are two challenges. First, a sensitive method must be found to detect the spins at the very low concentrations required. Second, the state of polarization of the nuclear spins must be controlled in order to minimize the dephasing that they produce. Previous work using electron paramagnetic resonance with microwave detection made use of a slight nuclear polarization and thus fell short of meeting these two challenges.[9] Here, we detect microwave-induced resonance using the magneto-optical Kerr effect[10] to show a fully satisfactory result. The sensitivity of this approach is adequate to observe the resonance without any dynamic nuclear polarization (DNP). In addition, other limits were explored such as enhancement of the resonance signal by DNP and pinning of the resonance to the external field. Samples with different doping concentrations confirm that the experiment has reached the limit where the spins are fully localized and noninteracting. The localized electrons maintain a long dephasing time even in a magnetic field around 6 T.

GaAs doped with donors undergoes a metal-insulator transition at an electron concentration of 2×10^{16} cm^{-3}. To study samples with localized electrons, we have chosen a set of three samples grown by molecular beam epitaxy (MBE) with concentrations of 3×10^{15}, 1×10^{15}, and 3×10^{14} cm^{-3}. These are layers 1 μm thick bounded by AlGaAs regions and show excellent mobilities and photoluminescence intensites.[11] The interactions between the donors decrease with decreasing concentration and are negligible for the lowest concentration. For photoexcitation by a HeNe laser at constant level of 0.5 mW, their emission spectra are shown in Fig. 1(a). The emission is dominated by neutral exciton (X) emission at 818.4 nm and donor-bound exciton (D^0X)

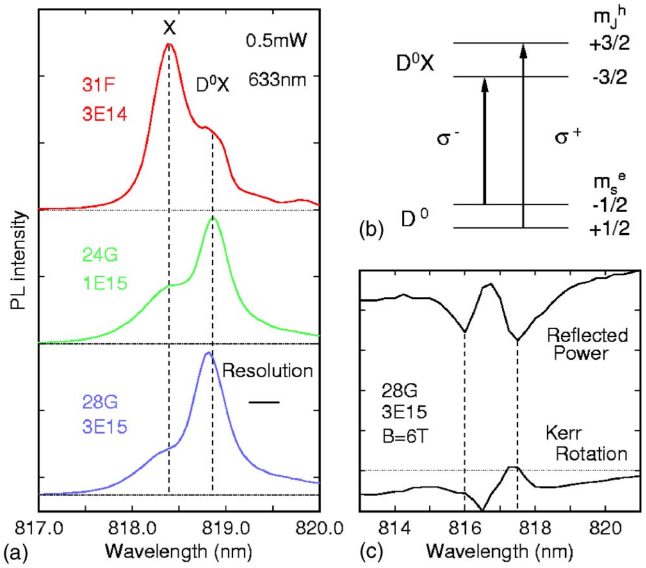

FIG. 1. (Color online) Photoluminescence, reflection, and Kerr rotation in doped GaAs. Part (a) shows the photoluminescence spectra for all three samples under 633 nm excitation of 0.5 mW. The relative contributions of X and D^0X change with donor concentration. Part (b) shows a simplified energy-level scheme for the D^0 and D^0X states in a magnetic field. Allowed transitions for σ^+ and σ^- light are indicated. Part (c) shows the reflected power spectrum and the Kerr rotation spectrum for the highest doped sample at 6 T and 1.5 K. Partially resolved features occur related to X and D^0X transitions. A diamagnetic shift in the transition energy is responsible for the difference in position between the photoluminescence and reflection results.

emission at 818.8 nm. The change in relative intensities shows the increasing importance of the D^0X emission with increasing donor concentration under the moderate photoexcitation.

To perform magnetic resonance detected by Kerr rotation, we first apply a magnetic field in the direction of light propagation. Both the ground and excited states for the D^0X transition split in an applied magnetic field as shown in Fig. 1(b). The ground state in this picture consists of the spin-up and spin-down states for the donor-bound electron. The excited state has two electrons in a spin singlet state and thus its spin derives from the spin of the hole. The level structure has been simplified considerably by taking only one orbital state and only the heavy hole. For a more detailed description see the work-by Karasyuk *et al.*[12] Kerr rotation is performed in reflection and is analogous to Faraday rotation that is performed in transmission. Both effects are proportional to the difference between the index of refraction for right and left circularly polarized light $[\eta(\sigma^+)-\eta(\sigma^-)]$. The experiments were performed with a 7 T Oxford Spectromag superconducting magnet and cryostat mostly at 1.5 K. The light source was a Spectra Physics Tsunami pulsed Ti-sapphire laser with a pulse width of 1.5 ps. The corresponding spectral width is 0.6 nm. The rotation was detected on the reflected beam using a balanced photodiode bridge. With a 6 T magnetic field applied, the most heavily doped sample shows two minima in the reflected power as a function of laser wavelength at 816.0 nm and 817.5 nm [see Fig. 1(c)]. At this field, features in the Kerr rotation are also seen in the same spectral range. The Kerr rotation depends strongly on the population difference between the two ground-state levels. This forms the basis for detecting magnetic resonance of the donor-bound electrons.

With the strong polarization produced by the high magnetic field and low temperature and the detection of polarization through Kerr rotation, we need only a method of altering the spin polarization to produce magnetic resonance. For this the sample was placed in the oscillating magnetic field produced by a 35 GHz microwave cavity with optical access, and this cavity was driven by an Agilent microwave signal generator. Square-wave modulation of the microwaves at 3 kHz allowed lock-in detection of the magnetic resonance signal. When the microwaves are on, the populations of the electrons spins are driven toward being equal. The off part of the modulation cycle is long enough for the electron spins to return to thermal equilibrium.

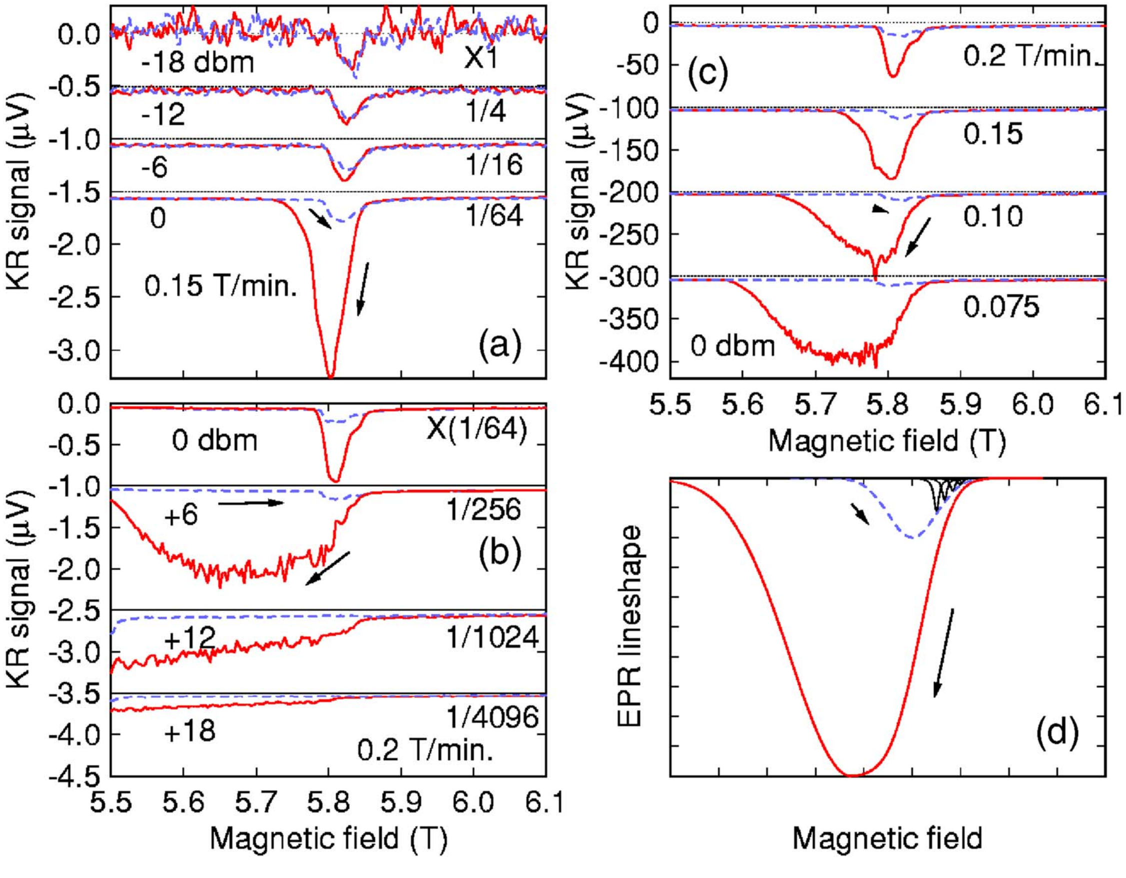

FIG. 2. (Color online) Magnetic resonance detected by Kerr rotation as a function of microwave power and magnetic-field sweep rate. The sample has 3×10^{14} cm^{-3} excess electrons and the light is at 817.8 nm. The microwave frequency is 34.69 GHz and the temperature is 1.5 K. Parts (a) and (b) show the power study with the data normalized such that it would appear constant for an unsaturated spin resonance. In part a (b), the magnetic field sweep rate is 0.15 T/min (0.20 T/min). Three distinct behaviors occur as described in the text. Part (c) shows the sweep rate study for a microwave power of 0 dbm. Part (d) presents a qualitative picture of the signal enhancement by dynamic nuclear polarization. Spin packets acquire effective nuclear fields and remain in resonance with the decreasing external field to produce a strong signal.

If there were no nuclear spins, increasing the microwave power would produce an increasing signal up to the point of saturation as the spin populations become equal. In a Bloch equation analysis, the dependence of the signal amplitude (S^{A}) on microwave power (P) is $S^{\mathrm{A}}=(\alpha P)/(1+\beta P)$, where α and β are constants. But in GaAs, there is nuclear spin on each host atom and dynamic nuclear polarization (DNP) occurs as the electron spins try to relax through the nuclear spins. In a steady state, the average nuclear spin is given by

$$\langle I\rangle = [I(I+1)/S(S+1)] \times [\langle S\rangle - \langle S_T\rangle], \qquad (1)$$

where $\langle S\rangle$ is the average electron spin and $\langle S_{\mathrm{T}}\rangle$ is the average electron spin in thermal equilibrium.[13] Since $\langle S_{\mathrm{T}}\rangle$ is strong in this experiment and the microwaves make $\langle S\rangle$ smaller than $\langle S_{\mathrm{T}}\rangle$, $\langle I\rangle$ is negative and can take on significant values when high microwave power reduces $\langle S\rangle$ significantly. To see the sign of the effective nuclear field B_{N} created by the DNP, we note that $B_{\mathrm{N}}=A\langle I\rangle/g\mu_B$, where A is the hyperfine constant for a particular nuclear spin. Since in GaAs, g is negative and $\langle I\rangle$ can become negative in this experiment, the effective field produced by the nuclei is positive. The appropriate equation for electron spin resonance when there is DNP becomes $h\nu=g\mu_B\ (B_{\mathrm{ext}}+B_{\mathrm{N}})$. Since DNP depends on the saturation of the electron spin polarization, the rate of nuclear polarization depends on the microwave power.

In the experiment, the external magnetic field is swept through magnetic resonance leading to a slow dynamics with respect to the nuclear spins. When the external field is swept to smaller values at a rate $|dB_{\mathrm{ext}}/dt|$ equal to or less than the rate of nuclear polarization dB_{N}/dt, strong enhancement[9] of the magnetic resonance signal or pinning[14] of the resonance to the changing magnetic field can occur.[15]

The dependence of the Kerr-detected magnetic resonance on microwave power is shown in Figs. 2(a) and 2(b). The data were taken on the lowest doped sample with the laser set at 817.8 nm on the long wavelength side of the D^0X transition. Results are shown for magnetic-field sweeps in both directions with a slight change in the rate between parts 2(a) and 2(b). The signals were normalized such that the displayed curves would be constant if the resonance were unsaturated. Three limits can be distinguished that differ in the rate on nuclear polarization (dB_{N}/dt) relative to the rate of change of the external magnetic field ($|dB_{\mathrm{ext}}/dt|$). First, the signals are unsaturated for powers from -18 to -6 dbm ($dB_{\mathrm{N}}/dt<|dB_{\mathrm{ext}}/dt|$). The up and down scans are very similar and the normalized signal amplitudes are constant. Kerr-detection provides enough sensitivity to detect the resonance under negligible saturation. Second, the signals are enhanced by DNP for down scans at powers from 0 to +6 dbm ($dB_{\mathrm{N}}/dt\sim|dB_{\mathrm{ext}}/dt|$). The down scans are broadened, shifted, and enhanced in amplitude. The up scans are essentially unchanged from the expectation for unsaturated signals. Noise from changing nuclear polarization appears on the down scan at 6 dbm. These signals are similar to those observed for donor spins by Seck *et al.*[9] using microwave detection. Third, the resonance is pinned to the external field for down scans at powers of +12 and +18 dbm ($dB_{\mathrm{N}}/dt>|dB_{\mathrm{ext}}/dt|$). Signals are observed in down scans that never reach maximum amplitude. On the up scans, there is signal at the starting field with nothing at the original resonance position. This limit was observed in studies of two-dimensional electron gases in GaAs.[14–16]

In the microwave power study, the rate of nuclear polarization changes since DNP depends on saturation of the electron spin polarization and saturation depends in turn on microwave power. Figure 2(c) shows the converse where the microwave power and, hence, dB_{N}/dt are held constant while the magnetic sweep rate ($|dB_{\mathrm{ext}}/dt|$) is varied. A series of curves is observed that mimics the data in the microwave power study. A simple explanation for these effects is given in Fig. 2(d). Individual spins packets occur for different local magnetic fields. For a down scan with $dB_{\mathrm{N}}/dt\sim|dB_{\mathrm{ext}}/dt|$, the spin packets are gaining additional local magnetic field from nuclear polarization at a rate that matches the change in external field and they stay in resonance. A large signal is produced that is something like the integral of the original Gaussian envelope. After a while, the rates become out of synchronization and the signal returns to zero. In contrast, when the external field is swept to larger values, spin packets go out of resonance since their total fields ($B_{\mathrm{ext}}+B_{\mathrm{N}}$) become too high.

Further insight on the detection process and DNP effects can be gained by studies of the optical parameters and samples with different doping densities. The dependence of the magnetic resonance on wavelength of the light is shown

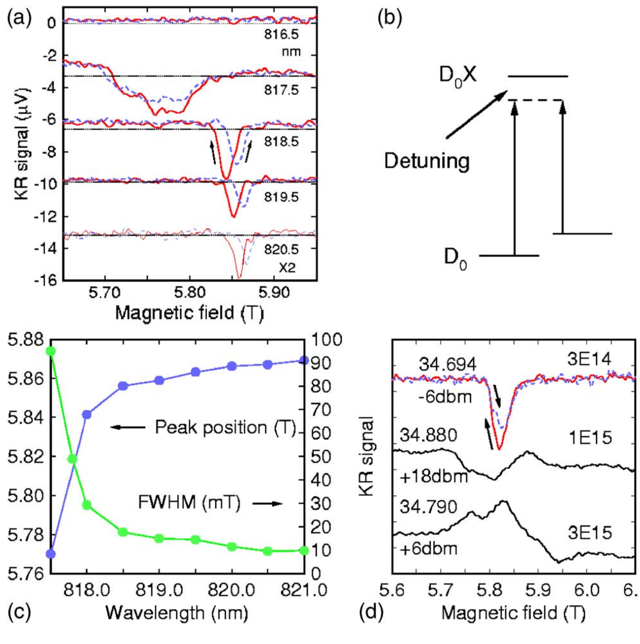

FIG. 3. (Color online) Dependences of the magnetic resonance on the wavelength of the light and on the electron density. Part (a) shows the resonance as the wavelength is varied for the lowest concentration sample and with the microwave power at −6 dbm. Part (b) is a schematic drawing showing the spectral study as detuning from the D^0X transition. Part (c) shows the shift in peak position and the narrowing that occur as wavelength increases. Both parameters approach limiting behavior with increasing detuning. Part (d) shows data taken at 817.8 nm for the three samples. For the higher concentrations, the lines have multiple features and consist of both increases and decreases in Kerr rotation.

in Fig. 3(a). This study reveals the effects of optical detuning from the spectral region of X and D^0X to lower energies, that is, farther from the GaAs band gap [see Fig. 3(b)]. For wavelengths shorter than that of D^0X, no magnetic resonance is observed. At D^0X (817.5 nm) the resonance is very broad. Continuing to longer (off optical resonance) wavelengths, the magnetic resonance narrows and shifts and then approaches limiting behavior [Fig. 3(c)]. This is the best condition since fewer photoexcited electrons and holes are produced. Most of the data were taken with a power in the unfocussed beam of 300 μW. Varying the optical power, the resonance also shifts to lower magnetic fields and the line broadens with increasing power. The samples with higher donor concentrations show multiple lines with signals that both increase and decrease the Kerr rotation [Fig. 3(d)]. These results indicate that, when there are electrons present with different degrees of localization and isolation, resonances occur at different positions. The differing degrees of localization may produce a difference in g factor, or a change in electron spin relaxation rate, and consequently a change in the degree of local nuclear polarization. The effects seen in the higher concentration samples are also similar to what is seen in the 3E14 sample with light resonant with D^0X [Fig. 3(a)]. Here, resonant excitation creates a significant population of excitons that interact with (ground-state) electron spins.

The best results occur for the sample with the lowest electron concentration and with the probing light off of optical resonance. In these conditions, the donors are widely separated, producing the most homogeneous ensemble, and the off-resonant light is a very weak perturbation. The limiting values of line position and linewidth seen for these conditions [Fig. 3(c)] allow measurements of the g factor and dephasing time (T_2^*). With the light at 820.5 nm, the g factor is −0.428 (negative sign added) and is slightly smaller than the value given by Seck *et al.* for this magnetic field (−0.433).[9] The linewidth of 10 mT converts to a T_2^* of 5.4 ns. This value is in agreement with the expected relaxation from the fluctuation in nuclear spin.[17] For comparison, it is better than that seen in the microwave-detection experiments of 1.1 ns. Both values are far better than the results in semi-insulating samples where g broadening is significant.[4,18] In the present case with the magnetic field around 6 T, the Kerr-detected resonance shows negligible g broadening and negligible DNP.

In summary, we have used Kerr rotation to detect magnetic resonance in GaAs at high magnetic field. In the lowest doped sample with low microwave power and detuned light, the linewidth converts to a T_2^* of 5.4 ns. This lifetime implies both negligible g broadening and negligible DNP. The Kerr-rotation approach should allow the detection of the homogeneous lifetime, T_2, for donors in GaAs through the use of a Hahn spin-echo experiment.[19]

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[1] S. A. Wolf, D. D. Awschalom, R. A. Buhrman, J. M. Daughton, S. von Molnár, M. L. Roukes, A. Y. Chtchelkanova, and D. M. Treger, Science **294**, 1488 (2001).

[2] S. E. Harris, Phys. Today **50**, 36 (1997).

[3] L. V. Hau, S. E. Harris, Z. Dutton, and C. H. Behroozi, Nature (London) **397**, 594 (1999).

[4] J. M. Kikkawa and D. D. Awschalom, Phys. Rev. Lett. **80**, 4313 (1998).

[5] I. Zutic, J. Fabian, and S. Das Sarma, Rev. Mod. Phys. **76**, 323 (2004).

[6] J. S. Colton, T. A. Kennedy, A. S. Bracker, and D. Gammon, Phys. Rev. B **69**, 121307(R) (2004).

[7] Kai-Mei Fu, C. Santori, C. Stanley, M. C. Holland, and Y. Yamamoto, Phys. Rev. Lett. **95**, 187405 (2005).

[8] R. I. Dzhioev, K. V. Kavokin, V. L. Korenev, M. V. Lazarev, B. Ya. Meltser, M. N. Stepanova, B. P. Zakharchenya, D. Gammon, and D. S. Katzer, Phys. Rev. B **66**, 245204 (2002).

[9] M. Seck, M. Potemski, and P. Wyder, Phys. Rev. B **56**, 7422 (1997).

[10] R. Romestain, J. Phys. C **13**, 1097 (1980).

[11] J. S. Colton, T. A. Kennedy, A. S. Bracker, and D. Gammon, Phys. Status Solidi B **233**, 445 (2002).

[12] V. A. Karasyuk, D. G. S. Beckett, M. K. Nissen, A. Villemaire, T. W. Steiner, and M. L. W. Thewalt, Phys. Rev. B **49**, 16381 (1994).

[13] D. Paget and V. L. Berkovits, in *Optical Orientation*, edited by F. Meier and B. P. Zakharchenya (North Holland, Amsterdam, 1984), p. 388.

[14] A. Berg, M. Dobers, R. R. Gerhardts, and K. v. Klitzing, Phys. Rev. Lett. **64**, 2563 (1990).

[15] E. Olshanetsky, J. D. Caldwell, A. E. Kovalev, C. R. Bowers, J. A. Simmons, and J. L. Reno, Physica B **373**, 182 (2006).

[16] C. Hillman and H. W. Jiang, Phys. Rev. B **64**, 201308(R) (2001).

[17] I. A. Merkulov, Al. L. Efros, and M. Rosen, Phys. Rev. B **65**, 205309 (2002).

[18] J. M. Trombetta and T. A. Kennedy, Phys. Rev. B **48**, 17031 (1993).

[19] T_2 has recently been measured for a single qubit. See J. R. Petta, A. C. Johnson, J. M. Taylor, E. A. Laird, A. Yacoby, M. D. Lukin, C. M. Marcus, M. P. Hanson, and A. C. Gossard, Science **309**, 2180 (2005).