

Dependence of optically oriented and detected electron spin resonance on donor concentration in *n*-GaAs

J.S. Colton*, T.A. Kennedy, A.S. Bracker, J.B. Miller, D. Gammon

Naval Research Laboratory, Washington, DC 20375, USA

Received 6 August 2004; accepted 31 August 2004 by S Das Sarma

Available online 11 September 2004

Abstract

Electron spin resonance of donors in GaAs has been observed through optical orientation and detection of spins. GaAs samples doped below the metal–insulator transition were studied. The resonance linewidth increases as the concentration of donors is reduced, due to the dependence of the T_2^* spin lifetime on correlation effects between donor electrons. The linewidth of the lowest doped sample ($3 \times 10^{14} \text{ cm}^{-3}$) corresponds to a T_2^* of 5 ns, which is the value predicted for electrons in the non-interacting, localized limit. The nuclei need to be simultaneously depolarized in order to make the electron resonance observable.

© 2004 Elsevier Ltd. All rights reserved.

PACS: 76.30.-v; 78.55.Cr; 72.25.Rb; 72.25.Fe

Keywords: A. Donors; A. GaAs; E. Optical; E. Spin resonance

Electronic spins in semiconductor quantum dots have been proposed in scalable potential quantum computing (QC) schemes [1]. In order for such schemes to be functional, the spin lifetime must be long compared to the time scale of computing operations [2]. GaAs and related materials are being considered as potential QC materials [1], and recent observations of electron spin dephasing times in the tens and hundreds of nanoseconds in *n*-type GaAs have been encouraging [3–6]. This paper deals with electrons in lightly doped *n*-GaAs, where the localization of the electrons on the donors gives rise to spin properties which are similar to electrons localized in quantum dots.

For localized electrons in GaAs quantum dots, the T_2^* spin lifetime has been theoretically predicted to be hyperfine interaction-limited to about 1–5 ns [6–8]. The lifetimes are

extended from this limit when some electron delocalization occurs through correlation between electrons and hopping between donors [6,9]. Due to the strong hyperfine interaction experienced by localized electrons and due to the low concentrations of electrons, resonance experiments in *n*-type GaAs below the metal–insulator transition have proven difficult to perform. For example, the classic resonance research by Weisbuch and Hermann in GaAs and other III–V compounds was typically with conduction-band electrons in *p*-type material [10,11].

Previous spin resonance experiments in lightly doped *n*-GaAs have been performed by two groups: Seck et al. [12], and Colton et al. [13]. In both cases, interaction between electron and nuclear spins proved to be a source of difficulties for the ESR, due to the electrons seeing a changing effective magnetic field produced by the Overhauser polarization of the nuclei. The two groups took varying approaches in dealing with the problem. Seck et al. performed a high field experiment (6–10 T) and controlled such nuclear effects by carefully timing their sweep rates. The result was a broad peak—the linewidth was about

* Corresponding author. Present address: Physics Department, University of Wisconsin–La Crosse, WI 54601, USA. Tel.: +1 6087858432; fax: +1 6087858403.

E-mail address: colton.john@uwlax.edu (J.S. Colton).

50 mT, presumably due to a combination of nuclear effects and g -broadening. Colton et al., conversely, performed a low field experiment (41 mT), limiting the possibility of g -broadening, and controlled the nuclear effects via NMR. The NMR served to depolarize the nuclear spins: when the NMR was set appropriately, the ESR peak proved to be quite narrow (2 mT width), but as NMR power was reduced, the ESR peak shifted, broadened, and decreased in amplitude. The 2 mT linewidth corresponds to a spin lifetime 28 ns, and is lengthened from the 1–5 ns localized limit by correlations between electrons.

This present work continues the previous experimental technique of Colton et al., by simultaneously resonating the nuclei while performing optically oriented and detected spin resonance on the electrons—a variation of optically detected magnetic resonance (ODMR). The dependence of the spin resonance on sample concentration has been investigated for three samples below the metal–insulator transition. A resonance was observed on even the lowest-doped sample, in which the donors are spaced far enough away from each other that the electrons are essentially fully localized. To our knowledge, this is the first report of ODMR performed on fully localized electrons in GaAs. The width of the resonance peak for this sample indicates a T_2^* spin lifetime of 5 ns, which agrees well with the theoretically predicted result for the localized limit.

The samples we investigated were 1 μm thick GaAs layers in an AlGaAs heterostructure, whose growth and characteristics are described elsewhere [14]. Three different doping levels were studied: 3×10^{14} , 1×10^{15} and $3 \times 10^{15} \text{ cm}^{-3}$. The samples are labeled 3E14, 1E15, and 3E15, respectively. The 3E15 sample is the same one studied in Ref. [13].

Our experiments were performed at 5–6 K in a liquid helium cryostat. Electronic spins in the samples were oriented through exchange interactions with photo-excited electrons and excitons; the excitation was done by circularly polarized light at 785 nm. The spin state was read out by measuring the polarization of luminescence of the free exciton [11,13,15,16]. A photoelastic modulator was used to alternate the exciting light between σ^+ and σ^- , and helped to reduce (though not completely eliminate) the build-up of nuclear polarization which would occur with cw σ^+ light. VHF frequencies to resonate the electrons were applied via a loop in series with a capacitor (forming an LC circuit); rf frequencies to simultaneously resonate the nuclei were applied by a coil positioned perpendicularly to the VHF loop. The loop and coil provide oscillating magnetic fields without accompanying oscillating electric fields that would couple strongly to the charge of the electrons.

The experimental conditions required for the best results were as follows: the rf was swept from 75 to 800 kHz (this is a wider sweep than had been used in Ref. [13]; some details on how sweep width affected the resonance are provided in Fig. 4), at a sweep rate of 5 Hz, and at a power level of 1–2 W producing a B_1^{HF} of ~ 0.3 mT. The VHF was applied at a

fixed 236 MHz—the resonant frequency of the LC circuit—and was at a power level of 5 W producing a B_1^{HF} of ~ 1 mT. The resonance peaks for each of the three samples under these conditions can be seen in Fig. 1.

The peaks in Fig. 1 have been fit to a Lorentzian function:

$$\frac{\Delta P}{P_0} = \frac{A}{B_{1/2}^2 + (B - B_0)^2} \quad (1)$$

where the center, B_0 , is determined by the g -factor:

$$B_0 = \frac{\hbar \nu_{\text{res}}}{g \mu_B} \quad (2)$$

and the half width at half max, $B_{1/2}$, is determined by the T_2^* spin lifetime:

$$B_{1/2} = \frac{\hbar}{g \mu_B T_2} \quad (3)$$

Eqs. (1)–(3), have been used with the fits to extract g -factor and the T_2^* spin lifetime for each of the three samples. The values for those fitted parameters are given in Table 1.

The most notable feature is the decrease in lifetime as the donor concentration is decreased, as seen by an increase in linewidth. This corresponds to the well-understood theory of interacting donor electrons: the interaction between electrons provides an averaging effect, which reduces the dephasing from the nuclei. The spin interaction can be characterized by a correlation time. As the donor concentration is decreased, the correlation time increases, and the spin lifetime decreases. Fig. 2 shows these measured T_2^* values superimposed on the results of Dzhirov et al. for n -GaAs samples [6]. The points labeled τ_S are the experimental T_2^* spin lifetimes measured by that group via the Hanle effect. The τ_C points are their deduced values for the correlation time based on τ_S measurements in longitudinal applied fields. Our measured T_2^* values are

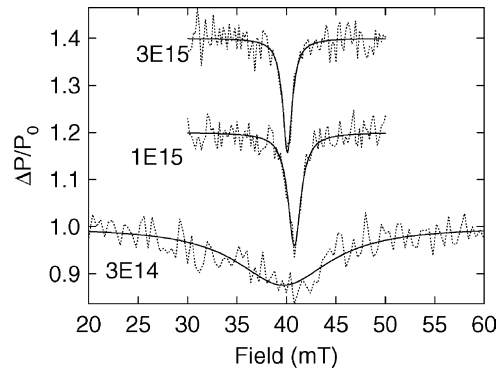


Fig. 1. Spin resonance for three different donor concentrations. The linewidth decreases by an order of magnitude with increasing concentration. The change in polarization at resonance is about 7% for the $3\text{E}15 \text{ cm}^{-3}$ sample. The 3E15 and 1E15 curves have been displaced vertically for clarity.

Table 1
|g| and T_2^* of the samples

Donor concentration (cm^{-3})	g -Factor	T_2^* (ns)
3E14	0.43	4.6
1E15	0.41	35
3E15	0.42	47

consistently below the Hanle values; this could have to do with differences in the samples or incomplete nuclear depolarization.

Two additional observations from our data should be pointed out. First, the extremely narrow line displayed in the 3E15 sample—close to 1 mT—is among the narrowest ever seen in GaAs. That demonstrates both the quality of our samples and the effectiveness of this technique. Secondly, the 4.6 ns lifetime measured for the 3E14 sample agrees well with the theoretical prediction for the fully localized case. Thus we believe we have successfully used optically detected electron spin resonance to determine the T_2^* of completely localized electrons—a first for GaAs.

As mentioned above, control of the nuclear spins via NMR was a critical aspect of this experiment. The three nuclear isotopes were successively resonated by sweeping the rf across the resonant nuclear frequencies. As the sweep passed through a nuclear resonance, the spin polarization of that isotope was effectively reset to zero. If, however, the NMR sweep rate was reduced, the nuclei would begin to polarize.

Uncontrolled nuclear polarization adversely affected the resonance experiment. When the electron spins are out of thermal equilibrium, one way they attempt to relax is through the hyperfine interaction, which produces a nuclear polarization. There are two aspects of the present

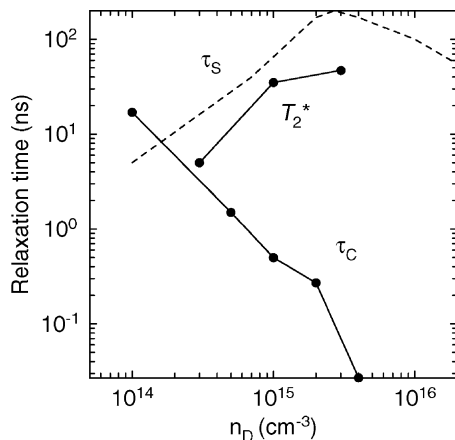


Fig. 2. Spin relaxation and electron correlation times for low donor concentrations. T_2^* is taken from the resonance data. τ_S and τ_C are the spin relaxation time and the electron correlation time from Ref. [5]. At $3 \times 10^{14} \text{ cm}^{-3}$, the T_2^* reaches the correlation time—indicating strong localization.

experiment, where the electron spins are taken out of equilibrium. First, excitation by circularly polarized light leads to polarization of the donor spins—nuclear polarization is avoided here by modulation of the polarization of the exciting light, as previously mentioned. Second, and more to the point here, at electron spin resonance the spin populations for spin packets in resonance are equalized—taken out of equilibrium—by the strong VHF field. The nuclei near the spin packets in resonance become polarized and produce a shift in the local field, which smears out a clean resonance. Controlling the nuclear polarization via appropriate NMR sweeps eliminated this smearing.

The resonance peak's dependence on NMR sweep rate is shown in Fig. 3 for the 3E15 sample. For sweep rates of less than $\sim 0.5 \text{ Hz}$, the electron resonance peak shifts, broadens, and is reduced in intensity precisely in the same way that has been previously reported for the peak's dependence on NMR power [13]. Thus the nuclei re-polarize on that time scale, and an estimate of the nuclear polarization time (T_1') can be made of $\sim 2 \text{ s}$.

A similar effect is seen when the NMR sweep limits are changed so as to not cover one of the nuclear isotopes. Fig. 4(a) shows the effect as the end of sweep was changed so that ^{71}Ga was not covered; Fig. 4(b) shows it as the start of sweep was changed so that ^{75}As was not covered. In both cases, as the isotope begins to cease being covered by the NMR, the electron resonance peak shifts, broadens, and reduces in intensity. The scale over which the resonance peak changes its behavior yields an estimate of the broadening of the NMR peaks: $\sim 50\text{--}100 \text{ kHz}$ in both studied isotopes.

In conclusion, we have measured electron spin resonance in three lightly doped *n*-GaAs samples, using optical methods for the alignment and detection of spins. The measurement on the lowest-doped sample indicates that we were successfully probing localized electron spins. Thus, optical polarization and photoluminescence detection might

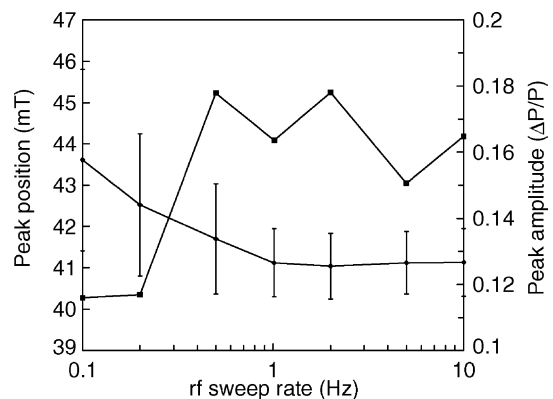


Fig. 3. Peak position and peak amplitude of the electron resonance peak of sample 3E15, plotted vs. the NMR rf sweep rate. The peak width is indicated by the error bars on the peak position plot.

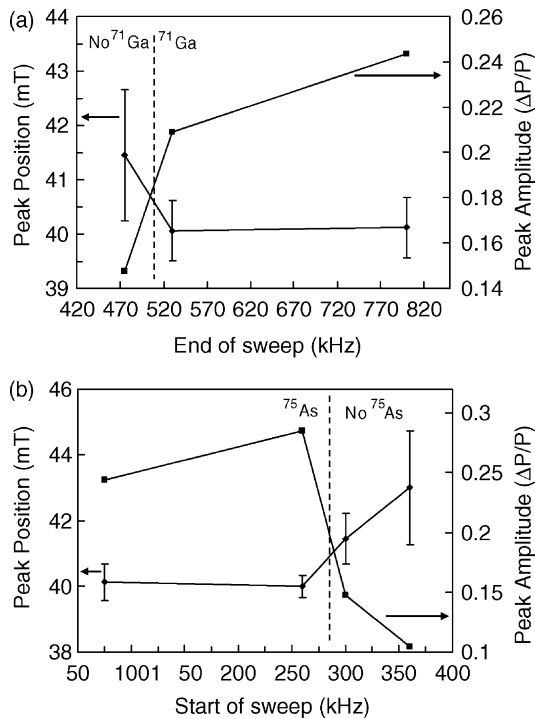


Fig. 4. Peak position and peak amplitude of the electron resonance peak of sample 3E15, plotted vs. the NMR rf sweep ending frequency (a) and rf sweep beginning frequency (b). The peak width is indicated by the error bars on the peak position plot.

be applicable to single dot measurements [17]. Future work will involve pursuing spin echo measurements of the homogeneous T_2 spin lifetime. The type of spin resonance described in this paper, combined with time-resolved optical spin measurements such as those that measured the T_1 spin flip time [18], should make a T_2 measurement possible. A prime consideration in a T_2 experiment will be the choice of magnetic field/ESR frequency. For example, the T_1 measurements indicate that fields of at least ~ 1 T must be used to have a hope of obtaining T_2 results above 100 ns or so—which is the time scale needed when using microwave pulse sequences at practical power levels.

Acknowledgements

We thank A.I.L. Efros for helpful discussions. JSC was an NRL-NRC Research Associate. Work has also been supported by DARPA and ONR.

References

- [1] D. Loss, D.P. DiVincenzo, Phys. Rev. A 57 (1998) 120.
- [2] M.A. Nielsen, I.L. Chuang, Quantum Computation and Quantum Information, Cambridge University Press, New York, 2002, p. 278.
- [3] R.I. Dzhioev, B.P. Zakharchenya, V.L. Korenev, M.N. Stepanova, Phys. Solid State 39 (1997) 1765.
- [4] J.M. Kikkawa, D.D. Awschalom, Phys. Rev. Lett. 80 (1998) 4313.
- [5] R.I. Dzhioev, V.L. Korenev, I.A. Merkulov, B.P. Zakharchenya, D. Gammon, A.I.L. Efros, D.S. Katzer, Phys. Rev. Lett. 88 (2002) 256801.
- [6] R.I. Dzhioev, K.V. Kavokin, V.L. Korenev, M.V. Lazarev, B.Ya. Meltser, M.N. Stepanova, B.P. Zakharchenya, D. Gammon, D.S. Katzer, Phys. Rev. B 66 (2002) 245204.
- [7] M.I. D'yakonov, V.I. Perel, Sov. Phys. JETP 38 (1974) 177.
- [8] I.A. Merkulov, A.I.L. Efros, M. Rosen, Phys. Rev. B 65 (2002) 205309.
- [9] K.V. Kavokin, Phys. Rev. B 64 (2001) 075305.
- [10] C. Weisbuch, C. Hermann, Phys. Rev. B 15 (1977) 816.
- [11] F. Meier, B.P. Zakharchenya (Eds.), Optical Orientation, North-Holland, Amsterdam, 1984.
- [12] M. Seck, M. Potemski, P. Wyder, Phys. Rev. B 56 (1997) 7422.
- [13] J.S. Colton, T.A. Kennedy, A.S. Bracker, D. Gammon, J.B. Miller, Phys. Rev. B 67 (2003) 165315.
- [14] J.S. Colton, T.A. Kennedy, A.S. Bracker, D. Gammon, Phys. Status Solidi B 233 (2002) 445.
- [15] D. Paget, Phys. Rev. B 24 (1981) 3776; D. Paget, Phys. Rev. B 25 (1982) 4444.
- [16] V.B. Vekua, R.I. Dzhioev, B.P. Zakharchenya, G. Fleisher, Sov. Phys. Semicond. 10 (1976) 210.
- [17] O. Gywat, H.-A. Engel, D. Loss, R.J. Epstein, F.M. Mendoza, D.D. Awschalom, Phys. Rev. B 69 (2004) 205303.
- [18] J.S. Colton, T.A. Kennedy, A.S. Bracker, D. Gammon, Phys. Rev. B 69 (2004) 121307(R).