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Nuclear effects in Kerr rotation-detected magnetic resonance of electrons in GaAs

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ABSTRACT

We have studied nuclear effects on electron spins in lightly doped n-GaAs bulk and quantum well samples using optically detected magnetic resonance, with the optical detection done via Kerr rotation. The magnetic resonance signal allowed us to measure the electron T_2^* lifetimes and g -factors. We observed strong dynamic nuclear polarization, which was controllable via nuclear magnetic resonance techniques. We measured the nuclear spin relaxation time (T_1) by monitoring the changing Overhauser field in time, and were able to perform combined electron and nuclear resonance.

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1. Introduction

Applications in spintronics and quantum information require electron spin states which can be manipulated and have good spin properties [1]. Long spin lifetimes are essential to prevent loss of information encoded in the electrons' spin. Electron spins in GaAs are good candidates for spin qubits, as has been shown by the long spin lifetimes previously reported for electrons in bulk n-type GaAs [2–6], electrons in GaAs-related self-assembled quantum dots [7], and electrons in gated GaAs [8–13] and GaAs-related [14] nanostructures.

Electron spin resonance techniques (ESR), including optically detected magnetic resonance (ODMR), provide a well-established tool for studying and manipulating spins [15,16]. As opposed to the now-standard technique of time-resolved Faraday or Kerr rotation [17], which observes the precession of spins in the transverse

plane of the Bloch sphere, ESR induces transitions between the spin-up and spin-down eigenstates and under the right conditions can make coherent rotations of the spin vector possible, for example, to observe Rabi oscillations and spin echoes.

Despite early success in p-type materials [18], ESR of electrons in n-type GaAs has proven more elusive. n-type material is desirable from a quantum information point of view, because only with added dopants can the spin lifetime extend beyond the optical lifetime. Aside from recent experiments in gated quantum dots [19,20], the increased hyperfine interaction between electrons and nuclei that is present when electrons are localized has seriously impacted ESR experiments in GaAs. Thus, the only ESR experiments in bulk n-GaAs of which we are aware are high-field measurements by Seck et al. [21], and Kennedy et al. [22], where the electron–nuclei interaction substantially impacted the resonance lineshapes, and low-field measurements by Colton et al., where deliberate steps were taken to remove that interaction [23,24]. The objectives of this work were to perform ODMR of electrons in n-type GaAs at moderate fields as a necessary prerequisite for coherent spin rotations, and to study and control the nuclear effects on the electron spins.

2. Material and methods

The samples described in this paper are an $n = 3 \times 10^{14} \text{ cm}^{-3}$ GaAs layer, and a multi-quantum well (QW) sample modulation-doped at $n = 3 \times 10^{10} \text{ cm}^{-2}$. The quantum well sample has wells of

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2.8, 4.2, 6.2, 8.4, and 14 nm [25]; all of our results reported here are from the 14 nm well, which was selected by tuning our probe laser to the optical transition of that well. These two samples have been studied by several groups which have reported on the exciton and trion fine structure [25], as well as spin properties of the electrons in the two samples [22,24,26,27].

Spin resonance experiments require an initial spin polarization, a modification of the spin polarization, and a continual detection of the spin polarization as the spins are being modified. We initialized the spins via thermal effects: the samples were placed in a 1.5–1.9 T magnetic field and cooled to 1.4–1.5 K. The magnetic field causes the electron spin states to split by $\Delta E = |g|\mu_B B$; simple two-level Boltzmann statistics predict the polarization of electron spins to be $\tanh(|g|\mu_B B_{\text{ext}}/2k_B T) = 14\%–20\%$ for typical experimental conditions.

We used microwaves at about 10 GHz to cause transitions between the two spin states when the resonance condition is fulfilled: $hf = |g|\mu_B B$. This changes the overall electron spin polarization. A custom variable-frequency microwave cavity was used to enhance the microwave field at the sample with a typical loaded Q-factor of about 1800 [28]. The microwaves were chopped at frequencies ranging from 1 to 100 kHz.

The electron spin polarization was monitored through Kerr rotation (KR), the rotation of the angle of polarization of a linearly polarized probe beam in response to the magnetization or spin polarization of a sample. The probe polarization is measured with a polarizing beam splitter and two balanced diode detectors. The signal from the balanced detectors is sent to a lock-in amplifier, using the microwave chop frequency as its reference. KR is measured in reflection geometry but is otherwise similar to Faraday rotation in transmission [29]. The probe laser was tuned to 807.5 nm for the QW sample, to be resonant with the QW optical transition; it was tuned to 821 nm for the bulk sample to be a few nm below the band-edge transition. When the electron spins are under resonance, the change in spin polarization is seen as a change in the KR. This detection technique is similar to that of Kennedy et al. [22] except our use of a cw rather than pulsed laser allows for a more precise tuning of the probe beam energy.

3. Results and discussion

In spin resonance experiments, the peak position yields the g -factor: $|g| = hf/\mu_B B$; and the peak width yields the T_2^* spin lifetime: $T_2^* = \hbar/(g\mu_B B_{\text{half-width}})$. Fig. 1 displays typical ODMR peaks of our two samples, with deduced QW g -factors of -0.444 and -0.346 for the bulk and QW samples, respectively (negative signs taken from the literature). The bulk value is consistent with other measured values by our group and others [18,22,24]. The QW value matches the value given by Snelling et al. [30]; the apparent disagreement of our g -factor and that of -0.29 measured by Kennedy, et al. for this same sample and quantum well – using time-resolved KR [27] – is likely explained by the difference between g_{\parallel} and g_{\perp} in quantum well samples as outlined by Malinowski and Harley [31].

The T_2^* spin lifetimes deduced from Fig. 1 are 6 ns and 9 ns for the bulk and QW samples, respectively. These are consistent with theoretical predictions for localized electrons dephasing from frozen fluctuations in the nuclear hyperfine field [32], and indicate that effects from the electron–nuclear interaction play a dominant role in the behavior of electron spins in these samples at these temperatures and fields. They also indicate that little inhomogeneous g -broadening is occurring.

The electron–nuclear interaction arises from the hyperfine coupling, $H = AS \cdot I$ [33]. This interaction creates an effective magnetic field seen by the electrons, often called the “Overhauser field”: $B_{\text{eff}} = A\langle I \rangle/g\mu_B$, where $\langle I \rangle$ is the average nuclear spin and A is the hyperfine coupling constant. This Overhauser field adds to the

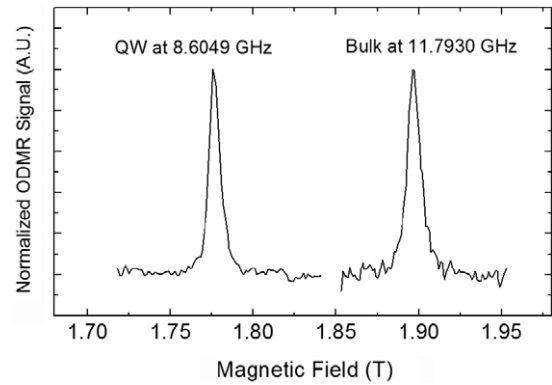


Fig. 1. Typical ODMR scans for each sample. The quantum well data was taken with 11 dBm microwaves at 8.6049 GHz and a 0.10 mW probe laser at 807.5 nm. The bulk data was taken with 22 dBm microwaves at 11.7930 GHz and a 0.2 mW probe laser at 821.0 nm. Although the peak positions are close together, the g -factors are very different—as is evidenced by the very different frequencies needed to resonate the spins at this field. The g -factors are -0.444 and -0.346 for the bulk and QW samples, respectively.

externally applied magnetic field such that $B_{\text{tot}} = B_{\text{ext}} + B_{\text{eff}}$. Variation in the Overhauser field from electron to electron is precisely what causes the dephasing which limits the T_2^* values given above. The Overhauser field also creates a shift in the external field needed to induce spin flips, as the resonance condition becomes $hf = |g|\mu_B(B_{\text{ext}} + B_{\text{eff}})$. Because the Overhauser field is proportional to $\langle I \rangle$, a shift in ODMR peak position is evidence of dynamic nuclear polarization (DNP), and is also a direct measure of the amount of nuclear spin polarization.

The electron–nuclear interaction causes the nuclei to become polarized when the electron spins are taken out of equilibrium [34]. Both the quantum well and the bulk sample displayed DNP; as seen in Fig. 2(a), it occurs with higher laser powers, where the probe laser injects more and more unpolarized (non-equilibrium) electrons and ceases to be a probe. Local inhomogeneity in the nuclear polarization, likely caused in part by the Gaussian profile of the probe laser beam, is manifested as broadening of the ODMR peak for medium powers. The flattening of the peaks at the highest laser powers indicates that electrons are being pinned in resonance: the Overhauser field is increasing at the precise rate at which the external field is decreasing (the field is swept downward), keeping the spins continually in resonance. DNP-related shifting and broadening of the peak also occurs with increasing microwave power; see Fig. 2(b). The peak shift could be towards lower or higher fields, depending on whether the nuclear-related B_{eff} is aligned with or against the external field. In general, we observed more DNP effects in the bulk sample than in the QW sample under similar conditions. This indicates a stronger nuclear interaction, likely meaning the electrons are more localized in the bulk sample (localization caused by donor impurities) than in the QW sample (no donor impurities present), and is consistent with the longer T_2^* value measured for the QW sample.

Since the Overhauser field is proportional to $\langle I \rangle$, successive measurements of the peak position can be used to observe the relaxation of the nuclear spins. To do this in the bulk sample we first polarized the nuclei by pumping the sample with a 25 mW circularly polarized laser at 781 nm. This caused the ODMR peak to shift by ± 35 mT, the sign depending on the pump laser helicity. The ODMR peak position changed back to equilibrium with a characteristic relaxation time of 2.7 min, which is the nuclear T_1 time; see Fig. 3. The technique of measuring the nuclear T_1 via shifted ODMR peak positions was also used by Schreiner et al. for undoped and p-type GaAs structures; their relaxation times ranged from 6.8 to 50.8 min [35]. The shorter relaxation time that we measured is indicative of the stronger electron–nuclear interaction that occurs in n-type samples because of the increased electron localization.

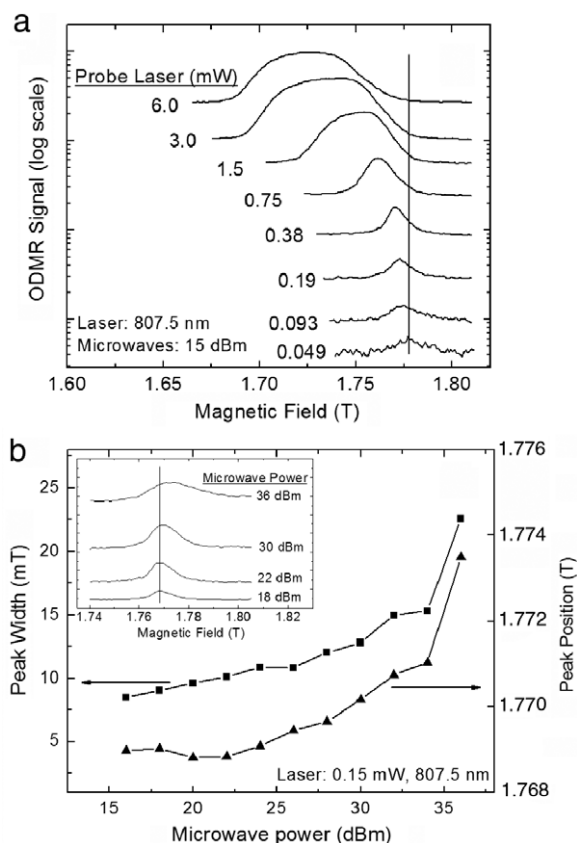


Fig. 2. ODMR of the quantum well sample at 8.6 GHz. When DNP becomes significant, the ODMR peaks typically broaden and shift as is evident at the higher probe laser and microwave powers. (a) Probe laser power dependence. (b) Inset: Microwave power dependence, 1% duty cycle. Main figure: Summary of peak positions and widths versus microwave power.

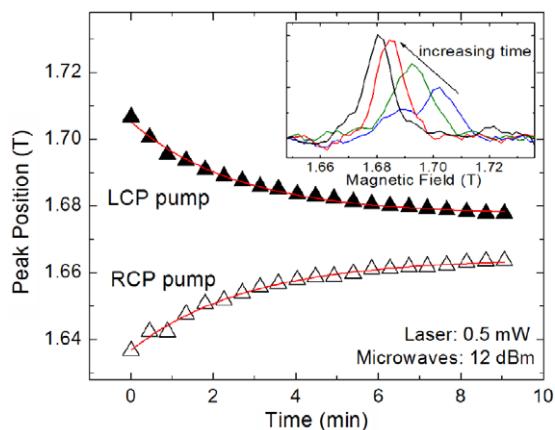


Fig. 3. The ODMR peak positions of the bulk sample at 10.5 GHz, after pumping by a left or right circularly polarized pump beam (labeled LCP and RCP). The peak positions shifted back to equilibrium as the Overhauser field relaxes, providing a time-resolved measure of the nuclear polarization. The characteristic nuclear spin relaxation time (T_1) was 2.7 min. The microwave and light powers were kept low so as not to perturb the nuclear polarization during the field sweeps. Inset: Representative ODMR peaks at various times after LCP pump laser.

By shifting and broadening the electron spin resonance, DNP seriously impacts the manipulation of electron spins. One tactic which has been previously employed by our group and others to reduce the effect of DNP is to continually “reset” the polarization of the nuclear spins via nuclear magnetic resonance (NMR) [18,23,24]. That was done in these experiments by adding a split Helmholtz coil to our resonant cavity. The coil was driven with rf at

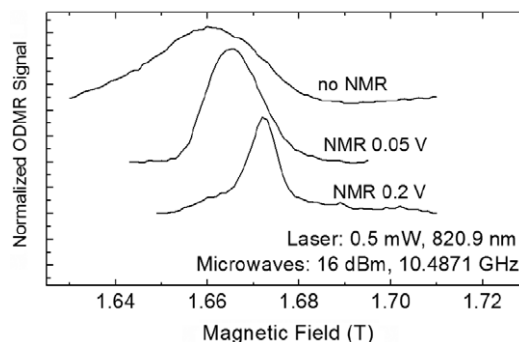


Fig. 4. Bulk sample ODMR peaks with varying NMR powers (function generator voltages shown). With sufficient rf (bottom curve), the dynamic nuclear polarization could be eliminated, shifting the ODMR peak back to its equilibrium position. Peaks are vertically offset for clarity.

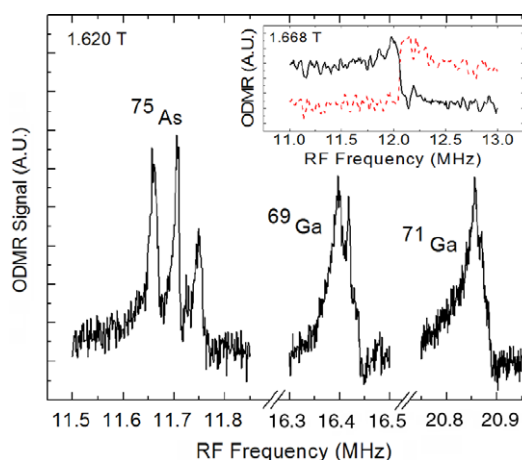


Fig. 5. Changes to the bulk sample Kerr rotation (electron spins) as the rf was scanned. As the nuclear polarization changed at the NMR conditions, the electron spins changed in response. Inset: As the rf was stepped through the ^{75}As resonance either upward (dashed) or downward (solid), the signal abruptly increased as the ODMR peak shifted back to equilibrium.

13, 18, and 23 MHz (at 1.76 T), the resonance frequencies for ^{75}As , ^{69}Ga , and ^{71}Ga , the three nuclear isotopes in our sample. We typically used a function generator to sweep the rf from the lowest to the highest frequency, repeated at about 20 Hz, and an rf amplifier between the function generator and our NMR coil. The nuclei could be depolarized via just a few seconds of applied rf, rapidly relaxing a DNP-shifted ODMR peak back to the equilibrium peak position. In many cases, applied rf should be used to prevent the nuclei from dynamically polarizing in the first place (see Fig. 4), although conditions such as high probe beam and/or microwave powers sometimes made it impossible to completely remove the DNP effects.

We also observed the electron–nuclear interaction via the nuclear resonance, by setting the microwaves and magnetic field to the electron resonance condition and scanning the rf through the three NMR frequencies. This is similar to the traditional optically detected electron–nuclear double resonance, ODENDOR, set-up. As the NMR conditions were met, the nuclear spin polarization changed, which then caused the electron spin polarization to change, and was detectable through a change in the KR of the ODMR peak; see Fig. 5. Note that in the figure the ^{75}As peak shows three sub-peaks, as is expected from the quadrupole splitting. However, the precise peak separation was not reproducible and is likely a complex result of feedback between electron and nuclear spins during the scan; a quadrupole splitting of 50 kHz for ^{75}As would indicate substantial strain [36], for which there is no other evidence in our bulk sample.

The inset to Fig. 5 illustrates another double resonance technique where we first polarized all three nuclei, shifting the ODMR peak away from equilibrium. We “reset” the nuclear polarization of the two Ga nuclei, leaving the ^{75}As nuclei still polarized. We then monitored the ODMR signal while stepping through the ^{75}As NMR frequency. When those nuclei were resonated, the ODMR peak shifted back to the equilibrium field position which we were monitoring, giving an abrupt increase in signal.

4. Conclusions

In summary, we have used Kerr rotation to measure electron and nuclear spin properties in two n-type GaAs samples. We have measured $g = -0.444$ and $T_2^* = 6$ ns for electrons in a bulk sample and $g = -0.346$ and $T_2^* = 9$ ns for a 14 nm quantum well. Dynamic nuclear polarization caused by the electron–nuclear hyperfine interaction was significant – once polarized, the nuclear spins relaxed with a characteristic time of 2.7 min – but was controllable by resonating the three nuclear isotopes.

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