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Spin Lifetime Measurements in MBE-Grown GaAs Epilayers

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Electron spin relaxation times in excess of the localized limit have been measured in MBE n-GaAs layers, with the times depending on the doping concentration. We have optically oriented the electrons in the samples, and measured spin lifetimes via luminescence depolarization in a transverse magnetic field (Hanle effect). The lifetimes thus obtained were 14 and 26 ns for samples nominally doped at 1×10^{15} and 3×10^{15} cm^{-3}, respectively. The dominant dephasing mechanism, which is the hyperfine interaction of localized electrons with lattice nuclei, is discussed. Our results are presented in the context of our larger goal, which is to use resonance techniques for spin measurements and control. In this context, the Hanle spin lifetime measurement is a necessary step to be followed by optically detected magnetic resonance in a longitudinal magnetic field.

Electronic spin in semiconductors is a well-known candidate for quantum computation [1]. In this work, we focus on low temperature spin lifetime measurements in n-type GaAs, with electron localization at donor sites being analogous to the electron localization present in quantum dots.

Long spin lifetimes have been reported for electrons in n-type GaAs. Dzhioev et al. [2] reported a Hanle effect spin lifetime measurement of 42 ns in a 35 μm GaAs layer grown by liquid phase epitaxy, doped with $n = 10^{15}$ cm^{-3}. Kikkawa and Awschalom [3] used time-resolved Faraday rotation to measure a spin lifetime of 130 ns in a bulk GaAs crystal (mechanically thinned to 50 μm), doped with $n = 10^{16}$ cm^{-3}.

Our objective is to measure and manipulate electron spins in n-GaAs. We orient the electronic spin optically; that is, we use polarized light to inject spin-polarized electrons. Comparing optical orientation of electrons in semiconductors to optical orientation of atoms in a gas, one may say that working with the spin of the electrons in n-type semiconductors is analogous to optically pumping the gas atoms in the ground state [4], whereas orienting the spin of minority electrons in p-type semiconductors is analogous to the orientation of excited atoms in a gas. Thus, in n-type semiconductors the doped electrons provide a means for extending the spin lifetime well beyond the radiative recombination time.

The main focus of this paper is the results of optical orientation and optical detection of spins. Spin lifetimes of 26 and 14 ns were observed. Spin manipulation through microwave techniques is also being explored, and some comments on those efforts will be given at the end of the paper.

We report measurements on GaAs samples grown by molecular beam epitaxy (MBE) at the Naval Research Laboratory in Washington, D.C. The sample epilayer structure is as follows: (starting with a semi-insulating GaAs substrate) a 100 Å GaAs layer, a 100 Å

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AlAs layer, a 2000 Å undoped GaAs buffer layer, a 1000 Å n-$Al_{0.3}Ga_{0.7}As$ layer, a 1000 Å undoped $Al_{0.3}Ga_{0.7}As$ layer, the 1 μm n-GaAs active layer, a 1000 Å undoped $Al_{0.3}Ga_{0.7}As$ layer, a 1000 Å n-$Al_{0.3}Ga_{0.7}As$ layer, and a 50 Å GaAs cap. The doping levels of the two doped AlGaAs layers were 7×10^{16} and $1.5 \times 10^{16}\,cm^{-3}$, respectively; they were chosen to minimize band-bending, and to place the Fermi energy of the active layer at the GaAs conduction band. Surrounding the active layer with AlGaAs layers has two effects: (1) it greatly enhances the photoluminescence (PL) due to elimination of surface channels for non-radiative recombination, and (2) it eliminates spin diffusion into the bulk. Undoped AlGaAs layers were used as immediate active-layer neighbors in order to prevent carrier transfer from the barriers into the active layer. The AlAs layer near the substrate was grown as a potential etch-stop so that the heterostructure may be removed from the substrate if desired, although all of the experiments described in this work have been performed with the substrate intact.

Three doping levels of the 1 μm GaAs layers were chosen: nominally undoped, $1 \times 10^{15}\,cm^{-3}$, and $3 \times 10^{15}\,cm^{-3}$. The latter two samples will be referred to as the "1E15" and "3E15" samples, respectively. The PL of all three samples is very strong. Free and donor-bound exciton PL peaks are visible at low temperature and low excitation power densities, with the bound exciton dominant in the doped samples and the free exciton dominant in the undoped sample. The 1.5 K PL is shown in Fig. 1, together with another, high quality, unbounded MBE thick layer sample with a similar doping level. With adjustments to excitation power density, the ratios between free and bound excitons in the samples can be varied. The PL of all three heterostructures is much stronger than that of the uncapped sample. The PL data shown here was excited with an unfocused HeNe laser at 633 nm and 3 mW.

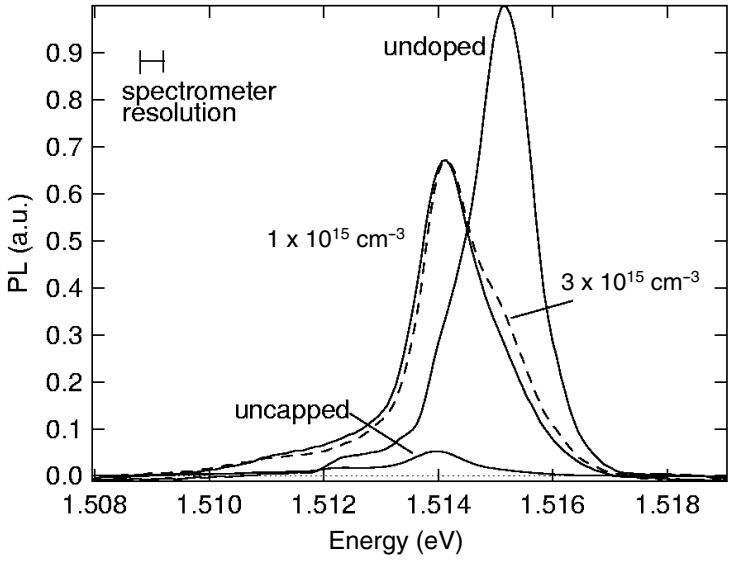

Fig. 1. Photoluminescence of four GaAs samples: samples 1E15 and 3E15, the undoped control sample, and a high quality doped uncapped sample. PL is apparent from both the free (higher energy) and bound (lower energy) excitons

The carrier concentrations and mobilities of the doped samples have been measured by the Hall effect. The room temperature carrier concentrations were $n = 1.6 \times 10^{15}$ cm^{-3} and 3.8×10^{15} cm^{-3} for samples 1E15 and 3E15, respectively. The 77 K mobilities were 43000 cm^2/Vs and 31000 cm^2/Vs, and estimations of the compensation factor N_A/N_D are 0.5 and 0.4 for the two samples.

Circularly polarized light was used to inject spin polarized electrons in the samples, through the well established technique of optical orientation, using photon energy between the band-gap and the split-off band [4]. In typical III–V semiconductors, electrons in the conduction band may be characterized with angular momentum $\pm 1/2$, holes in the heavy hole valence band $\pm 3/2$, and holes in the light hole valence band as $\pm 1/2$. Thus, a photon with angular momentum +1 can inject an electron with either spin down (if an electron is removed from the heavy hole valence band) or spin up (if an electron is removed from the light hole valence band). If the transition probabilities were the same for the heavy and light hole valence bands, no electronic spin polarization would be produced. However, the heavy hole transition is favored over the light hole, in a 3 to 1 ratio. With 3 spin downs produced for every spin up, a maximum of 50% electronic polarization may be obtained. The same selection rules apply to emission, so a maximum of 25% polarization may be obtained in the emitted light. In practice, the observed polarizations are smaller than that.

Our experiment consisted of optical excitation and optical detection in a cryogenic magnet. For excitation, we used a New Focus external cavity tunable diode laser, operating at 785 nm. A photo-elastic modulator (PEM) was used as a quarter wave plate to switch the polarization between σ^+ (+1 angular momentum) and σ^- (−1 angular momentum) at 20 kHz. The samples were placed in an Oxford Instruments liquid helium cryostat integral with a split-pair superconducting magnet in transverse field geometry (Voigt). Photoluminescence for the samples was collected, passed through a circular polarization analyzer (composed of a quarter wave retarder and a linear polarizer), and then spectrally dispersed with a SPEX 1680 0.22m double spectrometer. A photomultiplier tube connected with a two-channel photon counter was used to measure the signal. The photon counter operated synchronously with the PEM to detect the results of σ^+ and σ^- excitation.

In n-type samples, injection of spin polarized photo-electrons gives rise to a polarization of the doped electrons [5]. This is easy to understand if one assumes that recombination is largely spin-independent. In that case, spin-polarized electrons replace the initially unpolarized, doped electrons, as some of the latter form excitons and combine radiatively with the photoexcited holes. As pumping power density is increased, more spin-polarized electrons are injected, and the polarization of the doped electrons, and hence of the emitted PL, increases. This is seen for samples 1E15 and 3E15 in Fig. 2.

The polarization thus produced and detected can be affected by applying a transverse magnetic field. This is the Hanle effect. As spin polarized electrons experience the transverse field, they begin to precess. This decreases the spin component along the axis of detection. A larger field produces more precession on average before recombination, so the observed luminescence decreases with field. If the spin lifetime is long, more spins will survive to recombine at a precessed direction, and the luminescence polarization will be reduced by more than in the short lifetime case. Thus, the rate at which the polarization falls off with field gives information on both the spin lifetime and the electron lifetime.

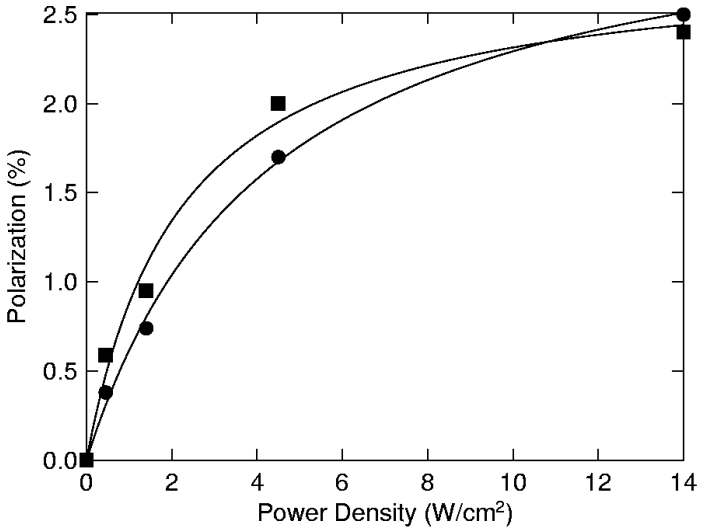

Fig. 2. PL polarization percentage vs. excitation power density for samples 1E15 (squares) and samples 3E15 (circles). The data has been fit according to Eqs. (1) and (2): $S = S_0\ (1 + n/G\tau_S)^{-1}$. One may estimate the relation between G and the laser power density P: $G/P = (1 - \mathrm{R})\ \alpha/E_{\mathrm{photon}}$, where R is the reflectivity ($\sim$0.3), and α is the absorption coefficient ($\sim 10^4\,\mathrm{cm}^{-1}$). Using the values of τ_S obtained from Fig. 3, the parameters obtained from the fit are $S_0 = 2.9\%$, $n = 8.7 \times 10^{14}\,\mathrm{cm}^{-3}$ (sample 1E15), and $S_0 = 3.3\%$, $n = 3.2 \times 10^{15}\,\mathrm{cm}^{-3}$ (sample 3E15). The PL was detected at the free exciton peak, at 1.5 K, and was excited at 785 nm

Measurements of the Hanle effect in samples 1E15 and 3E15 are displayed in Fig. 3 for various excitation power densities. For comparison purposes, the curves have been normalized to the same maximum polarization. The polarization was detected in the PL of the free exciton line. That the free exciton PL reflects the doped electron spin lifetime, is explained by Vekua et al. [5] by noting that the lifetime of oriented electrons in the exciton state is much less than the time between generation of oriented spin and incorporation into an exciton.

The depolarization curves are well fit by Lorentzians. This is in contrast to typical Hanle curves for n-type semiconductors, which have a non-Lorentzian shape characterized by a large polarization tail for high values of B [4]. This is evidence of a lack of spin diffusion, which is a result of the confining AlGaAs layers. In our samples, the spin orientation is homogeneous throughout the layer thickness.

In the absence of spin diffusion or other spin relay mechanisms, the steady-state situation in n-type semiconductors may employ the standard optical orientation and Hanle effect theory. The zero-field average electron spin depends on spin relaxation time (τ_s) and electron lifetime (τ_J) in this manner:

$$S(B = 0) = S_0\ (1 + \tau_J/\tau_S)^{-1}\,, \tag{1}$$

where S_0 is a constant. The steady-state electron lifetime (τ_J) depends on the electron concentration (n) and the electron–hole pair generation rate (G, number of pairs generated per unit volume per unit time):

$$\tau_J = n/G\,. \tag{2}$$

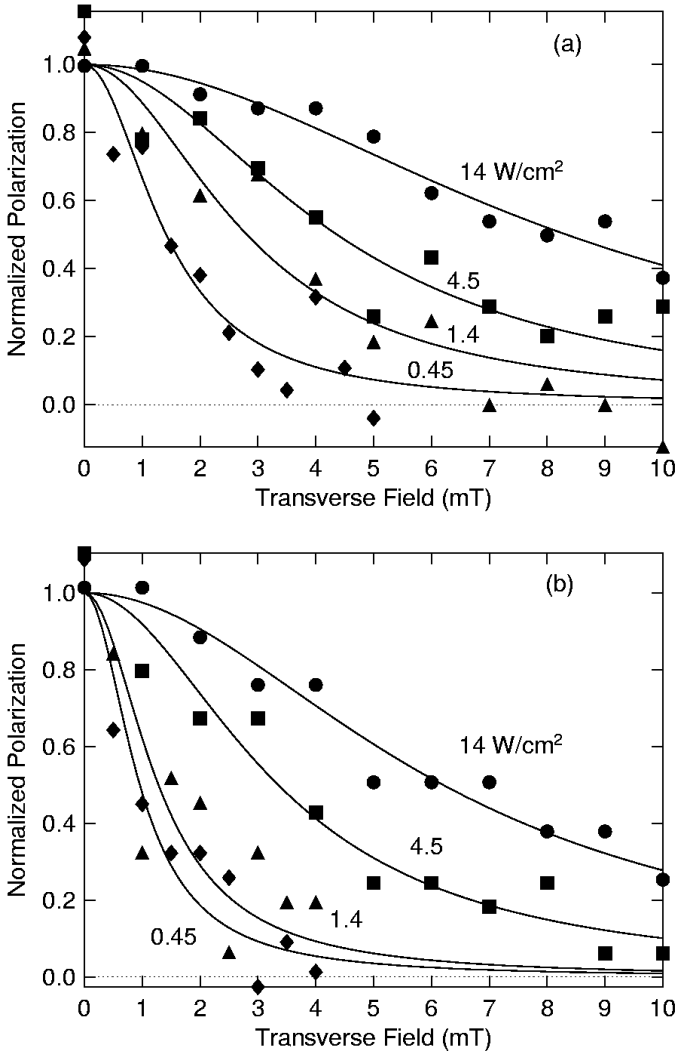

Fig. 3. Depolarization of the PL in the presence of a transverse magnetic field (Hanle effect), for various excitation power densities, in a) sample 1E15 and b) sample 3E15, fit according to Eq. (3). The low power extrapolation of the Hanle linewidth yields the spin relaxation time, which is 14 ns and 26 ns for samples 1E15 and 3E15, respectively

In a transverse field the electron spin is depolarized with a Lorentzian dependence:

$$S(B) = S(0)/(1 + (\Omega T_S)^2) \,, \tag{3}$$

where Ω is the precession frequency: $\Omega = g\mu_B B/\mathrm{h}$, and T_S (the measured spin lifetime) depends on both the electron lifetime and the spin relaxation time: $1/T_S = 1/\tau_J + 1/\tau_S$.

The measured spin lifetime T_S is thus obtained from the Hanle halfwidth: $B_{1/2} = \hbar/(g\mu_B T_S)$. Since G is proportional to the excitation power density, the steady state equations show that the inverse measured lifetime plotted vs. power density should be

a straight line: $1/T_S = G/n + 1/\tau_S$. The low power limit of the linear fit yields the true spin relaxation time (τ_S), which was for our doped samples 14 ns and 26 ns (for samples 1E15 and 3E15, respectively). The point at which $\tau_J = \tau_S$ is the excitation power density where the inverse lifetime is twice the low power limit, and was approximately 3.8 and 2.6 W/cm^2 for the two samples. The relaxation time of the undoped sample was difficult to quantify, due to a much smaller PL polarization, but was approximately an order of magnitude shorter than in the doped samples.

The main dephasing mechanism at low temperature, low excitation power density, and low doping density is due to the contact hyperfine interaction between electrons and the lattice nuclei [6]. The hyperfine interaction with a single nucleus produces an effective field; the average field experienced by each donor electron is a sum of the fields produced by the more than 10^5 nuclei within the donor Bohr radius. This average effective nuclear field is different for each donor site, and the electrons' precession in their individual fields leads to decoherence within the ensemble of electrons. If there is some correlation between electrons at different donor sites, then the decoherence is reduced. This may be the case, for example, if electrons can hop from one site to another. Dyakonov and Perel derive a spin relaxation time for this case: $\tau_S^{-1} = 2/3\ \omega_c^2\ \gamma^{-1}$, where ω_c is the precession frequency in the nuclear field (they estimate it to be about 5×10^8 s^{-1}), and γ^{-1} is the correlation time (i.e., the hopping time). If the electrons are completely localized, then the correlation time becomes the time of precession, and the limit of spin lifetime in this regime thus becomes about 3 ns. A more recent estimate of spin relaxation in this regime was made by Merkulov et al. [7]: a relaxation time of 1 ns is given for the case of GaAs quantum dots containing 10^5 nuclei.

The spin relaxation time of the undoped sample is consistent with these estimates; however, both of the doped samples displayed spin lifetimes in excess of this limit. This shows that there is some correlation between donors in those samples. In an estimation of the correlation time dependence on donor concentration, Kavokin [8] predicted that spin lifetimes in lightly doped samples may be increased, from the localized limit up to around 200 ns (at which point other relaxation mechanisms become dominant), with the maximum being found at $n \sim 5 \times 10^{15}$.

The lifetime measured for our 3E15 sample was well below Kavokin's estimate for that concentration ($\sim$100 ns). There are a few possible explanations for this. One thing to consider is compensation from unwanted acceptors may play a role in reducing the hopping rate (i.e., increasing the correlation time). An effect from compensation has been seen in p-type samples, where the spin relaxation time in the low-temperature, localized electron regime, was shorter for more compensated samples [9]. GaAs samples grown by other methods may prove to have less unwanted impurities and possibly a longer relaxation time. For example, GaAs layers grown by liquid phase epitaxy with carrier concentrations as low as 10^{12} cm^{-3} and having 77 K mobilities of 240000 cm^2/Vs have been reported [10]. In comparison, nominally undoped GaAs layers grown by metal–organic chemical vapor deposition (MOCVD) [11], and MBE typically have carrier concentrations in the range of 10^{14} cm^{-3} and mobilities in the range of 100000 cm^2/Vs. In particular, an undoped sample (a single thick GaAs layer) grown at the Naval Research Laboratory shortly after our heterostructures, has an electron concentration of 9×10^{13} cm^{-3} and a mobility of 140000 cm^2/Vs, and likely reflects both the quality of the samples discussed in this work and the doping level of our nominally undoped sample. MBE, it should be added, does offer some unique advantages over other

growth techniques; for example, interface fluctuation quantum dots grown by MBE have long been produced and studied at NRL [12]. This easy extension into quantum dots is a primary reason why we have focused on MBE GaAs.

The structure of the heterostructures themselves may also be playing a role in determining the spin lifetime. For example, in a 100 nm MBE GaAs layer, similarly surrounded by AlGaAs layers, Dzhioev et al. [13] saw large effects on the spin relaxation time due to changes in the correlation time caused by electrons transferred from the barrier mediating spin exchange between donors. In that work, the correlation time was greatly reduced, and a relaxation time of 290 ns was measured for an n-type GaAs epilayer doped around $10^{14}\,\mathrm{cm}^{-3}$. Thus, in that case, the heterostructure played a vital role in greatly *extending* the lifetime. Although our structure has been designed to minimize effects from the non-active layers, it is conceivable that a reduction in lifetime could come from effects such as band-bending.

One final possibility considered here is that the excitation conditions may be limiting the lifetime. It has been recently observed, by some of our collaborators at the Ioffe Institute in St. Petersburg, that relaxation times of electrons in n-type GaAs may be reduced significantly when the excitation light is above the band-gap. When excitation is between the band-edge and the exciton resonance, longer relaxation times are observed. This may be due to production of faster-relaxing free carriers in the above band-gap case, and to their interaction with the localized electrons that we desire to probe [14]. Investigations are underway to test the excitation energy dependence of the lifetimes in these samples.

Along with our optical injection and measurement of electronic spin in GaAs, we have also made experimental efforts at spin control with microwave and rf fields. The 10–20 ns spin relaxation times that we have measured are long enough to enter the regime where optically detected magnetic resonance (ODMR) becomes possible. The optical detection can conceivably be done in a variety of ways. For example, one can detect the polarization of PL emission, similar to the experiments described in this work. On the other hand, one can also probe the spin orientation of free electrons via absorption effects such as Faraday rotation or magnetic circular dichroism [15].

An important consideration is the frequency at which to apply the microwave/rf field. Low frequencies (100–500 MHz), with the corresponding low resonant magnetic fields (20–100 mT), have the advantage of reducing the effects of g-broadening. Some estimates and effects of g-broadening have been reported by Kikkawa and Awschalom [3], who estimate $\Delta g < 0.005$ in a $10^{16}\,\mathrm{cm}^{-3}$ sample, and by Seck et al. [16], who used conventional electron spin resonance on n-type GaAs ($\sim 10^{15}\,\mathrm{cm}^{-3}$) to measure the resonance linewidth of about 50 mT at high fields ($\sim$9 T). Seck's width is much larger than the 2–4 mT linewidth we expect to see in these samples at low fields (twice the Hanle width). Low fields also make an easier extension into quantum wells and quantum dots, since the GaAs g-factor has a large variation with quantum well width [17]. On the other hand, higher fields (larger than $\sim$1–2 T), combined with low temperatures, can produce a thermal polarization of the electrons larger than that obtainable with optical methods at low fields, and may eliminate the necessity of optically orienting the electrons. Unsuccessful attempts at ODMR have been made using many of the above parameters (low field and high field, thermal polarization and optical polarization, photoluminescence and Faraday rotation). From the considerations outlined above, the current plan is to concentrate on low field resonance for the present.

In summary, we have grown high quality GaAs epilayers by MBE. The spin relaxation lifetimes have been measured at 14 and 26 ns for two doped samples, and are in excess of the completely localized limit. There may be an opportunity to see longer lifetimes in other samples, if the correlation time between electrons on donors can be shortened. However, extensions into quantum dots may push us toward the localized (long-correlation time) limit. In that case, longer lifetimes may possibly be obtained by minimizing the effects of nuclear hyperfine fields through polarizing the nuclei. Combined extensions into quantum dots and microwave resonance techniques are particularly appealing because it has been noted that this type of dephasing in quantum dots arises from inhomogeneity and may be compensated for using the spin echo technique [7]. The lifetimes observed in these samples should be sufficient to allow optically detected magnetic resonance, and such experiments are proceeding.

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