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Spin coherence and dephasing in GaN

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Time-resolved Faraday rotation is used to measure electron spin coherence in *n*-type GaN epilayers. Despite densities of charged threading dislocations of $\sim 5\times 10^8\,\mathrm{cm}^{-2}$, this coherence yields spin lifetimes of ~20 ns at $T=5$ K, and persists to room temperature. Spin dephasing is investigated in the vicinity of the metal-insulator transition. The dependence on both magnetic field and temperature is found to be qualitatively similar to previous studies in *n*-type GaAs, suggesting a common origin for spin relaxation in these systems.

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There is an emerging interest in the exploitation of electron spins in semiconductors to realize all-semiconductor electronic devices such as spin diodes[1] or nonvolatile spin memory.[2] The successful realization of these devices necessitates the ability to inject and preserve spin information over practical device length and time scales. Recent observations revealing extremely long spin coherence times for optically injected spins in nonmagnetic semiconductors,[3] as well as spin transport over macroscopic distances[4] and through semiconductor heterointerfaces,[5] has additionally raised the possibility that these spin coherent properties may eventually enable quantum computational operations in solid state systems.[6] In this context, the III-V semiconductor GaN and its related ternary compounds are intriguing in that they combine a high density of charged threading dislocations (typically $10^8-10^{10}\,\mathrm{cm}^{-2}$)[7,8] with high optical quality.[9,10] As a result, it is possible to optically create and monitor coherent electronic spin states in a system where crystalline defects may play an important role in spin dephasing and decoherence mechanisms.

Here we use the optical pump/probe technique of time-resolved Faraday rotation (TRFR)[11] to systematically investigate spin coherence in a series of *n*-type GaN epilayers grown by metal-organic chemical vapor deposition (MOCVD), with carrier concentrations ranging from the insulating to the metallic regime. Despite the dislocation density in these samples, the data reveal a triexponential decay with surprisingly long spin lifetimes of up to ~20 ns observed at $T=5$ K. The spin coherence persists to room temperature, and exhibits similar behavior to other III-V and II-VI semiconductors grown by molecular-beam epitaxy with many orders of magnitude fewer defects,[3,11] suggesting that the spin degree of freedom is largely insensitive to momentum scattering from the charged threading dislocations.

Si-doped GaN epilayers 3 μm in thickness are grown on *c*-axis sapphire substrates by MOCVD.[12] Room temperature electron carrier densities n of two unintentionally doped samples are 3.5×10^{16} and $1\times 10^{17}\,\mathrm{cm}^{-3}$, with carrier activation energies of 14.2 and 11.1 meV, respectively, demonstrating insulating behavior. A third Si-doped sample with $n=9\times 10^{17}\,\mathrm{cm}^{-3}$ is metallic, as indicated by the absence of thermally activated behavior in both carrier density and resistivity at low temperatures. All samples have a threading dislocation density of $\sim 5\times 10^8\,\mathrm{cm}^{-2}$, as determined by atomic force microscopy.[13] The importance of scattering from the charged threading dislocations in determining the mobility in GaN (typically $\sim 500\,\mathrm{cm}^2\,\mathrm{V}^{-1}\,\mathrm{s}^{-1}$ at room temperature) has been extensively investigated both theoretically[14] and experimentally.[15] This scattering was found to be relevant only at low carrier densities where charge screening is not effective, while at higher carrier densities conventional ionized impurity scattering is predominant. Given the sample parameters listed above, as well as an observed monotonic decrease in mobility with increasing carrier density (not shown), we conclude that these samples are in the high carrier density scattering regime.

For time-resolved measurements both pump and probe pulses are derived from the frequency-doubled output of a mode-locked Ti:sapphire laser with a temporal resolution of 120 fs at $E=3.481$ eV. A mechanical delay line generates time delays between pump and probe pulses of up to 3 ns. For TRFR the pump pulse is circularly polarized and focused normal to the sample surface to a spot size of 75-μm diameter. The Larmor precession of the optically injected carrier spins in the presence of a magnetic field oriented perpendicular to the surface normal (Voigt geometry) is subsequently monitored as oscillatory changes in the Faraday rotation of the linearly polarized probe pulse. Although the carrier density differs by more than an order of magnitude between samples, we do not observe a significant shift of the optical absorption edge within the energy width of the laser (the full width at half maximum is 16 meV). The laser energy is therefore kept constant for all three samples with excitation densities more than two orders of magnitude less than the carrier concentration.

Figure 1(a) shows the oscillatory Faraday response at $T=5$ K in a transverse magnetic field of $B=1.5$ T for GaN samples of different carrier concentrations. The oscillations appear at the Larmor frequency ω_L directly proportional to the energy splitting, $\Delta E=\hbar\omega_L$, between the two spin eigenstates parallel and antiparallel with the field. This frequency is a direct measure of the electron g-factor, $g=\hbar\omega_L/\mu_B H$ (μ_B is the Bohr magneton, and H the magnetic field). Fast Fourier transforms of the spectra in Fig. 1(a) give $g=1.94$

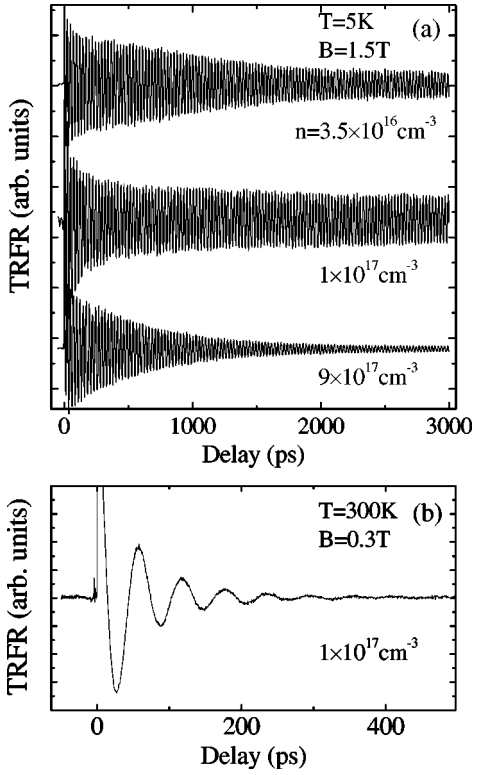

FIG. 1. (a) Time-resolved Faraday rotation (TRFR) for *n*-doped GaN epilayers at $T=5$ K and $B=1.5$ T. Carrier densities n are room-temperature values. (b) TRFR taken at a magnetic field of 0.3 T and a temperature of 300 K in the sample from (a) with $n=1\times10^{17}\,\mathrm{cm}^{-3}$.

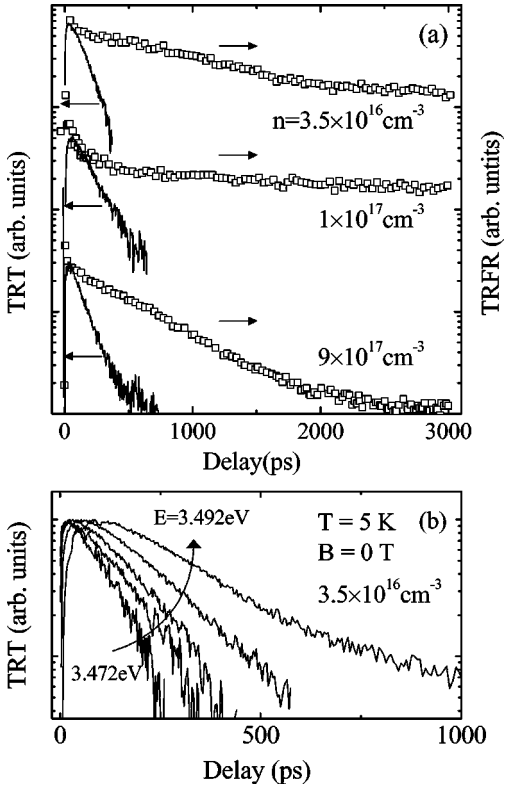

FIG. 2. (a) Envelopes of the data shown in Fig. 1(a) (open squares) superimposed with zero-field time-resolved transmittance (TRT) (lines) plotted on a semilog scale. All data were taken at $T=5$ K and at a laser energy $E=3.481$ eV. (b) TRT for the sample with doping $n=3.5\times10^{16}\,\mathrm{cm}^{-3}$ at laser energies of 3.492, 3.487, 3.481, 3.477, and 3.472 eV at $T=5$ K. Curves are plotted on a semilog scale, and are normalized to their maximum value.

±0.01 independent of doping, which is consistent with values from electron spin resonance measurements and reflects the small spin-orbit coupling in GaN.[16] The spin coherence persists to room temperature [Fig. 1(b)], and yields a spin lifetime of $\sim$35 ps.

An intriguing feature of the spin coherence is the nonmonotonic variation of its decay with carrier concentration, with the slowest decay observed for the sample with doping in the vicinity of the metal-insulator transition (MIT). In order to more easily visualize this decay, we plot the envelopes of the TRFR data on a semilog scale, showing a multicomponent exponential decay of the spin coherence [Fig. 2(a)]. On the same semilog scale we include measurements of the time-resolved transmission (TRT), which is a measure of the carrier lifetime, taken at the same temperature and laser energy. The decay of the TRT signal closely matches that of the initial rapid decay of the TRFR (τ_1), which we thus conclude is limited by electron-hole recombination. As recombination is known to be strongly energy dependent, and as our laser bandwidth is 16 meV, we investigate the evolution of the TRT in the vicinity of the band edge for all samples. Representative data are shown in Fig. 2(b). We find that the carrier lifetime varies with energy, but does not exceed 400 ps.[17] This supports the notion that carrier recombination does not play a role in the spin-dephasing mechanisms at longer times.

To further explore the spin dephasing at longer time delay, we focus on the magnetic-field dependence of the TRFR. Figure 3(a) shows the TRFR at selected fields for the sample with $n=3.5\times10^{16}\,\mathrm{cm}^{-3}$. There are two primary effects of increasing the magnetic field: the spin precession frequency increases, and the spin coherence decays more rapidly. The former can be understood from the linear dependence of the Larmor frequency on an applied field, while the latter indicates a change in the spin-dephasing time. To determine the changes in dephasing time directly, Fig. 3(b) shows several TRFR envelopes from the samples with lowest and highest carrier concentrations. As is seen in the doped sample, the increasing magnetic field reveals two distinct decays at long-time delay. While the shorter of the two becomes more rapid with increasing field, the longer appears unchanged.

The TRFR envelopes can be fit by a simple double-exponential decay of the form $\Sigma_{i=2,3}A_i e^{-\Delta t/\tau_{(i)}}$ for delays larger than 400 ps. However, a reliable value for the long-lived decay lifetime τ_3 is prohibited by the length of our delay line, and by the large amplitude of A_2. We provide a lower limit for the lifetime of $\tau_3\geqslant8$ ns from fitting and an upper limit of $\sim$20 ns from resonant spin amplification measurements.[3] In fact, allowing τ_3 to vary from 8 ns to infinity does not alter the value of τ_2 or diminish the quality of the fits from 400 ps to 3 ns. Thus we set τ_3 to infinity, and

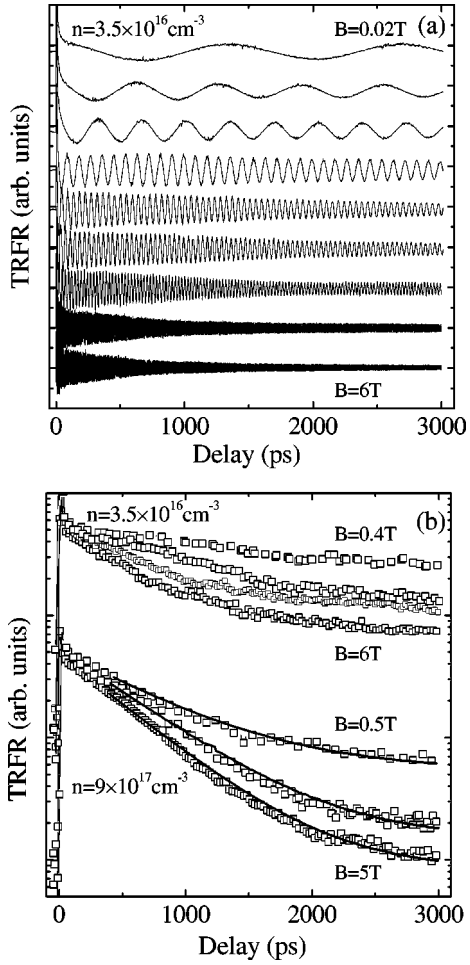

FIG. 3. (a) TRFR rotation taken at $T=5$ K for the sample with $n=3.5\times10^{16}\,\mathrm{cm}^{-3}$ at magnetic fields of $B=0.02$, 0.05, 0.1, 0.4, 0.8, 1.0, 1.5, 4, and 6 T (from top to bottom). (b) Selected envelopes plotted on a semilog scale for samples with $n=3.5\times10^{16}\,\mathrm{cm}^{-3}$ ($B=0.4$, 1.5, 4, and 6 T) and $n=9\times10^{17}\,\mathrm{cm}^{-3}$ ($B=0.5$, 1.5, and 5 T). Solid lines are fits to the data (see text).

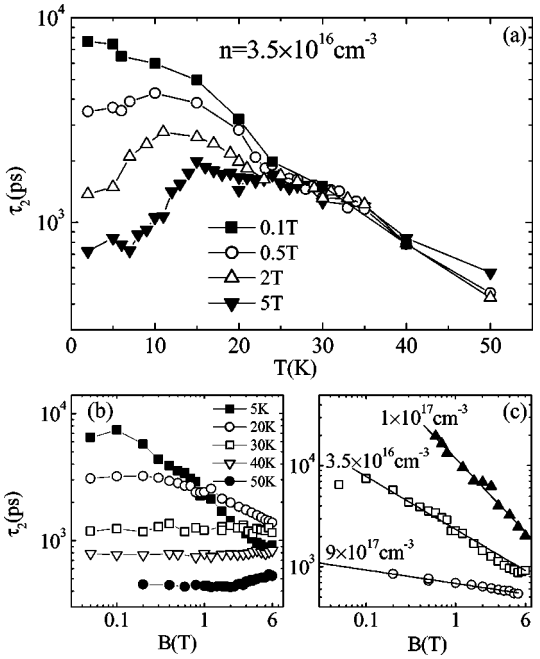

FIG. 4. (a) Spin scattering time τ_2 vs temperature at various fields for the sample with $n=3.5\times10^{16}\,\mathrm{cm}^{-3}$. (b) The magnetic-field dependence of τ_2 for the same sample. (c) τ_2 vs magnetic field on a log-log plot at $T=5$ K for all samples. Solid lines are power-law fits to $\tau_2\propto B^{-\alpha}$, yielding $\alpha=0.52$ (open squares), 0.93 (filled triangles), and 0.13 (open circles).

effectively have a three-parameter fit, with the resulting curves shown as solid lines in Fig. 3(b).

According to Elliot-Yafet[18] spin-scattering theory, momentum scattering plays a crucial role in the dephasing, and scattering from the charged threading dislocations should lead to spin decoherence or dephasing via local spin-orbit interactions. Although momentum scattering is strongly enhanced by the presence of these defects, as seen in the carrier mobility,[14] the measured spin coherence appears to be robust to their presence. A possible explanation for this surprising behavior lies in the fact that the valence-band spin-orbit coupling is ~100 times weaker than in GaAs,[16] resulting in a decoupling of momentum- and spin-scattering processes.

The temperature dependence of τ_2 is shown in Fig. 4(a) at a variety of magnetic fields for the sample with lowest doping. At high temperatures ($T>25$ K), τ_2 decreases monotonically with increasing temperature. Further, τ_2 is either constant or increasing with increasing magnetic field [Fig. 4(b)] in this temperature range. This dependence is expected, and agrees qualitatively with the existence of anisotropic internal magnetic fields such as caused by either the noncentrosymmetric crystalline structure (D'yakonov-Perel)[19] or the strong spontaneous piezoelectric fields (Rashba effect)[20] observed in GaN. Below $T=25$ K this behavior changes qualitatively. Whereas at low magnetic fields τ_2 still increases with decreasing temperature, at higher fields it goes through a field-dependent maximum and subsequently decreases. This indicates the onset of an additional field- and temperature-dependent spin scattering (or dephasing) mechanism. The same magnetic field induced scattering mechanism is observed in all samples, and is depicted on a log-log scale in Fig. 4(c) at $T=5$ K, revealing a simple power law behavior, $\tau_2\propto B^{-\alpha}$, with α largest close to the MIT.

Summarizing the experimental features of τ_2, we find that they are all in qualitative agreement with results obtained in GaAs:[3] (i) the zero-field spin lifetime is longest at intermediate doping, i.e. in the vicinity of the MIT. (ii) An additional spin-dephasing mechanism evolves at low temperatures and nonzero magnetic field. It is enhanced with *decreasing* temperature and *increasing* field. (iii) This field-induced suppression of the spin lifetime is strongest in the vicinity of the MIT.

In order to understand this additional spin scattering mechanism, we consider possible origins for the dephasing of τ_2. Field-induced spin relaxation is known to arise from inhomogeneous dephasing due to a Gaussian distribution of g factors, resulting in a simple inverse relationship between τ_2 and $B(\alpha=1)$.[21] This is not found experimentally in all samples. Additionally, spin-scattering mechanisms such as the Elliot-Yafet or D'yakanov-Perel mechanisms are un-

likely to be responsible for the non-monotonic behavior of the spin lifetime, as both depend on the mobility, which varies monotonically through the MIT. Alternatively, the divergence of the carrier localization length at the MIT (Ref. 22) indicates some connection between spin dephasing and the carrier length scale. This suggests the existence of spin scattering mechanisms whose strength is sensitive to the carrier localization length, such as nuclear hyperfine coupling[23] or anisotropic electron spin exchange.[24]

In conclusion, we have observed spin coherence in *n*-type MOCVD-grown GaN epilayers extending to long times and high temperatures despite the presence of several orders of magnitude greater defect densities than in previously measured semiconductors such as *n*-GaAs[3] or *n*-ZnSe.[5] A similar spin coherence was also observed in $In_xGa_{1-x}N$, suggesting that it is robust to high defect densities in a variety of materials.[25] These findings are promising for the potential development of optoelectronic devices that rely on the coherent properties of the electron spin.

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