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Room temperature electron spin relaxation in GaInNAs multiple quantum wells at 1.3 μm

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The authors report a direct measurement of electron spin relaxation in GaInNAs semiconductor multiple quantum wells at room temperature. Multiple quantum wells of widths 5.8, 7, and 8 nm exhibiting excitonic absorption around 1.3 μm have been studied. Spin relaxation times were found to increase with well width in the range of 77–133 ps. The spin relaxation time dependence on first electron confinement energy suggests the Elliot-Yafet mechanism [A. Tackeuchi et al., Physica B 272, 318 (1999)] as the dominant relaxation process. © 2006 American Institute of Physics. [DOI: 10.1063/1.2396901]

The dilute nitride alloy GaInNAs has great potential for optoelectronic applications. The bowing parameter in N-containing alloys is unusually large; substitution of nitrogen for arsenic in Ga1−xInxNAs1−y decreases the band gap energy by ~0.1 eV per percent of N for x<3%.1 The emission wavelength of GaInNAs-based devices can thus be tailored to optical fibers communication at 1.3–1.5 μm while staying lattice matched to GaAs. This has advantages over InP-based systems operating in the same wavelength range, including realization of devices based on the use of high refractive index contrast AlGaaS/GaAs Bragg reflectors. Stronger electron confinement in quantum wells is also expected in the case of nitrides, due to the larger conduction-band offset ratio of GaInNAs/GaAs compared to GaInAsP.2

Spin relaxation has been studied at room temperature (RT) in a range of low-dimensional III-V semiconductor materials, particularly GaAs,3,4 InGaAs5 InGaAsP,5 and more recently, InGaN.7 The unusual band structure of narrow band gap dilute nitrides may have implications for electron spin relaxation, yet to date little work investigating spin relaxation mechanisms in these materials has been published. In this letter, we present optical time-resolved measurements of electron spin relaxation as a function of well width in GaInNAs multiple quantum wells (MQWs) at communication wavelengths.

A series of three undoped MQW samples was studied, each comprising five periods of Ga0.653In0.347N0.0124As0.9876 separated by 20 nm GaAs barriers grown on a 001 n-GaAs substrate by molecular beam epitaxy, with samples A, B, and C having nominal well widths of 5.8, 7, and 8 nm, respectively. Pump and probe pulses of 1 ps duration and 82 MHz repetition rate were produced by a Ti:sapphire-pumped optical parametric oscillator tuned to wavelengths close to the respective peaks of the heavy hole excitonic absorptions, as shown in Fig. 1. Quarter wave plates were used to create circular polarization. 122 pJ pump pulses generated an estimated carrier density of 1011 cm−2, and were spatially overlapped with a weak probe at the sample using a focusing lens. The pump beam was chopped, and the modulation transferred to the probe detected using phase-sensitive detection.

Figure 2 shows results for sample B for same circular polarization (SCP) and opposite circular polarization (OCP) of the probe relative to the pump. The difference, SCP-OCP, gives the spin relaxation time and a longer decay due to carrier recombinaton. We see that electron spin relaxation times (τe) are on the same order as the carrier recombinatn in the samples studied here. The change in probe transmission is therefore also dependent on carrier recombinaton. The nonzero transmission of the OCP probe at zero time delay can be attributed to the screening and broadening components of excitonic saturation.

Due to the lifting of the degeneracy of light and heavy hole energies at k=0 in quantum wells, the selection rules for optical transitions allow excitation of 100% spin-polarized electrons using circularly polarized light resonant with the heavy hole exciton. Electronic spin polarization is defined as

\[ P = (N^+ - N^-)/(N^+ + N^-), \]

where \( N^+ \) and \( N^- \) represent populations of spin-up and spin-down photogenerated carriers, respectively. Upon excitation, the spin states of these carriers relax to equilibrium producing equal spin-up and spin-down populations according to the following rate equations:

![Image of linear absorption spectra showing the peak of the heavy hole excitonic absorption for samples A (1.26 μm), B (1.29 μm), and C (1.30 μm), of quantum well widths 5.8, 7, and 8 nm, respectively.](image-url)
is effectively described by Eq. (2) in the spin-up state, and this state is then probed by a SCP pulse. If a circularly polarized pump pulse generates carriers with time delay for sample B when probe is same (SCP) and opposite (OCP) circularly polarized relative to the pump.

\[
\frac{dN^+}{dt} = -\frac{N^+}{\tau_e} + \frac{N^+}{\tau_s},
\]

(2)

where \(\tau_e\) is the recombination time and \(\tau_s\) the spin relaxation time. If a circularly polarized pump pulse generates carriers in the spin-up state, and this state is then probed by a SCP pulse, the evolution of the population of carriers in this state is effectively described by Eq. (2). If spin-up carriers are generated by the pump, but the spin-down state is probed by OCP light, the change in the population of spin-down carriers with time is given by Eq. (3).

\[
\frac{dN^-}{dt} = -\frac{N^-}{\tau_e} + \frac{N^+}{\tau_s} - \frac{N^-}{\tau_s},
\]

(3)

The solutions to the addition and subtraction of Eqs. (2) and (3), respectively, are

\[
S = S_0 \exp(-t/\tau_e),
\]

(4)

\[
D = D_0 \exp(-t/\tau_s),
\]

(5)

where \(S = N^+ + N^-\), \(D = N^+ - N^-\), and

\[
\tau_s^{-1} = \tau_e^{-1} + 2\tau_s^{-1}.
\]

(6)

Adding the changes in probe transmission as a function of time delay for SCP and OCP configurations results in an exponential decay that can be fitted with Eq. (4). Similarly, subtracting the change in OCP probe transmission from the change in SCP transmission results in a decay which may be fitted with Eq. (5). \(\tau_e\) and \(\tau_s\) can then be extracted from the fits (Fig. 3), giving a carrier recombination time and a spin relaxation time, respectively.

In a quantum well (QW), the spin relaxation rate due to the D’yakonov-Perel (DP) process is given by

\[
1/\tau_r = 16kTm^*E_{1c}\gamma E_{1c}\hbar^5,
\]

(7)

where \(\gamma\) is a spin-splitting factor, \(m^*\) the electron effective mass, \(E_{1c}\) the first electron confinement energy in the QW, and \(\tau_r\) the electron momentum relaxation time. Previous studies found the DP process to be the main electron spin relaxation mechanism in GaAs/AlGaAs MQWs at RT, while the Elliot-Yafet (EY) process dominates in InGaAs/InP MQWs.

Tackeuchi et al. modified the expression for spin relaxation rate by the EY mechanism for a QW (Ref. 11 and 13–15) as follows, where \(\Delta\) is the spin-orbit splitting energy:

\[
\frac{1}{\tau_s} = \frac{8}{9} \left( \frac{\Delta}{E_g + \Delta} \right)^2 \left( 1 - \frac{m^*}{m} \right)^2 \frac{E_{1c}kT}{E_g^2} \frac{1}{\tau^*},
\]

(8)

Spin relaxation rates predicted by the EY process are proportional to the quantum confinement energy \(E_{1c}\). Both DP and EY theories predict more efficient spin flip for narrower wells. This was borne out experimentally in this study, with \(\tau_r\) of 77, 116, and 133 ps for QW widths of 5.8, 7, and 8 nm, respectively. These results are consistent with trends observed in other material systems. \(E_{1c}\) values were obtained with an analytical model, using effective masses calculated using the band anticrossing model for the conduction band, and a 6 × 6 Hamiltonian including the influence of the spin-orbit band for the valence band. A band offset ratio of 80:20 for GaInNAs MQWs with GaAs barriers was assumed. The dependence of \(\tau_s\) on \(E_{1c}\) is shown in Fig. 4, with the gradient of the fitted line suggesting that the EY process may have significance for electron spin relaxation in this material. No postgrowth annealing was carried out on these samples, and nitrogen concentration was nominally the same for all MQWs. However, it was well known that the incorporation of nitrogen into a GaInNAs matrix has a profound effect on the alloy band structure, and that thermal annealing greatly improves crystal quality. Thermal annealing is expected to be accompanied by increases in electron mobility and \(\tau^*\), a fact used by Lombrez et al. in their interpretation of time-resolved photoluminescence studies on spin dynamics in dilute nitrides. As \(\tau_s\) is directly proportional to \(\tau_s\) according to EY theory, but inversely proportional to \(\tau_e\) in the DP model, a study of spin relaxation times including the role of nitrogen and the effects of rapid thermal annealing may yield more insight into spin relaxation in GaInNAs. A calculation taking into account electron mobilities would be.

![FIG. 2. Observed change in probe transmission as a function of time delay for sample B when probe is same (SCP) and opposite (OCP) circularly polarized relative to the pump.](image1)

![FIG. 3. Sum and difference of SCP and OCP probe transmissions as a function of time delay for sample B.](image2)

![FIG. 4. Spin relaxation time as a function of first electron confinement energy for MQW samples C, B, and A.](image3)
appropriate in order to provide a more complete description.\textsuperscript{20}

In summary, we have investigated electron spin relaxation in GaInNAs MQWs at an optical communication wavelength, as a function of well width and first electron confinement energies, at RT. The observed dependence of $\tau_s$ on $E_{1e}$ suggests that the EY mechanism may have significance for spin relaxation in GaInNAs. Future work will consider the effects of varying nitrogen concentration and thermal annealing, in order to more fully appreciate the interplay of mechanisms responsible for spin relaxation in this material.

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