Probe of Interband Relaxations of Photo-excited Carriers and Spins in InSb Based Quantum Wells

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Abstract

In this work we report the dynamics of photo-excited carrier/spin in several InSb/Al\textsubscript{x}In\textsubscript{1-x}Sb based quantum wells using differential transmission spectroscopy. The InSb quantum well layers were selectively pumped and probed by mid-infrared pulses to avoid possible contributions from the barrier materials. Our observations are suggesting that the initial distribution function strongly influences the carrier relaxation dynamics. In addition, we observe spin relaxation times ranging from 1.2-4.2 ps, longer than the earlier observations in similar structures. We compare our results with our earlier measurements using magneto-optical Kerr effect.

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Keywords: Narrow Gap Semiconductors; Time Resolved Spectroscopy; Spin Relaxation

1. Introduction:

In light of the growing interest in spin-related phenomena and devices, there is now a renewed interest in the science and engineering of narrow gap semiconductors (NGS) such as InSb. NGS offer several scientifically unique electronic features such as a small effective mass, a large g-factor, a high intrinsic mobility, and large spin-orbit coupling effects. In semiconductors with large spin-orbit interaction the coupling of electron spin polarization with electric fields or currents can provide new opportunities for spin manipulation in both electronic and optoelectronic devices. The samples studied in this work are InSb quantum wells (QWs) grown on GaAs (001) substrates by MBE at the University of Oklahoma. The Al\textsubscript{x}In\textsubscript{1-x}Sb barrier layers are delta-doped with Si, located either on one side of the QW (asymmetric sample) or equidistant on both sides of the QW (symmetric sample) \textsuperscript{[1-2]}. The samples’ characteristics are summarized in Table 1.

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doi:10.1016/j.phpro.2010.01.156
The strain in the InSb QWs can remove the degeneracy of the heavy hole and light bands, resulting in two different gaps at the $\Gamma$ point, with the light hole gap being larger.

**Table 1:** Characteristics of the samples studied in this work. The densities and mobilities are from the measurements at 290 K.

| Sample   | Density cm$^{-2}$ | Mobility cm$^2$/Vs | $x$ % | CB1-HH1 meV ($\mu$m) | CB2-HH2 meV ($\mu$m) | CB1-LH1 meV ($\mu$m) | CB2-LH2 meV ($\mu$m) |
|----------|------------------|-------------------|------|---------------------|---------------------|---------------------|---------------------|
| S3(S939) | 2.2x10$^{12}$    | 18,000            | 15   | 322 (3.8)           | 449 (2.8)           | 386 (3.2)           | N/A                 |
| A1(S360) | 6.4x10$^{11}$    | 21,000            | 9    | 265 (4.7)           | 330 (3.8)           | 302 (4.1)           | 360 (3.45)          |
| M1(S591) | Undoped          |                   |      | 265 (4.7)           | 330 (3.8)           | 302 (4.1)           | 360 (3.45)          |

2. Measurements:

We probed the interband relaxation of photo-excited carriers and spins in doped and undoped InSb QWs by employing several pump/probe schemes. The laser pulses were from an optical parametric amplifier (OPA) pumped by a chirped pulse amplifier (CPA), from a difference frequency generator (DFG), which mixes the signal and idler beams from an OPA, and the CPA itself. The pulses had a repetition rate of 1 KHz and duration of ~ 100 fs defining the resolution of the measurements. For 9% alloy concentration, the band gap of Al$_x$In$_{1-x}$Sb at 300 K is ~ 362 meV (3.48 $\mu$m) and only the MIR pulses fixed at 3.467 $\mu$m can potentially create carriers in the barrier layer. This fact is supported by earlier measurements that determined the concentration and temperature dependence of the fundamental energy gap in Al$_x$In$_{1-x}$Sb [3]. For S3, with 15% alloy concentration, the absorption of the selected MIR pulses by the barrier can be ignored.

Understanding the dynamical behaviour of nonequilibrium carriers generated by intense pump pulses can provide valuable information about different scattering mechanisms, carrier phonon coupling, and band structures. Here we present carrier dynamics in several pump/probe schemes tuned in the vicinity of several possible interband transitions listed in Table 1. Figure 1 shows the examples of these measurements where the carriers are created by NIR pulses fixed at 800 nm above the band gap of Al$_x$In$_{1-x}$Sb and InSb and probed by MIR pulses. The pump fluence is on the order of 5 mJ-cm$^{-2}$ corresponding to a photo-excited carrier density of ~5x10$^{18}$ cm$^{-3}$. The carriers were captured in the QW in a time scale of ~ 800 fs and not fully relaxed in a time scale longer than 20 ps. Electrons that are sufficiently energetic have the possibility to scatter between the X, L, and $\Gamma$ valleys in the conduction band, resulting in a longer relaxation time and a more complex relaxation pattern [4]. The initial distribution function influences the evolution of carrier populations [5], examples of this fact can be observed in a series of measurements, presented in Fig. 1b, where the pump pulses are selectively tuned close to several interband transitions in InSb layer with a similar laser fluence as the two-color measurements.

![Fig. 1: a) Two-color differential transmission measurements at 290 K. The pump excitation was fixed at 800 nm and the MIR probe beams were tuned at different possible interband transitions. b) Carrier relaxation in a degenerate MIR pump/probe scheme close to interband transitions.](image-url)
For a transition at 3.467 μm (LH2-CB2), we observed an initial increase in the differential transmission followed by a change in the sign of the signal. The initial sharp increase in the differential transmission can result from free carrier Drude absorption, whereas the alteration of the dielectric function of the film through changes in the electron and hole distribution functions can change the sign of the differential transmission. The signal in the time scale larger than 3 ps did not relax to its original value at the negative time delay. This slow relaxation component, observed in all traces, can be ascribed to interband recombination. In addition, the presence of the generated nonequilibrium carriers can affect the probe absorption through the Pauli blocking of interband optical transitions [4,5]. The relaxation of the photo-excited carriers demonstrated different characteristics for 3.8 μm (HH2-CB2) and 4.1 μm (LH1-CB1) where the effect of the barrier layer can be ignored. For these two wavelengths, the carrier dynamic, except for the initial 0.5 ps, is dominated by the free carrier absorption followed by a slower relaxation component.

In order to probe the relaxation of photo-induced spin polarized carriers in the InSb QWs, pump/probe pulses were tuned close to several interband transitions of the InSb QWs. By monitoring the transmission of a weaker, delayed probe pulse that has the same circular polarization (SCP) or opposite circular polarization (OCP) as the pump pulse, the optical polarization \( P = (SCP - OCP)/(SCP + OCP) \) can be extracted. The optical polarization \( P \) is decaying exponentially with a decay constant related to the spin lifetime as following: \( P = P_0 \exp(-t/T_1) \), where the magnitude of \( P_0 \) is a constant and can be 0.25 at best for bulk III-V semiconductors [6]. In order to measure the optical polarization, the differential transmissivity was recorded as a function of the time delay between the pump and probe pulses, using a liquid nitrogen cooled MCT detector. Figures 2a-c are examples of the polarization-resolved differential transmission measurements at 4.1 μm to extract the spin relaxation (\( T_1 \)).

Table 2 summarizes the results of the measurements for several MIR wavelengths in the vicinity of interband transitions. We observe spin relaxation times ranging from 1.2-4 ps, longer than the earlier observations in Te-doped InSb/Al\(_{0.15}\)In\(_{0.85}\)Sb QWs at room temperature [7,8]. We compared the measured \( T_1 \) in our samples with the theoretical models in two dimensional systems based on the Dyakonov-Perel (DP) relaxation mechanism [8-10]. Using this model, the spin relaxation rates for the QWs with 30 nm (11.5 nm) width, are predicted to range from 3-6 ps (0.5-1 ps) depending on the choice for a dimensionless parameter \( C_{DP} \) in the model, being 16 or 32 [8-10]. For the 30 nm QWs, the Elliot-Yafet (EY) model [8-10], predicts relaxation rates ranging from 5 to 32 ps, depending on a dimensionless parameter, \( C_{EF} \), to be either 7.5 or 1. For the narrower QW, the EY model predicts the relaxation times ranging from 2 to 16 ps for the same choice of parameters.

In the DP model, the spin relaxation time is inversely proportional to \( E_{1e}^2 \), where \( E_{1e} \) is the confinement energy for the lowest conduction subband. The scaling of the spin relaxation time with \( E_{1e} \) can serve as a reference for distinguishing between different spin-relaxation mechanisms for a constant momentum relaxation time. In the EY model [8-10], the spin relaxation is inversely proportional to \( E_{1e} \) and linearly proportional to momentum scattering time. We observed faster relaxation times for the transitions in the vicinity of first subband transitions (3.467 μm and 3.8 μm) in S3 compared to the relaxation times corresponding to the first subband transitions (4.1 μm and 4.68 μm) of A1 and M1. In S3, the relaxation time is faster by about a factor 2; whereas, \( E_{1e} \) is 3 times larger than the other two samples. This fact will be examined by further measurements on a series of InSb QWs with different well widths, when they become available.

The relaxation dynamics in undoped QWs can be more complex. In undoped QWs, the electron-hole pair near the band gap forms an exciton. The exciton spin can relax via spin relaxation of either hole or electron, or spin flip of both entities simultaneously. The main mechanism, which simultaneously flips the electron and hole spins, is the exchange interaction [11]. The complex nature of the valance band and the strong non-parabolicity in this material system can result in a short range exchange interaction in which the light hole and heavy hole mixing can flip the exciton spin [11]. InSb QWs are the narrowest band gap for which room temperature excitons have been observed [12]. In the undoped sample, M1, we measured longer \( T_1 \) in the vicinity of the LH1-CB1 and HH1-CB1 transitions, close to excitonic transitions, compared to the measurements in the vicinity of the second subband transitions. In order to probe the excitonic effects, we will study the relaxation times as a function of the temperature and the well width, in a series of undoped InSb QWs.
Table 2: Spin relaxation times measured for three InSb QWs at 290 K in the vicinity of several interband transitions. The observed measurements are best described by a theoretical model on the basis of the DP relaxation mechanism.

| Wavelengths | 3.467 μm | 3.8 μm | 4.1 μm | 4.68 μm |
|-------------|----------|--------|--------|---------|
| Samples     | T1 (ps)  | T1(ps) | T1(ps) | T1(ps)  |
| S3 (S939)   | 2.3      | 1.6    | 2.0    | 1.5     |
| A1 (S360)   | 1.2      | 1.2    | 4.2    | N/A     |
| M1 (S591)   | 2.0      | 1.5    | 4.0    | 3.5     |

Fig. 2: The optical polarization for three different QWs using the differential transmission technique and two-color MOKE. a) The optical polarization is decaying exponentially with a decay constant related to the spin lifetime. b) Same as a), only this time an undoped multi quantum well was probed. c) Same as a) and b), for a symmetrically doped QW. d) Probing spin relaxation of a symmetrically doped QW using a two-color MOKE technique. The sample is 11.5 nm wide QW and has a 15% alloy concentration. By tuning the pump at 2.6 μm it was possible to excite carriers only in the InSb layer. Figure 2d presents MOKE measurements in S3, where the use of pump pulses close to the HH2-CB2 transition at 2.6 μm, made it possible to excite carriers only in the InSb layer. We observed much longer relaxation time
compared to the differential transmission measurements which might be related to the nature of the measurement. For MOKE measurements on layered structures, one has to include multiple reflections of the probe beam (800 nm in this case) at the QW interfaces. In multi-layer structures, the signal is in general a mixture of "absorptive" (resonant) and "dispersive" contributions, so that the response has a component related to the average magneto-optical activity which in turn is some weighted average of spin polarization over the bands [13-14].

Summary:

In summary, we measured the carrier and spin relaxation times employing several pump/probe schemes in three InSb QWs. Our observations are suggesting that the initial distribution function strongly influences the evolution of the carrier populations. Using the polarization-resolved differential transmission measurements, the spin relaxation times in the vicinity of several interband transitions were measured. The observations in the 30 nm wells can be best described by the DP model. For the narrower well, there is a cross over where both EY and DP are predicting a relaxation time in the same order of our observations. Some of the observed wavelength dependences can be explained by the differences in the samples' structures. In addition, the formation of the excitons, in the undoped sample, can influence the picture of the relaxation mechanisms.

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Acknowledgment:

Supported by: NSF-DMR-0507866, NSF-DMR-0520550