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Time Resolved Spectroscopy of InMnAs Using Differential Transmission Technique in Mid-Infrared

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Abstract

InMnAs grown by MOVPE is a room temperature ferromagnetic semiconductor with a T_c of 330 K. The origin of the ferromagnetism and the interactions between itinerant carriers and localized spins in these structures are open questions. To address these questions, the carrier and spin life time in these structures were probed in mid-infrared region. The approach in this work was focused on the time and polarization-resolved differential transmission measurements suggesting a T_1 of ~ 1 ps. We compare our results with reported spin relaxations in InAs and MBE grown InMnAs.

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1. Introduction:

We have investigated carrier and spin relaxation dynamics in ferromagnetic $\text{In}_{1-x}\text{Mn}_x\text{As}$ films that were grown by metalorganic vapor phase epitaxy (MOVPE). The first III-V ferromagnetic semiconductor alloy, $\text{In}_{1-x}\text{Mn}_x\text{As}$, was prepared by molecular beam epitaxy (MBE) with a Curie temperature (T_c) on the order of 10 K. Subsequent studies demonstrated a T_c closer to 90 K [1]. Low temperature MBE was nearly exclusively used to prepare InMnAs thin films; although, MOVPE, an alternative technique demonstrated that single phase magnetic InMnAs compound could be deposited at 500°C, about 200°C higher than that used in MBE. Furthermore, MOVPE technique subsequently demonstrated that the films are ferromagnetic with a T_c of 330 K. X-ray diffraction and TEM analysis of the layers have ruled out the presence of a second phase with a different crystal structure. The mean field theory predicts that T_c should scale with Mn concentration (x) and hole concentration (p) as $xp^{1/3}$. Reasonable agreement between theory and experiments has been noted for GaMnAs. In contrast, the observation of a T_c of 10 K for

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InMnAs films with carrier concentration of 10^{18} cm^{-3} is inconsistent with calculations. Based on the calculations, InMnAs with a hole concentration of 10^{18} cm^{-3} should have a T_c of 8 K instead of the experimentally observed 90 K. The case for InMnAs grown by MOVPE is even more complex [2]. Films with a carrier concentration of 10^{18} cm^{-3} have T_c of 330 K and the T_c is nearly independent of carrier concentration.

This fact motivated us to probe Cyclotron Resonance (CR) in this material system to understand the nature of the carrier states in the valence band. CR is a direct and accurate method for determining the effective mass and therefore the energy dispersion of carriers. The measurements were performed in the High Magnetic Field Laboratories, Dresden, Germany. A free electron laser coupled to a pulsed magnet generating fields up to 50 Tesla, provided a tunable radiation source in the Far-Infrared. Unlike the reported measurements on the MBE grown InMnAs [3], we did not observe CR in MOVPE grown InMnAs up to 50 Tesla. The measurements at higher magnetic fields will be pursued. In addition to high T_c , the other significance of the MOVPE grown structure is the direct growth of the magnetic layer on the GaAs substrate. The MBE grown ferromagnetic InMnAs films with the GaSb layers below the magnetic layer resulted in a type II band alignment providing localized regions at the interface for the accumulation of holes. The holes at the interface could have been responsible for the observation of CR in the MBE grown InMnAs [3].

Recently, the magnetoresistance of InMnAs/InAs *p-n* heterojunctions for magnetic fields of up to 18 T has been measured. At 300 K, giant magnetoresistance effects (2680 %) at 18 T were observed [4]. A crucial factor in the design of any spin-sensitive devices is the electron-spin-relaxation time (T_1) which must be reasonably long to allow for transport and processing of the spins. The origin of ferromagnetism along with the interactions between itinerant carriers and localized spins in these structures are open questions. In this work, we probed the carrier and spin life time in the MOVPE grown InMnAs structures. The sample is an 800 nm thick film with a Mn content of ~4%, a hole concentration $p=1.35 \times 10^{18} \text{ cm}^{-3}$, and a mobility $\mu=142 \text{ cm}^2/\text{Vs}$. The details of the growth conditions are described in Ref. [2].

Although the s-d coupling with the localized Mn ions is considered to be significantly weaker compared to the p-d exchange coupling that characterizes the valence band, the holes are known to relax faster than electrons (<0.5 ps). Most of the understanding of the spin relaxation in narrow gap (III,Mn)V ferromagnetic structures has been on the basis of two color magneto-optical Kerr (MOKE) time-domain spectroscopy [5-7]. In multi-layer structures, the signal in general, is a mixture of "absorptive" (resonant) and "dispersive" contributions. Therefore, the response has a component related to the average magneto-optical activity, which can be some weighted average of spin polarization over the bands [8,9]. In general, there is no guarantee that MOKE signals would be proportional to the actual instantaneous spin polarization. Altering a magnetic system with short laser pulses can strongly modify the thermodynamic equilibrium among the carriers, spins, and ultimately the lattice.

2. Experimental Approach:

The approach in this work employed the time and polarization-resolved differential transmission measurements to provide insight into the timescales and the nature of microscopic interactions in MOVPE grown InMnAs. Using this technique and by tuning the pump-probe radiations from 3-3.5 microns we mainly probed the relaxation of the photo-induced electrons. We compare our results with reported spin relaxations of photo-excited electrons in InAs and MBE grown InMnAs. In our measurements, the MIR laser source was a difference frequency generator (DFG), which mixes the signal and idler beams from an optical parametric amplifier (OPA). The OPA itself was pumped by an amplified Ti:sapphire oscillator with a repetition rate of 1 KHz. The pulses had a duration of ~ 100 fs defining the resolution of the measurements. Spin-polarized electrons were created/probed close to the fundamental gap of the InMnAs film using circularly polarized pulses in the mid-infrared (MIR) ranging from 3.1-3.5 μm . 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 $P=(SCP-OCP)/(SCP+OCP)$ which is proportional to the spin polarization can be measured [10]. The differential transmissivity as a function of the time delay between the pump and probe pulses, using a liquid N_2 -cooled MCT detector, was measured. Similarly, differential transmission measurements can extract the carrier relaxation time.

3. Results and Discussions:

Examples of our measurements on a MOVPE grown InMnAs film with 4% Mn content are presented here. Figure 1a demonstrates the relaxation of photo-excited carrier generated/probed at $3.467\ \mu\text{m}$ (close to the fundamental gap of InMnAs) at 290 K. Here, we observed several relaxation regimes with an initial sharp increase in differential transmission followed by a change in the sign of the signal. Optical transitions in semiconductors can be influenced strongly by the distribution of carriers in the conduction and valence bands. The initial sharp increase in the differential transmission results from free carrier Drude absorption; whereas, the alteration of the dielectric function of the film through changes in the electron and hole distribution function can be responsible for the sign change of the differential reflectivity. By tuning the pump/probe above the gap at $3.1\ \mu\text{m}$, demonstrated in Fig. 1b, the differential transmission did not change the sign. In both cases the differential transmission in the time scale larger than 3 ps didn't fully relax to its initial value at the negative time delay.

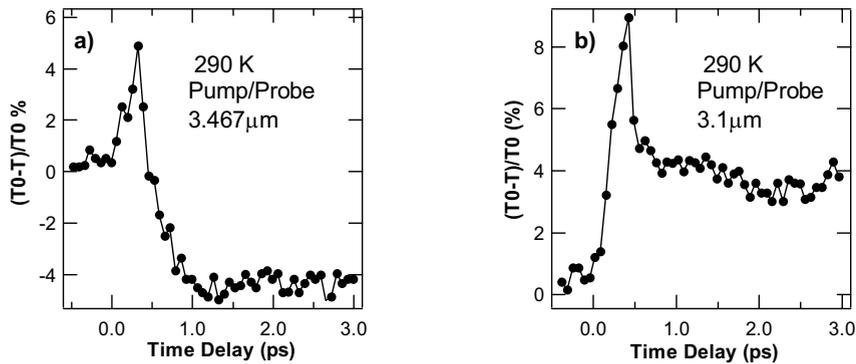


Fig. 1: Differential transmission in MOVPE grown ferromagnetic InMnAs at 290 K. a) By tuning the pump/probe at $3.467\ \mu\text{m}$, close to the fundamental gap of InMnAs, we observed several relaxation regimes with an initial sharp increase in differential transmission followed by a change in the sign of the signal. b) A different characteristic in the relaxation of the photo-induced carriers was observed by pumping/probing the carriers at a slightly different wavelength.

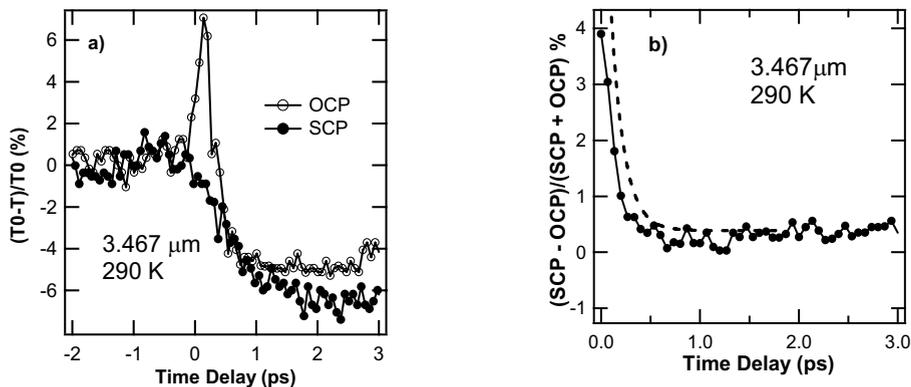


Fig. 2: Polarization-resolved differential transmission measurements. a) The normalized induced-transmission change of the probe beam as a function of the time delay between pump and probe pulses having the SCP and the OCP at $3.467\ \mu\text{m}$. b) The optical polarization P is decaying exponentially with a decay constant related to the spin lifetime. This observation suggests a spin relaxation of $\sim 1\text{ps}$. The exponential fit to the trace is shifted for clarity.

Recently, various measurements using differential transmission techniques to probe spin relaxation in InAs based systems have been performed. Hall *et al.* studied spin relaxation in (001) InAs/GaSb heterostructures and observed a spin lifetime of ~ 700 fs at 117 K [11]. Litvinenko *et al.* [12] studied spin lifetime in three undoped InAs films of thicknesses 0.15, 0.27, and 1 μm , for temperatures ranging from 77 K to 290 K. Several spin relaxation regimes were observed and the spin lifetimes were ranged from 1 ps for a 0.15 μm thick layer to 20 ps for a 1 μm thick layer, at 77 K [12]. The strong temperature dependence of the spin lifetime was observed only in the thickest films and a spin lifetime shorter than 1 ps was estimated in the surface layer. MOKE studies of InAs films in a similar laser fluence regime of this work demonstrated fast spin relaxation shorter than 2 ps [13]. The reported photo-induced time resolved MOKE in the MBE grown InMnAs demonstrated several relaxation regimes depending on the pump fluence, including a fast (< 1 ps) relaxation, interpreted as the dynamics through sp-d exchange interaction of photo-carriers and Mn ions, and a ~ 100 ps component, interpreted as a manifestation of spin-lattice relaxation [7].

In order to measure T_1 , we performed the polarization-resolved differential transmission measurements in MIR. Figure 2a shows an example of these measurements at 3.467 μm , demonstrating the normalized induced-transmission change of the probe beam as a function of the time delay between pump and probe pulses having the SCP and the OCP. As shown in Fig. 2b, the optical polarization P is decaying exponentially with a decay constant related to the spin lifetime as follows: $P = P_0 \exp(-t/\tau_s)$, where the magnitude of P_0 is a constant and can be 0.25 at best for bulk III-V semiconductors [9]. The observations in this work suggest spin relaxation times (T_1) to be in the range of ~ 1 ps at 290 K. We haven't observed significant temperature or wavelength dependence in the observed spin relaxations.

4. Summary:

In summary, we employed the time and polarization-resolved differential transmission measurements to provide information on the carrier/spin relaxation dynamics in MOVPE grown InMnAs. Our measured T_1 is comparable to the reported MOKE measurements in MBE grown InMnAs and several time resolved measurements on InAs. In order to provide complementary information, time resolved MOKE studies on this material system are underway. In addition, we performed high magnetic field CR up to 50 Tesla and unlike the reported measurements on the MBE grown InMnAs, we did not observe CR in MOVPE grown InMnAs. The measurements at higher magnetic fields will be pursued.

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