Propagating Coherent Acoustic Phonon Wavepackets in InMnAs/GaSb

J. Wang, Y. Hashimoto,* J. Kono†
Department of Electrical and Computer Engineering, Rice Quantum Institute, and Center for Nanoscale Science and Technology, Rice University, Houston, Texas 77005

A. Oiwa‡
PRESTO, Japan Science and Technology Agency, 4-1-8 Honcho, Kawaguchi 332-0012, Japan

H. Munekata
Imaging Science and Engineering Laboratory, Tokyo Institute of Technology, 4259 Nagatsuta, Yokohama 226-8503, Japan

G. D. Sanders, C. J. Stanton
Department of Physics, University of Florida, Gainesville, Florida 32611
(Dated: November 23, 2018)

We observe pronounced oscillations in the differential reflectivity of a ferromagnetic InMnAs/GaSb heterostructure using two-color pump-probe spectroscopy. Although originally thought to be associated with the ferromagnetism, our studies show that the oscillations instead result from changes in the position and frequency-dependent dielectric function due to the generation of coherent acoustic phonons in the ferromagnetic InMnAs layer and their subsequent propagation into the GaSb. Our theory accurately predicts the experimentally measured oscillation period and decay time as a function of probe wavelength.

Recently, there has been much interest in (III,Mn)V dilute magnetic semiconductors (DMS) with carrier-mediated ferromagnetism, a promising system for the realization of future semiconductor spintronic devices capable of performing information processing, data storage, and communication functions simultaneously. InMnAs is the prototypical III-V DMS, being the first DMS to exhibit ferromagnetism, and the first DMS in which cyclotron resonance was observed, evidence that at least some of the carriers are itinerant.

Despite the fact that time-domain studies of the dynamic aspects of (III,Mn)Vs are more informative than static magnetization or electrical transport measurements, relatively few time-dependent studies have been attempted. In this paper, we report on our time-dependent femtosecond transient reflectivity measurements on an InMnAs/GaSb heterostructure using two-color pump-probe spectroscopy. In addition to changes in the reflectivity associated with ultrashort carrier lifetimes (∼2 ps) and multi-level carrier decay dynamics which we attribute to a large density of bound states and a high concentration of Mn p-type dopants, we observe pronounced oscillations in the differential reflectivity signal. While originally the oscillations were believed to originate from the ferromagnetism (since they were not observed in similar systems without Mn doping), a systematic study shows that they instead result from the generation of coherent phonons in the InMnAs layer which then propagate into the GaSb. The period and the characteristic decay time of the transient reflectivity oscillations are consistent with a propagating coherent acoustic phonon wavepacket model.

While femtosecond spectroscopy has previously been used for generating and detecting coherent phonons in many materials (e.g., optical phonons in bulk semiconductors, metals (Bi, Sb) and superconductors; acoustic phonons in InGaAs/GaAs-based semiconductor heterostructures), only recently has there been interest in the application of this technique to probing coherent excitations (e.g., phonons and magnons) in strongly-correlated electronic systems. These innovative studies further demonstrate the potential of transient reflectivity spectroscopy to unravel the subtle interplay between lattice and spin excitations.

The main Mn-doped sample studied was a p-InMnAs/GaSb heterostructure grown by low-temperature molecular beam epitaxy (LT-MBE). The sample [shown schematically in Fig. 1(a)] consisted of a 25-nm p-type InMnAs layer and an 820-nm GaSb buffer layer grown on top of a (100) GaAs substrate. The hole density and mobility in the InMnAs layer, as estimated by Hall measurements, were 1.1 × 10¹⁰ cm⁻³ and 323 cm²/Vs, respectively. Detailed growth conditions and sample information can be found in Ref. 17.

In order to separate various nonlinear effects and extract information on the dynamical response of the InMnAs/GaSb heterostructure, we used a two-color, selective pumping scheme shown schematically in Fig. 1(a). A 140 fs, midinfrared (MIR) 2 μm pump beam from an optical parametric amplifier (OPA) was used to create carriers in InMnAs with an excess kinetic energy of ∼0.2 eV.
InMnAs

25 nm

GaSb

820 nm

Pump,

2 µm, ~150 fs

Probe,

0.5-1.4 µm, ~340 fs

FIG. 1: (a) Schematic diagram of two-color pump-probe differential reflectivity experiment in an InMnAs/GaSb heterostructure. The pump photoexcites carriers only in the InMnAs layer and the transient differential reflectivity is probed at higher energies. (b) Typical time-resolved differential reflectivity data, showing an initial drop followed by oscillations in reflectivity.

Pumping at this photon energy allowed us to selectively photoexcite carriers in the InMnAs layer but not in the GaSb buffer layer or GaAs substrate. A near-infrared (NIR) (775 nm) probe beam from a chirped pulse amplifier (Model CPA-2010, Clark-MXR, Inc.) allowed us to probe energies far above the quasi-Fermi level of the optically excited carriers.

Typical time-dependent differential reflectivity data are shown in Fig. 1(b). After photoexcitation, the reflectivity shows a sharp drop followed by a rapid rise and sign change in a time less than ~2 ps. At longer times (several hundred ps) periodic oscillations are observed with a period of ~23 ps superimposed on a very slow decay. The initial sharp drop in reflectivity is believed to result from free carrier Drude absorption by the hot photogenerated carriers. The carriers relax back to quasi-equilibrium distributions through the emission of confined LO phonons and the ultrafast trapping of electrons (by AsGa antisite defects) and holes (by Ga vacancies) by the mid-gap states introduced by LT-MBE growth. This alters the dielectric function of the heterostructure through changes in the electron and hole distribution functions and gives rise to the sharp increase and sign change in the differential reflectivity seen in Fig. 1(b). Over much longer times, the quasi-equilibrium electrons and holes recombine across the gap causing the differential reflectivity to return to zero. The beginning of this slow decay can be seen in Fig. 1(b).

Initially, the oscillations were thought to originate from the Mn ions and possibly the ferromagnetism since they were not observed in similar samples without the Mn. However, when we applied an external magnetic field of 0, 5, and 9.5 Tesla, the oscillation period did not change, as shown in Fig. 2(a). This rules out magnons or quantum beats between Landau levels as a source of the oscillations.

Next, we varied the pump fluence as shown in Fig. 2(b). For pump fluences of 0.7, 3.5, and 10 mJ/cm², the intensity of the oscillation changed but the frequency did not. Varying the pump fluence increases the photoexcited carrier density. Since the plasma frequency increases with the total carrier concentration, we would expect the oscillation period to increase with increasing pump fluence if it were related to plasmons. Since the oscillation period

FIG. 2: Two-color pump-probe differential reflectivity oscillations in an InMnAs/GaSb heterostructure. The dependence of the oscillations on magnetic field is shown in (a) while the dependence on pump power is shown in (b). The oscillation period does not change as shown by the vertical dashed lines.
is independent of pump fluence, we rule out plasmons as the cause of the oscillations.

A final possibility is that coherent phonons could be excited in the InMnAs epilayer, since they have previously been observed in InGaN/GaN epilayers [8,11], though InGaN/GaN has a wurtzite structure and as a result, strain can lead to large built-in piezoelectric fields (unlike InMnAs which has zinc-blende structure). The theory for the generation and propagation of coherent acoustic phonons in InGaN/GaN nanostructures having the wurtzite structure has been described in Ref. [17] (see the erratum in Ref. [19]). Reference [20] provides a good review of both experimental and theoretical aspects of coherent acoustic phonon generation in piezoelectric nitride-based semiconductor heterostructures. Generation of coherent acoustic phonons comes about from the electron-phonon interaction [21,22,23]. In nitride-based heterostructures such as GaN/InGaN large amplitude coherent acoustic phonon wavepackets can be generated through the piezoelectric and deformation potential electron-phonon interactions with the piezoelectric interaction being dominant.

In contrast to the nitrides, the InMnAs/GaSb heterostructure has zinc-blende structure. Consequently, coherent acoustic phonons cannot be generated (for [100]-grown samples) through the piezoelectric electron-phonon interaction. They can however be generated through the deformation potential electron-phonon interaction, but their amplitudes are weak. However, we can detect the coherent phonon oscillations in the differential reflectivity through a fortuitous circumstance. The wavelength-dependent GaSb dielectric function is very sensitive to small changes in strain in the vicinity of the E_1 (L-valley) transition centered at approximately 600 nm, which is close to our probe region.

We developed a theoretical model for the transient differential reflectivity in our InMnAs/GaSb structure based on a Boltzmann equation formalism. The photoexcited carriers in the ferromagnetic quantum well are assumed to be completely confined and electronic states near the band edge are treated in an eight-band effective mass model including conduction electrons, heavy holes, light holes, and split-off holes. The effect of Mn impurity spins on the itinerant carriers is also included. Photogeneration of carriers in the quantum well is treated using Fermi’s Golden Rule and carrier scattering by confined LO phonons in the quantum well is accounted for. From the time-dependent carrier densities, the generation and propagation of coherent acoustic phonons are modelled by solving a loaded string equation as described in Ref. [18]. Changes in the position- and frequency-dependent dielectric function due to coherent acoustic phonons are computed and the time-dependent reflectivity at the probe wavelength is obtained by globally solving Maxwell’s equations in the entire structure.

In Fig. 3 the coherent phonon differential reflectivity oscillations are shown as a function of time delay for probe wavelengths of 650, 775, and 850 nm. The theoretical differential reflectivity curves in Fig. 3(b) agree well with the experimentally measured differential reflectivity seen in Fig. 3(a) after subtraction of the transient background signal. As the probe wavelength approaches the GaSb E_1 transition near 600 nm, the amplitude of the differential reflectivity oscillations increase due to the enhanced sensitivity of the GaSb dielectric function to the localized propagating strain pulse.

The damping of the differential reflectivity oscillations seen in Fig. 3 is due to the finite penetration length of our probe inside the GaSb layer. The periodic oscillations in the differential reflectivity can be modelled by

$$\frac{\Delta R}{R}(t) \sim \alpha \cos \left( \frac{4\pi C_s n}{\lambda} t - \Phi \right) e^{-t/T},$$

where $\Phi$ and $T$ are the phase and decay time, $\lambda = 2\pi c/\omega$ is the probe wavelength, $C_s$ is the acoustic sound speed, and $n$ is the refractive index at the probe wavelength. From fits to the data, the decay time for a probe wavelength of 650 nm is found to be $\sim 45$ ps. The decay time is approximately $T = \lambda/(4\pi C_s k)$, where $k$ is the extinction coefficient at the probe wavelength. A numerical estimate yields a value of 41 ps for the decay time in

FIG. 3: Experimental (a) and theoretical (b) coherent phonon differential reflectivity oscillations for probe wavelengths of 650, 775 and 850 nm.
The differential reflectivity oscillation period increases with wavelength from 650 to 850 nm. The period of the reflectivity oscillations can be easily understood. The propagating strain pulse gives rise to a perturbation in the GaSb dielectric function which propagates at the acoustic sound speed. The sample thus acts as a Fabry-Perot interferometer and a simple geometrical optics argument shows that the period for the reflectivity oscillations due to the propagating coherent acoustic phonon wavepacket is approximately

$$T = \frac{\lambda}{2 C_s n(\lambda)}$$

where $C_s = 3.97 \times 10^5$ cm/s is the LA sound speed in GaSb, $\lambda$ is the wavelength dependent refractive index. At the wavelength of the probe (850 nm), $n(\lambda) \approx 4.24$ and we calculate the period of the differential reflectivity oscillations to be $T = 25.2$ ps, which is in close agreement with the observed oscillation period of 24 ps seen in Fig. 1(b). In Fig. 4 we have plotted the experimentally measured coherent phonon differential reflectivity oscillation periods as a function of probe wavelength as open circles. The solid line shows the oscillation period vs. probe wavelength estimated using the simple geometrical optics argument and taking the wavelength dependence of the refractive index in GaSb into account. The excellent agreement between theory and experiment clearly demonstrates that the reflectivity oscillations are the result of propagating coherent acoustic phonons in the GaSb barrier.

In summary, we have performed time-dependent two-color differential reflectivity measurements on a ferromagnetic InMnAs/GaSb heterostructure. In addition to observing changes in the reflectivity due to ultrashort carrier dynamics, we have observed reflectivity oscillations resulting from coherent phonon generation in the InMnAs layer. The propagation of these coherent, localized "strain pulses" into and through the GaSb buffer layer results in a position and frequency-dependent dielectric function. Our theoretical calculations accurately reproduce the experimental results. These results offer the intriguing possibility of future devices using coherent phonons to interact with the Mn spins in much the same manner that photoexcited carriers have been used to manipulate ferromagnetism.

We thank Xiangfeng Wang for technical help. This work was supported by NSF (through DMR-0134058, DMR-0325474, and INT-0221704), DARPA (through MDA972-00-1-0034), and ONR (through N000140410657).

[1] I. Žutić, J. Fabian, and S. Das Sarma, Rev. Mod. Phys. 76, 323 (2004).
[2] H. Ohno, Science 281, 951 (1998).
[3] S. A. Wolf et al., Science 294, 1488 (2001).
[4] M. A. Zudov et al., Phys. Rev. B 66, 161307(R) (2002).
[5] G. D. Sanders et al., Phys. Rev. B 68, 165205 (2003).
[6] Y. H. Matsuda et al., Phys. Rev. B 70, 195211 (2004).
[7] R. Liu et al., cond-mat 0310654.
[8] G. C. Cho, W. Küt, and H. Kurz, Phys. Rev. Lett. 65, 764 (1990).
[9] A. Bartels et al., Phys. Rev. Lett. 82, 1044 (1999).
[10] C. K. Sun, J. C. Liang, and X. Y. Yu, Phys. Rev. Lett. 84, 179 (2000).
[11] J. S. Yahng et al., Appl. Phys. Lett. 80, 4723 (2002).
[12] T. K. Cheng et al., Appl. Phys. Lett. 59, 1923 (1991).
[13] J. M. Chwalek et al., Appl. Phys. Lett. 58, 980 (1991).
[14] D. Lim et al., Appl. Phys. Lett. 83, 4800 (2003).
[15] R. Arita et al., Phys. Rev. Lett. 88, 127202 (2002).
[16] T. Shupinski, A. Oiwa, S. Yanagi, and H. Munekata, J. Cryst. Growth 237-239, 1326 (2002).
[17] J. Wang et al., J. Supercond. 16, 373 (2003).
[18] G. D. Sanders, C. J. Stanton, and C. S. Kim, Phys. Rev. B 64, 235316 (2001).
[19] G. D. Sanders, C. J. Stanton, and C. S. Kim, Phys. Rev. B 66, 079903(E) (2002).
[20] G.-W. Chern, C.-K. Sun, G. D. Sanders, and C. J. Stanton, in Topics in Applied Physics, edited by K.-T. Tsen (Springer-Verlag, New York, 2004), vol. 92, pp. 339-394.
[21] A. V. Kuznetsov and C. J. Stanton, Phys. Rev. Lett. 75, 3243 (1994).
[22] A. V. Kuznetsov and C. J. Stanton, Phys. Rev. B 51, 7555 (1995).
[23] A. V. Kuznetsov and C. J. Stanton, in Ultrafast Phenomena in Semiconductors, edited by F. Tsen (Springer-Verlag, New York, 2001), pp. 353-403.