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Ultrafast carrier dynamics in ferromagnetic InGaMnAs

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Abstract

We have carried out an ultrafast time-resolved differential reflectivity study of a ferromagnetic semiconductor InGaMnAs and made a systematic comparison with low-temperature grown and high-temperature grown InGaAs reference films. Very short carrier lifetimes ($\sim$2 ps) were observed in InGaMnAs and the low-temperature grown InGaAs film, but not in the high-temperature grown InGaAs film. We attribute the short lifetimes to carrier trapping by mid-gap states introduced during low-temperature MBE growth. Furthermore, at long times, we observed periodic oscillations in the differential reflectivity signal with period $\sim$20 ps, which we interpret as coherent acoustic phonons.

Keywords: III–V ferromagnetic semiconductors; Ultrafast carrier dynamics

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Recent developments in semiconductor spintronics promise future "multifunctional" devices which have capabilities of information processing, storage and communications. In particular, carrier/photo-induced ferromagnetism in (III, Mn)V semiconductors opened up new possibilities for manipulating long-range magnetic order [1, 2]. However, although their electrical transport and magnetic properties have been extensively studied, there have been only limited studies to explore their optical properties.

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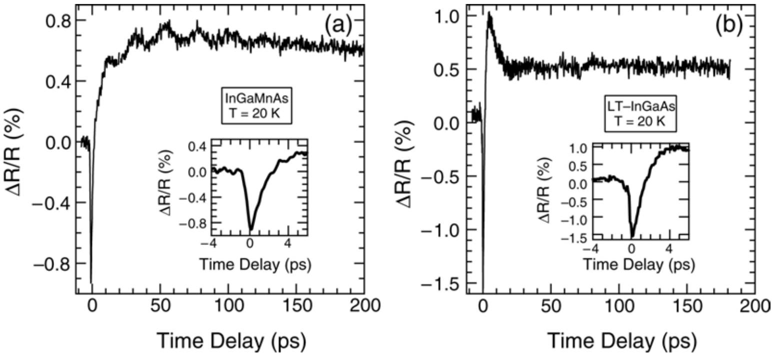

Fig. 1. Differential reflectivity of InGaMnAs (a) and LT-InGaAs (b) at 20 K. The 1.2 μm pump of 5 mW was linearly polarized and the probe wavelength was 775 nm. Insets: initial decay of differential reflectivity.

Study of ultrafast dynamics in (III, Mn)V systems can be interesting due to their low-temperature (LT) MBE growth and high Mn doping. It is known that As-based III–V semiconductors grown at low temperatures contain large density As-related point defects that act as carrier trapping centers, strongly shortening carrier lifetimes [3]. However, microscopic understanding of the observed complex transients and the effects of doping are still elusive [4]. Ultrafast study of carrier dynamics in (III, Mn)V ferromagnetic semiconductors should provide new insight into those partially understood issues.

Here, we report on the first time-resolved optical measurements of hot carrier dynamics in InGaMnAs. A very short carrier lifetime (∼2 ps) was observed in differential reflectivity, which we attribute to low temperature MBE growth. Complex multi-level hot electron and hole decay dynamics was observed, which we discuss via a careful comparison with data for low- and high-temperature grown InGaAs reference samples. Finally, we report on the first observation of coherent acoustic phonons in a magnetic semiconductor.

We performed femtosecond two-color time-resolved differential reflectivity spectroscopy. Experimental details were described previously [5]. At the pump wavelength (1.26 μm), the photon energy (0.98 eV) was just above the band gap of InGaMnAs (∼0.88 eV at 20 K), so the created carriers had only a small amount of extra kinetic energy (0.1 eV), minimizing contributions from intervalley scattering and intraband relaxation.

Typical data showing the ultrafast carrier dynamics of InGaMnAs are presented in Fig. 1(a). The initial fast change in differential reflectivity is negative (see inset of Fig. 1(a)), after which a fast rise occurs, exhibiting a quick sign change, followed by a slow recovery with periodic oscillations with a period ∼24 ps.

For the low temperature InGaAs (LT-InGaAs) (Fig. 1(b)), we observe a similar initial negative change in reflectivity and a subsequent fast rise with a sign change. However, no clear oscillations were observed. Another interesting feature for the LT-InGaAs sample is the "overshoot" that exists right after the sign change and before the very slow recovery starts. On the other hand, in the high-temperature InGaAs (HT-InGaAs) sample (Fig. 2(a)), the reflectivity change only shows a single exponential decay with a decay time ∼104 ps.

Below we discuss the origins of the series of transient phenomena observed for InGaMnAs: (1) an initial ∼1% decrease in reflectivity, (2) a very fast (∼2 ps) decay of the

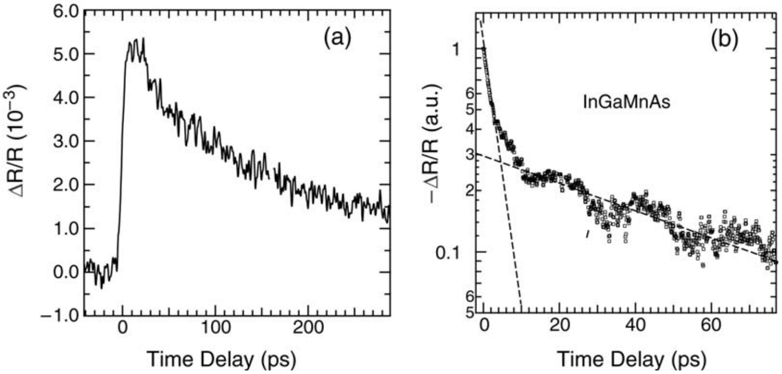

Fig. 2. (a) Differential reflectivity of HT-InGaAs taken under the same conditions as Fig. 1. A single exponential decay with the relaxation time ∼104 ps is observed. (b) The fast-rising components after the initial negative transients in reflectivity of InGaMnAs are plotted in log scale. A bi-exponential curve fitting yields relaxation times 2 ps for the fast and 23 ps for the slow component.

reflectivity decrease, (3) a sign change (to a reflectivity *increase*), (4) periodic oscillations (with period ∼24 ps), and (5) a very slow decay towards the equilibrium reflectivity.

The initial, pump-induced decrease in reflectivity is due to the photo-created transient free carriers. Three main types of free carrier effects are known in semiconductors: free carrier absorption (FCA), band filling (BF), and band gap renormalization (BGR). Among these we believe that the FCA mechanism dominates in the observed initial negative transient for the following reasons. First, according to dielectric theory, simple estimations predict the effects of FCA, BF, and BGR on reflectivity to be negative, positive, and negative at our probe wavelength (775 nm), so BF can be excluded. Second, in the case of BGR, the change in refractive index is largest near the band edge and should be substantially reduced at wavelengths away from the band edge. Third, in LT-grown semiconductors, band edge absorption is usually larger but smeared out by a large density of near band edge states introduced by defects. This will drastically reduce the importance of BGR (and BF). Therefore, we conclude that the photoinduced change in the Drude dielectric constant due to FCA is the origin of the initial negative transient.

We attribute the subsequent fast (∼2 ps) decay to the ultrafast trapping of electrons (by $\mathrm{As_{Ga}}$ antisite defects) and holes (by Ga vacancies), as is usually seen in transient reflection/transmission measurements of LT MBE semiconductors. Both the InGaMnAs data and the LT-InGaAs data can be interpreted in this fashion. However, upon close examination, we notice subtle differences between the two. In LT-InGaAs, the data shows a single exponential rise with decay time ∼2 ps. In InGaMnAs, on the other hand, a non-single exponential rising process is observed, which can be seen better in Fig. 2(b), where we plotted the rising component for InGaMnAs in log scale. A bi-exponential curve fitting yields decay times of 2 and 23 ps. Since the present InGaMnAs sample is highly p-type, we tentatively ascribe the differences between InGaMnAs and InGaAs to the different trapping rates for electrons and holes. Namely, the fast component (which is also seen in LT-InGaAs) is due to electron trapping and the slow component (which is seen only in InGaMnAs) is due to hole trapping.

At long times, when the differential reflectivity signal becomes positive, we see that there are periodic oscillations (Fig. 1(a)). We believe that these oscillations are associated with coherent acoustic phonon wavepackets that are generated in the InGaMnAs layer and propagate into the InGaAs layer. The pump pulse creates carriers only in the magnetic layer. Screening of the surface electric fields generates a coherent acoustic phonon wavepacket which propagates into the buffer layer. These oscillations are similar to those seen in GaN/InGaN epilayers [6]. As discussed by Liu et al., the period of the oscillation is related to the wavelength of the probe pulse by $T = \lambda_{\text{probe}}/2C_s n_{\text{ref}}$, where C_s is the speed of sound and n_{ref} the index of refraction in the InGaAs layer. Using values for these parameters, and the wavelength of the probe, we expect the period to be approximately 23–25 ps which is in close agreement with the observed oscillation.

The final decay process after the rising in reflectivity suggests slow recombination of the trapped carriers, which finally vacates the carrier traps. We should also notice that the decaying process of LT-InGaAs is remarkably different from InGaMnAs (Fig. 1(a)). At present, we do not have a picture of why this difference arises.

In summary, we have observed a series of intriguing dynamical processes in ferromagnetic InGaMnAs: (1) an initial decrease in reflectivity due to free carrier effects, (2) a very short decay due to carrier trapping, (3) periodic oscillations in reflectivity due to acoustic phonons, and (4) a final slow recombination process. Through a systematic comparison with low-temperature grown and high-temperature grown InGaAs reference films, we conclude that much of the rich dynamics is due to mid-gap defect states that are introduced in low-temperature growth. Furthermore, the observed oscillations represent the first observation of coherent phonons in a ferromagnetic semiconductor.

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