Ultrafast Dynamics of Coherent Acoustic Phonons in the Ferromagnetic Ga$_{1-x}$Mn$_x$As/GaAs System

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We observed pronounced oscillations in the reflectivity curves of ferromagnetic Ga$_{1-x}$Mn$_x$As/GaAs heterostructures using pump-probe spectroscopy that are caused by coherent acoustic phonons propagating through the sample. The changes in the oscillation period, damping and amplitude as the phonons travel across the Ga$_{1-x}$Mn$_x$As/GaAs interface reflect strong differences in the electronic structures and optical properties of these materials. Analysis of the oscillation amplitude indicates a transition region from Ga$_{1-x}$Mn$_x$As to GaAs substrate.

The introduction of III-V diluted magnetic semiconductors (DMSs) opens up promising opportunities to combine semiconducting properties and robust magnetism into conventional electrical and optical devices, leading to the future development of spin-based devices [1, 2]. (Ga,Mn)As structures are among the highlights of these materials due to their relatively high Curie temperature, and have been extensively studied both experimentally and theoretically [2, 3]. Whereas many time-domain carrier and spin dynamics studies that have been done on (III,Mn)V structures over the years, no systematic time-resolved experiments on phonon dynamics have been attempted. Recently, pronounced coherent acoustic phonon (CAP) oscillations have been reported in GaSb with an InMnAs capping, which, however, only played the role of the source layer in which coherent phonons were generated. In non-magnetic materials, CAPs have been widely observed using pump-probe spectroscopic technique [5, 6, 7, 8, 9, 10]. Here we report the first measurements of acoustic phonons traveling in (III,Mn)V structures that reveal important properties of these materials.

In our setup, the coherent phonons are generated in a thin over-layer of gold by femtosecond pump pulse. The subsequent CAP wave (strain pulse), which alters locally the optical properties, propagates in the Ga$_{1-x}$Mn$_x$As/GaAs and is detected by monitoring the reflected probe pulse. The ferromagnetic Ga$_{1-x}$Mn$_x$As thin layer is grown by a low temperature molecular beam epitaxy (LT-MBE) method. For the samples studied, electron beam evaporation was used to deposit a 5 nm thick film of gold on top of the Ga$_{1-x}$Mn$_x$As in order to generate acoustic waves by optical excitation. Here, we focus primarily on the Ga$_{1-x}$Mn$_x$As (x = 0.024) sample, with a Curie temperature $T_C$ $\approx$ 30 K. Similar results were observed for other samples with Mn concentrations (x=0.018,0.023). All the studied Ga$_{1-x}$Mn$_x$As thin films have a thickness $\sim$ 1$\mu$m.

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All pump-probe experiments measuring transient reflectivity changes $\Delta R/R$ are performed at a temperature of 10 K, employing a Ti:Sapphire laser with a repetition rate of 76 MHz, which produces $\sim$150 fs-wide pulses. Both pump and probe beams were focused onto the sample at the same spot with a diameter of around 100 $\mu$m and an intensity ratio of 10:1. The pump light typically had a fluence of 1.7 $\mu$J/cm$^2$.

Figure 1 shows the transient reflectivity signal $\Delta R/R$ measured in the Ga$_{0.976}$Mn$_{0.024}$As/GaAs at 10 K for 830 nm. Inset: the subtracted oscillatory response divided into three regimes: regimes 1 and 2 with damped oscillations, and transitional regime indicated by the shadow area.

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Figure 1 shows the transient reflectivity signal $\Delta R/R$ measured in the Ga$_{0.976}$Mn$_{0.024}$As/GaAs at 10 K. The inset is the measured oscillatory response, which is obtained by subtracting the thermal relaxation background. Both pump and probe pulses are centered at 830 nm, with a photon energy somewhat below the band gap of GaAs (820 nm). It can be seen that the total response consists of a fast transient (on the order of a few picoseconds) followed by a tail superimposed by two distinct damped oscillatory regimes, separated by a nar-
row transition region. The initial fast transient is typical of the electronic contribution to the pump-probe signal.

In order to determine whether the oscillations originate from ferromagnetism in the Ga$_{0.976}$Mn$_{0.024}$As, an external magnetic field of 0.15 T was applied. No changes in the period of the oscillations were observed. Thus, the oscillatory behavior is not due to a magnetically related mechanism. However, the observed oscillations may be explained by a propagating strain pulse model [4], where the oscillations originate from the interference of probe lights reflected from the top sample surface and the propagating strain pulse, respectively. The reflectivity change $\Delta R/R$ for the oscillatory behavior and its period $T$ take the following forms [2]

$$\Delta R/R \propto Ae^{-t/\tau} \cos(2\pi t/T + \phi), \quad T = \lambda/(2nV_s \cos \theta),$$

where $A$ is the amplitude, $\tau$ is the damping time, $\phi$ is phase shift, $n$ is the refractive index, $V_s$ is speed of sound, and $\theta(=\pi/2$ in our experiment) is the angle of incidence of the probe light in (Ga,Mn)As. In the 'near-field' approximation [5], the damping time $\tau$ is further related to the absorption properties of the material by $\tau = 1/(2nV_s) = \lambda(4\pi V_s \kappa \cos \theta)^{-1}$, where $\alpha$ is the absorption coefficient and $\kappa$ is the imaginary part of the complex refractive index $N(=n+ik)$. The amplitude $A$ is further connected to the change of local complex refractive index in terms of the strain by the following expression [2]

$$A \propto |\frac{\delta N}{\delta \eta_{33}}| \propto |\frac{\delta N}{\delta E_g} \frac{\delta E_g}{\delta \eta_{33}}| \propto \sqrt{\left(\frac{\partial n}{\partial E_g}\right)^2 + \left(\frac{\partial \kappa}{\partial E_g}\right)^2} \frac{\delta E_g}{\delta \eta_{33}},$$

where $\eta_{33}$ is the $z$ component of the strain tensor, and $E_g$ is the bandgap.

The thickness of the top gold layer is small (5 nm), so that we assume the entire Au layer is excited to generate the CAP wave. The generated CAP wave first travels through Ga$_{0.976}$Mn$_{0.024}$As along the normal direction at the speed of the longitudinal acoustic phonon (LAP) $V_s$(Ga$_{0.976}$Mn$_{0.024}$As) before it reaches GaAs. After that, it continuously propagates into the GaAs with LAP speed $V_s$(GaAs). We estimate that it takes about $\Delta t$ ($\approx 210$ ps) for the strain wave to reach GaAs layer if assuming that $V_s$(Ga$_{0.976}$Mn$_{0.024}$As) is approximately equal to $V_s$(GaAs) ($4.78 \times 10^7$ m/s at 10 K [12]). It can be seen from experimental results that 210 ps is around the transition region between the two distinctive damped regimes. So we may reasonably conclude that the two oscillatory regimes 1 and 2 represent propagation of the strain wave inside the Ga$_{0.976}$Mn$_{0.024}$As and GaAs layers, respectively. When the photon energy is less than 1.59 eV (780 nm), the oscillations damped very fast and could not be found in regime 2.

We performed wavelength dependent studies of the oscillations near the bandgap of GaAs(see Figure 3). It can be seen that oscillations in Ga$_{0.976}$Mn$_{0.024}$As decay markedly at all the wavelengths studied. In contrast, the GaAs oscillatory response may persist for very long times at probe wavelengths below the bandgap of GaAs. Applying Eq.(1) separately to the two regimes with damped oscillations, we can numerically fit our experimental data at different wavelengths. Some fitted parameters as a function of wavelength (or photon energy) are given in Figure 3.

Figure 3(a) shows that the oscillation periods for both materials are close to linear versus the probe light wavelength, in good agreement with Eq.(2). This agreement between experiment and theory confirms that the oscillations are only the result of traveling CAPs propagating through Ga$_{0.976}$Mn$_{0.024}$As and GaAs layers continuously. It can be apparently seen from Fig. 3(a) that the period in Ga$_{0.976}$Mn$_{0.024}$As is systematically larger than the period in GaAs at the wavelengths around the bandgap of GaAs. However, the change is astonishingly small (less than 2%) compared to the large doping levels (> 20% cm$^{-3}$). As we know, the speed of LAP waves propagating along [100] is given by $V_s = (C_{11}/\rho)^{1/2}$ for zincblende-structure materials with elastic constant $C_{11}$ and density $\rho$. In the as-grown Ga$_{1-x}$Mn$_x$As ($x > 0.01$) sample, considering the primary substitutional MnGa, and the ~ 20% interstitial Mn [2], its density is roughly the same as that of GaAs. If we further assume that Ga$_{1-x}$Mn$_x$As has the same elastic constant as GaAs [12], the LAP velocity $V_s$ of Ga$_{1-x}$Mn$_x$As approximately equals that of GaAs. Therefore, our results, together with Eq.(2), indicates that introducing Mn into GaAs leads to the reduction of the refractive index $n(\omega)$ compared to that in GaAs. A kink-like feature in $n(\omega)$ profile of GaAs expected by $T(\lambda)$ curve was also demonstrated by previous work [11].

Another experimental result is that the absorption coefficient can be directly obtained from our measured damping times. Analysis of the absorption resonance/band of $\alpha(\omega)$, attributed to different interband or intraband transitions, may reveal the electronic structure of the ferromagnetic Ga$_{1-x}$Mn$_x$As, which has been an important and lively issue over the last several years.

![FIG. 2: Temporal profiles of subtracted oscillatory response for different wavelengths at 10 K.](image-url)
Fig. 3(b) shows our experimental absorption coefficient $\alpha$ in terms of different photon energies for Ga$_{0.976}$Mn$_{0.024}$As and GaAs provided that $V_s = 4.78 \times 10^5$ m/s. As expected, for GaAs it was clearly seen that the sharp step-like absorption feature happens around 1.51 eV, below which $\alpha$ quickly drops close to zero. This observation is in agreement with canonical characteristics of a band model for GaAs with a direct band gap. In contrast, $\alpha(\omega)$ for Ga$_{0.976}$Mn$_{0.024}$As changes rather smoothly in the whole spectral range studied. Especially, no strong abrupt variation was observed at around the bandgap of GaAs. The absorption coefficient below 1.51 eV is still very large and stays almost constant ($\sim 0.66 \times 10^4$ cm$^{-1}$) down to 1.34 eV. The big difference in the light absorption between the ferromagnetic Ga$_{1-x}$Mn$_x$As and the GaAs can be due to the L T-MBE growth technique, where large unintentional defects such as As$_{Ga}$antisites and Mn$_{Ga}$interstitials acting as double donors are introduced except the main substitutional Mn$_{Ga}$. These donor levels provide additional excitation energy levels in the band structure, and lead to the strong broadening of the gap and absorption edge. Our measurements imply that the ferromagnetic Ga$_{1-x}$Mn$_x$As has very different electronic structure and optical properties than those of GaAs. This method, in principle, can be further employed to directly get the information of $n(\omega)$ and $\alpha(\omega)$ in the Ga$_{1-x}$Mn$_x$As system by measuring the period and damping time of the oscillations in the visible, mid-infrared or far-infrared spectra range, and provide an alternative way to understand the band structure of this material in more detail.

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From Figs. 1 and 2 it is clearly seen that at wavelengths close the bandgap of GaAs (a) the oscillation amplitude as a function of wavelength for GaAs suddenly increases, and (b) in the time domain the oscillation amplitudes experience abrupt changes when the strain pulse traveling from Ga$_{0.976}$Mn$_{0.024}$As to GaAs. As shown in Eq. (2), the strain pulse ($\delta N/\delta E_g$) introduces a strong perturbation to the bandgap $E_g$ and induces the local change of the optical properties. Due to the dielectric constants various rapidly around the bandgap $E_g$ of GaAs, $\partial N/\partial E_g$ can experience a fast change as the photon energy $E$ passes through the absorption onset. According to Eq. (2), in the vicinity of the bandgap the amplitude $A$ for GaAs manifests itself a strong peak in $\alpha$ As also discussed above, the optical properties for Ga$_{1-x}$Mn$_x$As are very different with GaAs, e.g. the absorption coefficient for ferromagnetic Ga$_{1-x}$Mn$_x$As varies smoothly in the wavelength range studied. Thus, the change in $\partial N/\partial E_g$ for Ga$_{0.976}$Mn$_{0.024}$As around $E_g$ is much smaller compared to that of GaAs. As a result, when the strain pulse propagates across the Ga$_{0.976}$Mn$_{0.024}$As/GaAs interface, the amplitude of the oscillations exhibits sudden increases for the photon energies around the bandgap of GaAs. This method may be employed to determine unknown film thickness or the longitudinal speed of sound, given that one of them is known.

Finally, we turn to the transition regime in the temporal oscillatory response as shown in Figs. 1 and 2. At wavelengths around the GaAs bandgap, the width of this regime in the time domain is roughly 45 ps, corresponding to a traveling distance of the strain pulse of about 200 nm. The oscillation amplitude in this region increases gradually as time increases. We therefore suspect the existence of a transition region between Ga$_{0.976}$Mn$_{0.024}$As and GaAs substrate. Actually, before depositing the studied Ga$_{1-x}$Mn$_x$As samples, a 100 nm LT-GaAs buffer layer is grown at low temperature (LT) conditions ($\sim 270$ C$^\circ$) following a normal ($\sim 600$ C$^\circ$) 100 nm GaAs buffer layer on GaAs (100) substrate. Due to the As$_{Ga}$antisite defects in LT-GaAs, the amplitude of the oscillatory response due to the strain pulse in the LT-GaAs and GaAs is expected to be different around GaAs bandgap. At the same time, the residual Mn ions inside the chamber lead to the incorporation of a few Mn impurities into these two GaAs buffer layers. Moreover, during growth of Ga$_{1-x}$Mn$_x$As, the Mn on the top could also back-diffuse into these two GaAs buffers. Therefore, all these factors might together provide reasonable explanations of the gradual increasing of the amplitude in the transition regions. Nevertheless, the analysis of the oscillation amplitudes evidently provides a sensitive way to detect the changes of material along growth direction in heterostructure systems.

In conclusion, we describe experimental investigations on pronounced oscillatory behavior in the reflection curves of ferromagnetic Au/Ga$_{1-x}$Mn$_x$/GaAs heterostructures using a pump-probe scheme. The observed oscillations are due solely to coherent acoustic phonon propagation in the (Ga,Mn)As and GaAs layers. Analysis of the oscillatory response indicates strong differences in the electronic structures between the ferromagnetic (Ga,Mn)As and GaAs. Our results are consistent with
the existence of a roughly 200 nm transition layer including LT-GaAs and GaAs buffers.

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