Studies of coherent acoustic phonons in CdMnTe diluted-magnetic single crystals

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Abstract. We report our studies of coherent acoustic phonons (CAPs), generated and time-resolved—detected in semimagnetic Cd$_{1-x}$Mn$_x$Te single crystals by means of femtosecond pump-probe spectroscopy. On crystals with concentrations $x = 0.01$, 0.05 and 0.09, we implemented one-colour spectroscopy, where photon energies of both the pump and probe beams were the same and either just above or below the energy gap of the crystals. For Cd$_{0.91}$Mn$_{0.09}$Te we have also performed two-colour spectroscopy, in which the pump/probe photon energies were far above/below the CdMnTe bandgap, respectively. The dependences of both the frequency and dephasing time of CAP oscillations on the probe wavelength were consistent with the propagating-strain—pulse model. In one-colour experiments, we observed a nonlinear dispersion relationship between the measured CAP frequency and the probe wave vector, which was due to a strong wavelength dependence of the probe beam on the refractive index $n$ near the CdMnTe bandgap. The functional dependence of $n$, extracted from our experiments, could be fitted very well by the Schubert model. Our two-colour pump-probe measurements showed that the penetration depth of probe light, rather than the intrinsic decay time of CAPs, was the dominant source of the oscillation dephasing time. The lifetime of CAPs in our crystals was found to be at least on the order of nanoseconds. We experimentally validated that the electronic stress was the dominant generation mechanism of CAPs in CdMnTe crystals and observed that the value of the sound velocity increased with the increase of Mn-ion concentration in our samples.

1. Introduction

Cd$_{1-x}$Mn$_x$Te (CdMnTe) is a well-known tertiary semimagnetic semiconductor with many desirable traits, such as a wide bandgap tuning range [1], a stable zinc-blend structure up to Mn concentrations $x$ above 0.70, and a possibility of growing samples with very high resistivities [2]. Coherent acoustic phonon (CAP) oscillations were studied in the past [3] and recently long-lived CAP oscillations have been explored in a variety of materials [4, 5]. In this presentation, we discuss our experimental results on optical generation and detection of CAPs in CdMnTe single crystals for a range of different $x$ and for different optical wavelengths of the pump and probe beams.
2. Fabrication and experimental setup

Our single-crystal samples were fabricated using a modified vertical Bridgman method in which the CdMnTe crystals were grown from a seed of (111) orientation in a melt of Cd, Te and desired concentration of Mn. The result is considered very homogeneous due to a near unity segregation coefficient. CdMnTe is $p$-type by nature due to Cd vacancies. These vacancies, and thus, the hole concentration, can be diminished by annealing the sample in Cd and/or by V doping, resulting in not only higher resistivity, but overall higher uniformity [2]. Samples tested include as-grown crystals with $x = 0.01$ and $0.05$, and a Cd-annealed and V-doped crystal with $x = 0.09$. For proper generation of CAPs, all samples had polished surfaces and had a thickness of ~0.7 mm.

Single-colour pump-probe spectroscopy was performed on all three samples in reflection mode. The generation and detection of CAPs were realized by using a 76-MHz repetition rate Ti:Sapphire laser providing 100-fs-wide pulses with a tunable wavelength between 745 nm and 850 nm. Pump pulses were acousto-optically modulated at 243 kHz for lock-in detection. The pump was incident upon the sample, normal to its surface, and with a fluence of ~0.4 $\mu$J/cm$^2$/pulse. The probe beam was incident at $-45^\circ$ to the normal, with a significantly lower fluence of ~0.016 $\mu$J/cm$^2$/pulse, and was considered non-invasive. In order to obtain minimal noise, the pump-beam was cross polarized to the polarizations of both the probe and the analyzer.

In the two-colour pump-probe spectroscopy performed on Cd$_{0.91}$Mn$_{0.09}$Te, the Ti:Sapphire laser was tuned lower than the energy gap $E_g$, and using a BBO crystal, the pump was frequency doubled high above the bandgap. This arrangement allowed probe beam for far below bandgap detection of CAPs, where the probe optical attenuation and dispersion is far less, while maintaining efficient generation of phonons. Furthermore, we decreased pump noise by implementing the optical filter before the analyzer. We measured the time-resolved normalized change in the reflectivity $\Delta R/R$ of the probe beam.
3. Results

The femtosecond pump pulses photoexcite both hole and electron carriers, leading to nonequilibrium conditions and resulting in electronic and thermal stress disturbances. These stresses then propagate as a strained layer of longitudinal acoustic phonons with velocity $v_s$ [3]. The sample’s optical properties within the CAP pulse are altered, causing a discontinuity in the refractive index $n$ of the material. The oscillatory response of $\Delta R/R$ seen in figure 1, is the result of interference between the probe beam’s reflection at the surface of the sample and its reflection at the CAP strained layer. The frequency $f$ of the oscillation is thus:

$$f = \frac{2n v_s}{\lambda_{prb}} \cos(\theta) = \frac{n(\lambda_{prb}) v_s}{\pi} \cos(\theta) k,$$  

where $\lambda_{prb}$ is the probe wavelength, $k$ is the probe wave number, $n(\lambda_{prb})$ is the sample’s real part of the wavelength dependent refractive index, and $\theta$ is the probe’s refracted angle within the sample [3, 5].

The oscillations seen in figure 2 were extracted from the full $\Delta R/R$ signal (figure 1) by removing the electronic exponential relaxation component of the response. The remaining $\Delta R/R$ is characterized by:

$$\frac{\Delta R}{R} = A \sin(2\pi f t - \phi) e^{-t / \tau_d},$$

where the amplitude $A$, and the phase $\phi$, are determined by various parameters and discussed by Wu et al. [6]. $\tau_d$ is the experimental decay time and $1/\tau_d = 1/\tau_{phonon} + \nu \alpha_{prb}$, where $\tau_{phonon}$ is the CAP intrinsic decay time, and $\alpha_{prb}$ is the probe light absorption coefficient. From the fitting of the experimental data (figure 2 solid line) and using equation (2), we can extract $f$ and $\tau_d$.

**Figure 3.** Experimental dispersion curves of $x = 0.01, 0.05,$ and $0.09$. CAP-oscillation frequency vs. probe beam wave number for single-colour (solid squares) and two-colour (empty squares) pump-probe spectroscopy. Dotted lines represent the best linear fit crossing the origin, to the long-wavelength data, and solid lines are fittings based on the Schubert $n$ model.
Figure 3 shows the CAP frequency dependence versus $k$ for crystals of $x = 0.01, 0.05,$ and $0.09$. We observed a dispersive effect when comparing our data to the simple linear fit through zero (dotted line), which we attribute to the wavelength dependence of $n(\lambda_{prb})$. In fact, we were able to obtain a very good, low error fit for $v_s$ (figure 3 solid line) when using equation (1) and Schubert’s dispersion relationship for the index of refraction $[7]$:  

$$n^2 = A + \frac{BE^2}{1 - (E/C)^2}, \tag{3}$$

where $E$ is the probe energy and is less than $E_g$, $A$ and $C$ are functions of $x$, and $B$ is a constant. The results are given in table 1, in which we see an increase in $v_s$ with $x$. For Cd$_{0.91}$Mn$_{0.09}$Te we obtained the most reliable results by implementing the two-colour pump-probe setup, and measuring CAP values in a less dispersive region of the spectrum.

| Crystal              | $E_g$ (eV) | $v_s$ (m/s) | $\alpha_{prb}$ for 0.97$E_g$ (cm$^{-1}$) |
|----------------------|------------|-------------|-----------------------------------|
| Cd$_{0.99}$Mn$_{0.01}$Te | 1.54       | 3444 ± 6.893 | 7080 ± 2165                      |
| Cd$_{0.95}$Mn$_{0.05}$Te | 1.59       | 3480 ± 3.632 | 5311 ± 687.3                     |
| Cd$_{0.91}$Mn$_{0.09}$Te | 1.65       | 3588 ± 2.040 | 4915 ± 572.1                     |

For probe beam energies near $E_g$, it was concluded, that the intrinsic lifetime of the CAP oscillations was much longer than the decay time of the probe, by measuring Cd$_{0.91}$Mn$_{0.09}$Te far below the bandgap where optical attenuation is negligible. The results showed no measurable decay of the oscillations throughout the 400-ps window, suggesting that CAP decay is on the order of at least nanoseconds. Although $\tau_{\text{phonon}}$ is still unknown, we can calculate $\alpha_{prb}$ from the measured values of $\tau_d$ and $v_s$ since the oscillation decay is now reduced to $1/\tau_d = v_s\alpha_{prb}$. In table 1 we present values of $\alpha_{prb}$ obtained at 3% below $E_g$.

In [8], the ratio of the electronic-to-thermal stress was calculated and the former was clearly the dominant generation mechanism of CAPs in CdMnTe. For all three concentrations this has been experimentally validated by the fact that we could detect pronounced CAP oscillations even when the pump-beam energy was below the sample’s bandgap, exciting the band-tail states of the crystal. Furthermore, for Cd$_{0.91}$Mn$_{0.09}$Te, the ratio of CAP oscillation amplitudes measured by a fixed $\lambda_{prb} = 750$ nm, and generated by 3.31 eV photons over those generated by 1.655 eV photons was only 2.6. For a system dominated by thermal stress, a much larger ratio is expected.

Although the current data on Mn concentration dependence is limited, our measurements show the strong dependence of $v_s$ and $\alpha_{prb}$ on $x$. More studies (including samples with high $x$) are needed to fully describe the role of Mn spins on the physics of CAPs within CdMnTe. Further experiments with two-colour pump-probe spectroscopy will also provide better characterization of the intrinsic $\tau_{\text{phonon}}$ of CAPs and further insight to the electronic and thermal stress dynamics.

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