Coherent acoustic phonons in hexagonal $HoMnO_3$ probed by femtosecond spectroscopy

2010 J. Phys.: Conf. Ser. 200 012107

(http://iopscience.iop.org/1742-6596/200/1/012107)

Journal of Physics: Conference Series **200** (2010) 012107 doi:10.1088/1742-6596/200/1/012107

Coherent acoustic phonons in hexagonal $HoMnO_3$ probed by femtosecond spectroscopy

C W Luo[1], H C Shih[1], K H Wu[1], J-Y Lin[2], J Y Juang[1], T M Uen[1], T Kobayashi[1], J M Lee[1,3] and J M Chen[3]

[1]Department of Electrophysics, National Chiao Tung University, Hsinchu, Taiwan, R.O.C.
[2]Institute of Physics, National Chiao Tung University, Hsinchu, Taiwan, R.O.C.
[3]National Synchrotron Radiation Research Center (NSRRC), Hsinchu, Taiwan, R.O.C.

E-mail: cwluo@mail.nctu.edu.tw

Abstract. We have observed the coherent acoustic phonons in the hexagonal manganite $HoMnO_3$ single crystals using wavelength-dependent femtosecond pump-probe spectroscopy. The oscillatory component of the photoinduced transient reflectivity change ($\Delta R/R$) at various pump-probe wavelengths is consistent with the model of strain pulse propagation. Furthermore, the oscillation period of the oscillation is strong temperature- and wavelength-dependent. In particular, the period of all oscillations in $\Delta R/R$ for various wavelengths are very similar in the vicinity of the Néel temperature (T_N), which associates with the appearance of the antiferromagnetic (AFM) long-range ordering.

1. Introduction

Recently, the coexistence of ferroic orders in $Re MnO_3$ with hexagonal (smaller ionic radius of rare-earth Re = Sc, Y, and Ho-Lu) or orthorhombic (larger ionic radius of rare-earth Re = La-Dy) structure not only gives rise to rich physics of the intimate interactions between charge, orbital, lattice, and spin degrees of freedom but also some fascinating emergent physical properties[1–5]. Moreover, the femtosecond optical spectroscopy has been proven a useful tool to resolve the dynamics of each ferroic order directly[6; 7]. In this paper, we performed time-resolved optical pump-probe spectroscopy in hexagonal $HoMnO_3$ (h-$HoMnO_3$) single crystals, where the coherent acoustic phonons are probed at energies close to the d-d optical transition for various temperatures.

2. Experiments

The h-$HoMnO_3$ single crystals were grown by using a traveling solvent optical floating zone method and cleaved into several platelets. The characteristics of crystallinity and magnetization were examined by XRD and SQUID, respectively. A commercial mode-locked Ti-sapphire laser system provided short ($\sim$ 30 fs) pulses with repetition rate of 80 MHz and tunable wavelength at 740 nm ($h\nu$ = 1.69 eV) and 770 nm ($h\nu$ = 1.61 eV). The spectral width of laser pulse (FWHM) is settled to 25 nm in two cases. A standard pump-probe setup has been employed with 40 mW and 2 mW for pump beam and probe beam, respectively. The optical pump beam was focused on the h-$HoMnO_3$ single crystal with the spot-diameter less than 500 μm, and the optical probe beam was overlapped with the spot of pump beam. The polarizations of pump beam and probe

beam perpendicular to each other were parallel to the ab-plane of h-$HoMnO_3$ single crystals (E $\perp$ c-axis). A mechanical delay line was used to vary the arrival time of pump pulses related to probe pulses at the sample. The reflectivity change of the probe beam was detected by using a photodiode detector and a lock-in amplifier.

3. Results and discussion

Figure 1 shows the typical transient reflectivity change (ΔR/R) curves with periodic oscillation. These oscillations in ΔR/R are associated with the propagating strain pulses generated by instantaneous thermal strain using pulsed lasers in an opaque sample[6]. Due to the self-interference of the reflected probe beam from the crystal surface and the surface defined by the propagating strain pulse, the periodic oscillation could be clearly observed in ΔR/R. Namely, the coherent acoustic phonons in h-$HoMnO_3$ single crystals could be generated by the intensive pump pulses and detected by the probe pulses in pump-probe measurements. It is worthwhile mentioning that the oscillation period (τ) is strong wavelength(λ)-dependent. For the case of T = 260 K, the oscillation period (between the dashed lines in Fig. 1) at λ = 770 nm is obviously smaller than that at λ = 740 nm, which is consistent with the results of $LuMnO_3$ reported by Lim *et al.* [6]. However, the oscillation period is wavelength-independent at low temperatures as shown in Fig. 1(b). For T = 100 K, the oscillation period at λ = 740 nm is almost equal to that at λ = 770 nm.

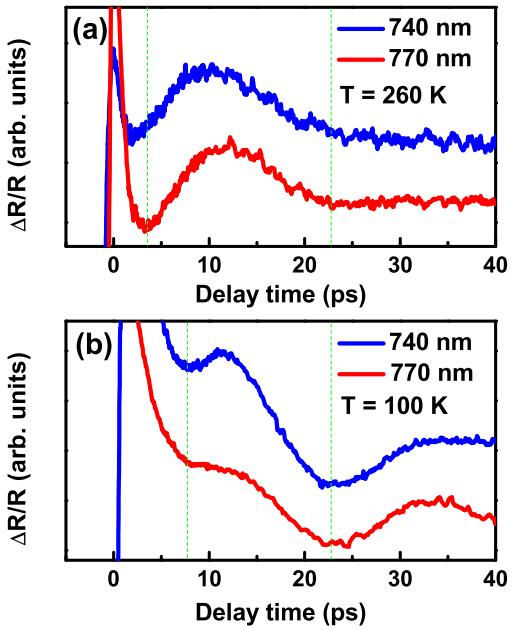

Figure 1. Oscillation in ΔR/R for λ = 770 nm and λ = 740 nm (a) at 260 K, (b) 100 K.

In order to figure out the temperature-evolution of the oscillation period in ΔR/R, all of the data at various temperatures for λ = 770 nm, 740 nm were fitted and quantitatively shown in Fig. 2. At higher temperature ($>$ 160 K), the τ_{740} ($\sim$ 30 ps) is larger than τ_{770} ($\sim$ 26 ps). Furthermore, the oscillation period gradually shrinks as decreasing temperatures. While the temperature close to the Néel temperature ($T_N \sim$ 76 K), both oscillation periods for λ = 770 nm and λ = 740 nm are around 20 ps. This abnormal change around T_N is consistent with the

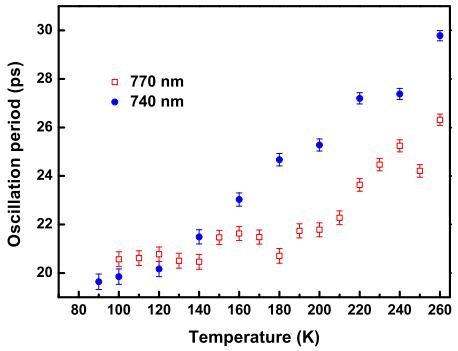

Figure 2. The temperature dependence of oscillation periods in Fig. 1

observed rise in demagnetization time (τ_{dm}) of temperature-dependent ΔR/R, which has been attributed to the coupling to the underlying magnetic order[7].

4. Summary

In summary, we have observed coherent acoustic phonons in hexagonal $HoMnO_3$ single crystals. The dependence of the oscillation periods on the pump-probe wavelengths could be explained by a propagating strain pulse model. It is found that the abnormal shrink of oscillation periods in the transient reflectivity curves associates with the temperature-dependent Mn^{3+} *d-d* optical transition that is coupled to the antiferromagnetic (AFM) long-range ordering.

Acknowledgements

This project is financially sponsored by the National Science Council (grant no. NSC 95-2112-M-009-011-MY3) and the Ministry of Education (2009 MOE ATU program at NCTU) of Taiwan, R.O.C.

References

[1] Hur N, Park S, Sharma P A, Ahn J S, Guha S and Cheong S W 2004 *Nature (London)* **429** 392

[2] Lottermoser T, Lonkai T, Amann U, Hohlwein D, Ihringer J and Fiebig M 2004 *Nature (London)* **430** 541

[3] Cheong S W and Mostovoy M 2007 *Nature Mater.* **6** 13

[4] Lorenz B, Litvinchuk A P, Gospodinov M M and Chu C W 2004 *Phys. Rev. Lett.* **92** 087204

[5] Kimura T, Goto T, Shintani H, Ishizaka K, Arima T and Tokura Y 2003 *Nature (London)* **426** 55

[6] Lim D, Averitt R D, Demsar J, Taylor A J, Hur N and Cheong S W 2003 *Appl. Phys. Lett.* **83** 4800

[7] Shih H C, Lin T H, Luo C W, Lin J Y, Uen T M, Juang J Y, Wu K H, Lee J M, Chen J M and Kobayashi T *Phys. Rev. B accepted*