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Ultrafast Dynamics of the Mn *d-d* Transition on Hexagonal $HoMnO_3$

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Abstract.

The photoinduced changes in reflectivity (ΔR/R) at energies closed to the Mn *d-d* optical transition of $HoMnO_3$ single crystal have been systematically measured from 300 K to 30 K by time-resolved optical pump-probe spectroscopy. The oscillation induced by the propagation of strain pulse was clearly observed in the ΔR/R signal with various probing wavelength. Moreover, the different relaxation processes of photoexcited carriers with in various wavelengths may be led by the blue shift of Mn^{3+} *d* level.

1. Introduction

Recently, the multiferroic rare-earth manganites (ReMnO$_3$) have attracted much attention due to their interesting coupling between the magnetic ordering and electric polarization. Depending on the rare-earth Re^{3+} ionic radius, these intriguing material systems could be classified to a hexagonal phase (Re = Sc, Y, Ho, Er, Tm, Yb, Lu) and an orthorhombic phase (Re = La, Pr, Nd, Eu, Gd, Tb). For hexagonal $HoMnO_3$ (h-HMO), the temperature of a ferroelectric phase transition T_C and an antiferromagnetic (AFM) transition T_N are $\sim$ 900 K and $\sim$ 70 K, respectively. In order to understand the dramatic properties in these multiferroic materials, many remarkable researches, e.g. first-principles calculations [1-3], optical spectroscopy [1, 4, 5], and second harmonic generation studies [6], have been performed unambiguously.

The optical conductivity measurement shows a well-defined absorption peak at photon energy $\sim$1.7 eV, which is assigned to Mn^{3+} $d_{(x^2-y^2),(xy)} \rightarrow d_{(3z^2-r^2)}$ transition in hexagonal manganite ReMnO$_3$. Furthermore, the blue shift of the Mn^{3+} *d-d* transition energy ($\sim$0.15 eV) appears as decreasing temperature [4]. Here, we report that the time-resolved ultrafast carrier dynamics behavior in h-HMO single crystal by using pump-probe technique. Due to the femtosecond time-resolved spectroscopy, the coupling among electrons, phonons, and spins can be directly revealed in the time domain.

2. Experiment

The single crystals of h-HMO_3 were grown by a traveling solvent optical floating zone method and cleaved into several platelet samples. All crystals cut to expose the face of c-axis were characterized by X-ray diffraction (XRD) and magnetization measurements.

For the transient reflectivity change measurements, the samples were mounted in an optical cryostat. A commercial mode-locked Ti-sapphire laser system provided short (∼30 fs) pulses with repetition rate of 80 MHz and tunable photon energy from 1.47 to 1.69 eV. A standard pump-probe setup has been employed with a pump power 50 mW and a probe power of 2 mW. The optical pump beam was focused on the h-HMO single crystal with the spot-diameter less than 1 mm, 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-HMO single crystals. 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.

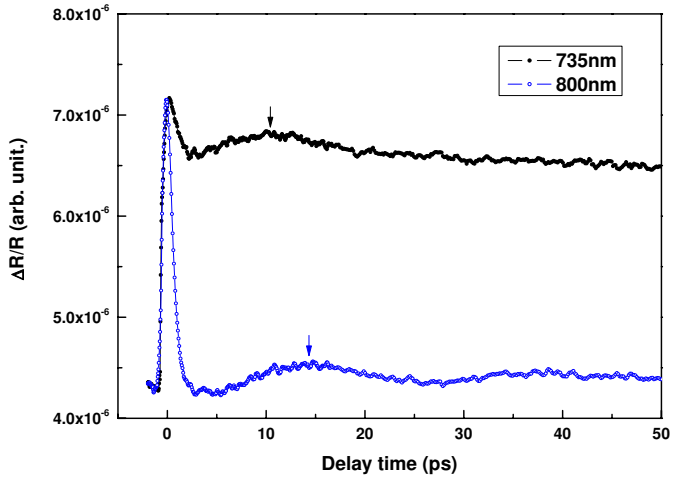

Figure 1. Comparison of the oscillations in transient reflectivity (ΔR/R) curves with pump-probe wavelength at 800 nm (open circles) and 735 nm (solid circles). These data were measured at room temperature.

3. Results and Discussion

Figure 1 shows the ΔR/R signals with periodic oscillations at various wavelengths. There so-called coherent acoustic phonon oscillations assigned to the model of strain pulse propagation have been also observed in many materials, such as multiferroic rare-earth manganites [7], semiconductors [8], and collosal magnetoresistive manganites [9], et al. In the model of strain pulse propagation [10, 11], the λ_{probe}-dependent oscillation period τ_{osc} due to self-interference of the reflected probe pulses from the surface and the bulk of crystals is given by $\tau_{osc} \cong (\lambda_{probe}/2\upsilon_{sound}\sqrt{n^2-\sin^2\theta})$, where the θ is the incident angle of probe beam, υ_{sound} is the sound velocity, and n is the index of refraction of the materials. Also, the ΔR/R in Fig. 1 could be fitted by the following equation to extract the information of oscillation period.

$$\frac{\Delta R}{R}(t, t>0) = A_1 exp(-t/\tau_1) + A_2 exp(-t/\tau_2)\cos(\frac{4\pi n \upsilon_s t}{\lambda} + \phi)$$

We can obtain τ_{osc} = 25.60 ps, and τ_{osc} = 29.52 ps at probe wavelength at 800 nm and 735 nm, respectively, which are consistent with Lim′s results on $LuMnO_3$ [7]. In Fig. 2, the

temperature-dependent ΔR/R curves of h-HMO samples have been systematically measured. The ultrafast responses exhibit totally different carrier dynamic behaviors between different pump-probe wavelengths. In the case of $\lambda_{probe} = 800$ nm, the ΔR/R signals shrink as decreasing temperatures, even the sign of ΔR/R becomes negative when the temperature is lower than 150 K. Similar temperature-dependence in ΔR/R was also observed in another case of $\lambda_{probe} = 735$ nm, but the temperature with sign change was 70 K. This implies that the energy gap between $d_{(x^2-y^2),(xy)} \rightarrow d_{(3z^2-r^2)}$ in Mn^{3+} ions becomes larger than the photon energy 1.55 eV of probe beam in our measurements.

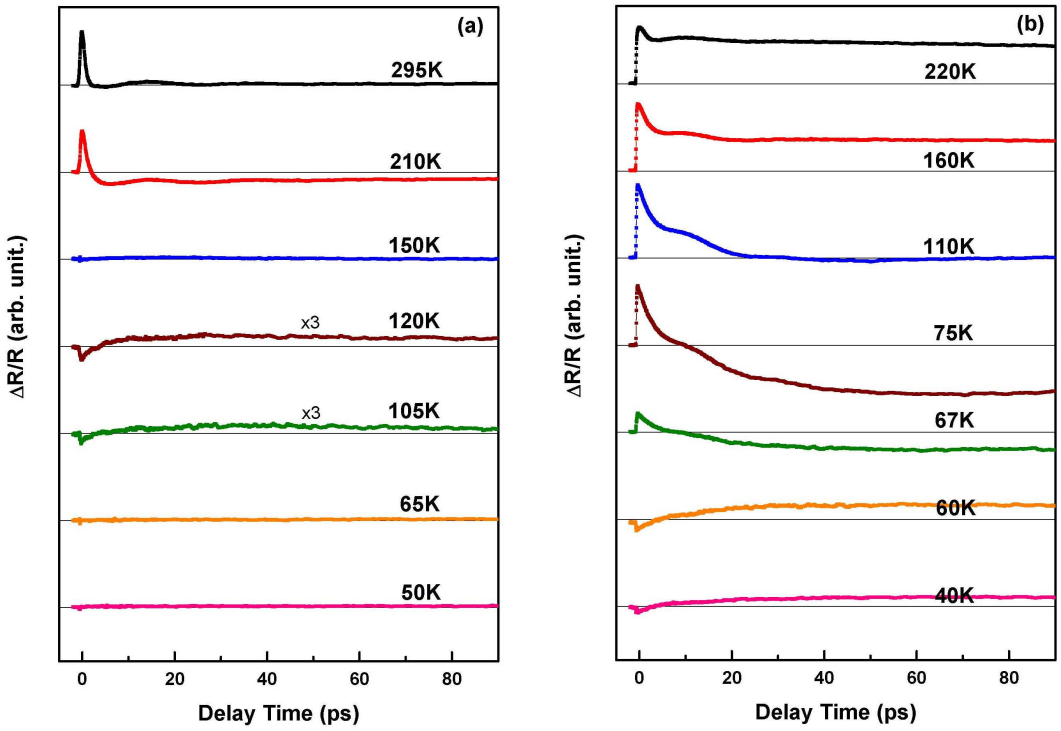

Figure 2. The temperature dependence of the ΔR/R curves for (a) pump at 800 nm and probe at 800 nm. (b) pump at 735 nm and probe at 735 nm.

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