Ultrafast polaron dynamics in $La_{0.7}Ca_{0.3}MnO_3$ thin films

2009 J. Phys.: Conf. Ser. 150 042230

(http://iopscience.iop.org/1742-6596/150/4/042230)

Journal of Physics: Conference Series **150** (2009) 042230 doi:10.1088/1742-6596/150/4/042230

Ultrafast Polaron Dynamics in $La_{0.7}Ca_{0.3}MnO_3$ Thin Films

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

[1]Department of Electrophysics, National Chiao Tung University, Hsinchu 300, Taiwan
[2]Institute of Physics, National Chiao Tung University, Hsinchu 300, Taiwan

E-mail: khwu@cc.nctu.edu.tw

Abstract.

In this work, we use ultrafast optical pump-optical probe (OPOP) spectroscopy to probe the polaron dynamics in $La_{0.7}Ca_{0.3}MnO_3$ (LCMO) thin films. The temporal evolution in transient reflectivity change $\Delta R/R$ exhibits two relaxing components: a fast component with a time constant of subpicosecond and a slow component with time constant ranging from tens of ps to hundreds of ps. The amplitude of the fast component exhibits the similar temperature depence with that of the resistivity, and the neutron scattering intensity due to nanoscale correlated polarons. The results strongly suggest that the fast photoinduced reflectivity change may have been due to the photoexcitation and trapping process of correlated Jahn-Teller (T-J) polarons in the paramagnetic (PM) and ferromagnetic (FM) phases.

1. Introduction

Recently electronic inhomogeneities with a wide variety of length scales (from sub-nanometers to microns) have been recognized as the intrinsic features of manganites [1-3]. These inhomogeneities may result from the coexistence of correlated and uncorrelated local lattice distortions (JahnTeller polarons) in the paramagnetic (PM) phase or the combination of charge-orbital ordered regions and spin ordering (magnetic polarons) domains in the ferromagnetic (FM)/antiferromagnetic (AFM) phases of manganites. Neutron and x-ray scattering techniques are frequently used to explore the existence of electronic inhomogeneities and the distribution of uncorrelated and correlated polarons in manganites at various temperatures. The results strongly suggest that the nanoscale correlated polarons is the dominant contribution to the CMR effect and the competition between various interactions in the electronic inhomogeneities are crucial for the colossal responses in magnetoresistance. Recently, researchers have paid close attention to investigate the strongly correlated electron materials by the ultrafast techniques, since the relative contributions of electron, phonon, and spin dynamics in these materials can be directly resolved in the time domain [4-7]. Moreover, the temperature dependence of the relaxation behavior always demonstrates a dramatic change near the transition temperature, which may reveal the valuable information about the physical mechanisms governing the intriguing properties of these materials. In this work, we use ultrafast OPOP spectroscopy to probe polaron dynamics in $La_{0.7}Ca_{0.3}MnO_3$ thin films. Although this manganite has been investigated extensively by many groups, most of the reports are concerned with a relative slow component, which corresponds to the spin-lattice interaction and the temperature dependence

of the slow relaxation time follows the spin specific heat [4]. There are seldom reports about the fast component. Therefore we will concentrate on the fast component of the relaxation process in the present work.

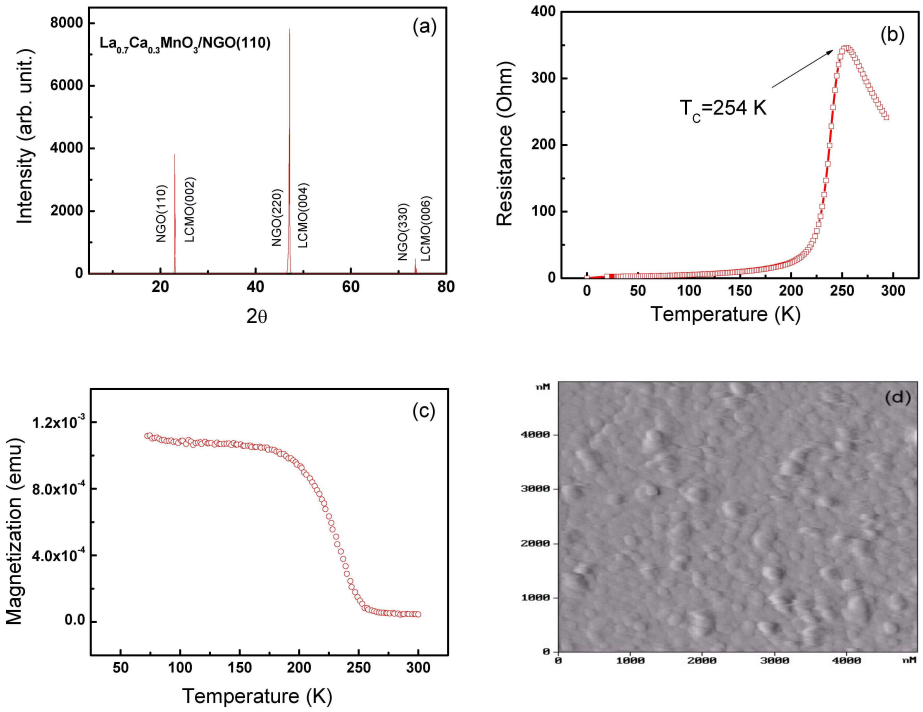

Figure 1. The (a) resistance versus temperature (R-T) curve, (b) magnetization versus temperature (M-T) curve, (c) X-ray diffraction (XRD) pattern, and (d) atomic force microscopy (AFM) image of the $La_{0.7}Ca_{0.3}MnO_3$ sample.

2. Experiment

The pure phase (001) $La_{0.7}Ca_{0.3}MnO_3$ thin film used in this study was prepared by pulsed laser deposition. The deposited LCMO thin film has a Curie temperature $T_C = 255$ K with a thickness of 250 nm. The detailed characteristics of the films, include resistance versus temperature (R-T) curve, magnetization versus temperature (M-T) curve, X-ray diffraction (XRD) pattern, and atomic force microscopy (AFM) image, are shown in Figure 1. The optical pulses were produced by a mode-locked Ti:sapphire laser (Femtolaser, Austria) with a photon energy of 1.55 eV and a 75 MHz train of 20 fs pulses. The ratio between the average power of the pump and probe beams was set at 50:2. The typical energy density of the pump pulses was ~ 5 μJ/cm^2, and the pulses were modulated at 87 KHz with an acousto-optic modulator. The small reflected signals were then detected by a lock-in amplifier [8].

3. Results and Discussion

Figure 2 shows the changes in transient reflectivity ΔR/R curves for a $La_{0.7}Ca_{0.3}MnO_3$ thin film measured at various temperatures. Each ΔR/R curve can be fitted by a superposition of two exponential terms (namely the fast, slow components in this paper) and a quasi-constant component, respectively. In the following, we will focus on the dynamics of fast component in

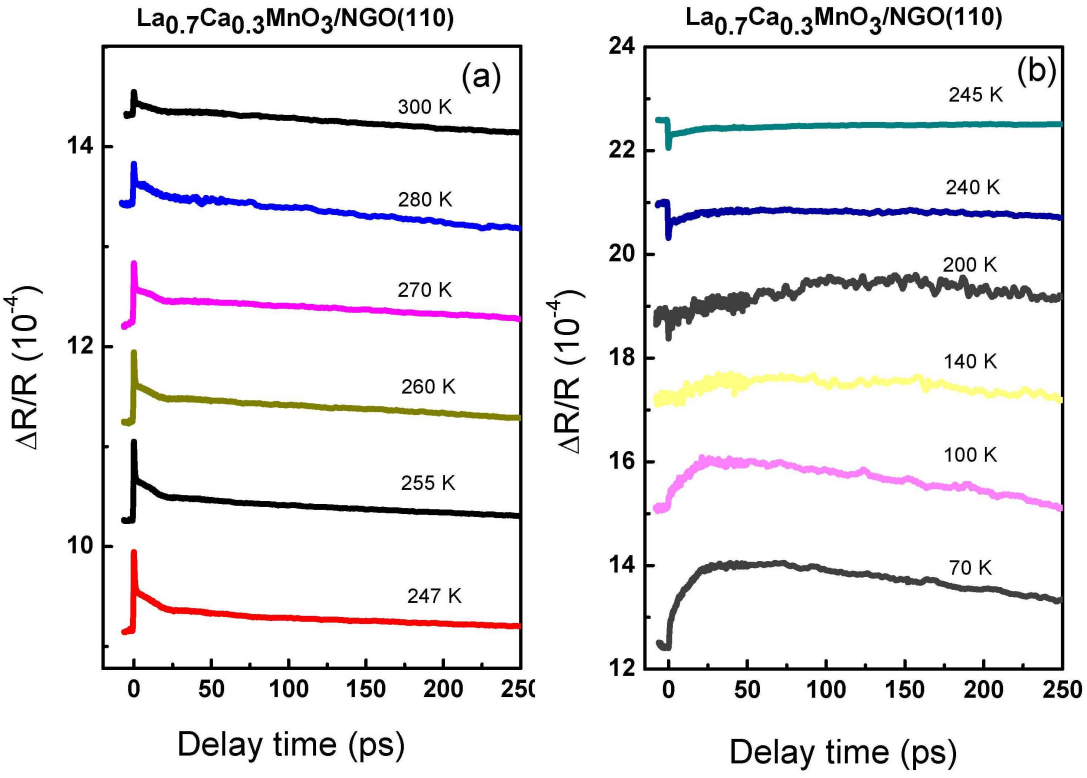

Figure 2. The transient reflectivity ΔR/R curves of a $La_{0.7}Ca_{0.3}MnO_3$ thin film measured at various temperatures.

LCMO. Figure 3 (a) and (b) show the temperature dependence of the amplitude A_{fast}(T) and the relaxation time τ_{fast}(T) of this component, respectively.

At T > T_C, the relatively weaker and gradual decrease of A_{fast}(T) with increasing temperature are primarily due to the decreasing population of the nanoscale correlated polarons and the lower hopping rate in the PM phase. Near T_C and in the PM phase, the number of nanoscale correlated polarons reaches maximum [2], causing A_{fast} to have a peak near the transition temperature. Upon lowering the temperature into the FM phase, Figure 3 shows the A_{fast} has large fluctuations in T_C > T > 220 K, and drop abruptly as temperature decreases to 160 K in the FM phase. The reason for the complicated behavior just below T_C may be due to the rapid changes of the electronic and phase inhomogeneities in this region. For T < 140 K, the spins are almost fully alignd and the double exchange becomes dominant for the electrical conduction, thus the itinerant e_g electrons hop freely between Mn ions. Then the population of polaron and A_{fast} almost disappear in this temperature range. The temperature dependence of the amplitude A_{fast}(T) of the fast component exhibits the similar temperature depencdence of the resistivity, and the neutron scattering intensity due to nanoscale correlated polarons. Therefore, the results strongly suggest that the fast photoinduced reflectivity change may have been due to the photoexcitation and trapping process of correlated Jahn-Teller (T-J) polarons in PM and FM phases and also reveals the electronic inhomogenity and percolative nature of the PM- FM /metal-insulator transition in LCMO maganites.

The relaxation time, which corresponds to the re-trapping time of a photoexcited carrier to its polaronic state, is about 450-600 fs. It is closed to the time predicted by neutron and x-ray

scattering measurements [1]. The relaxation time reduces slightly at T < 160 K, which is the typical value of a conventional metal.

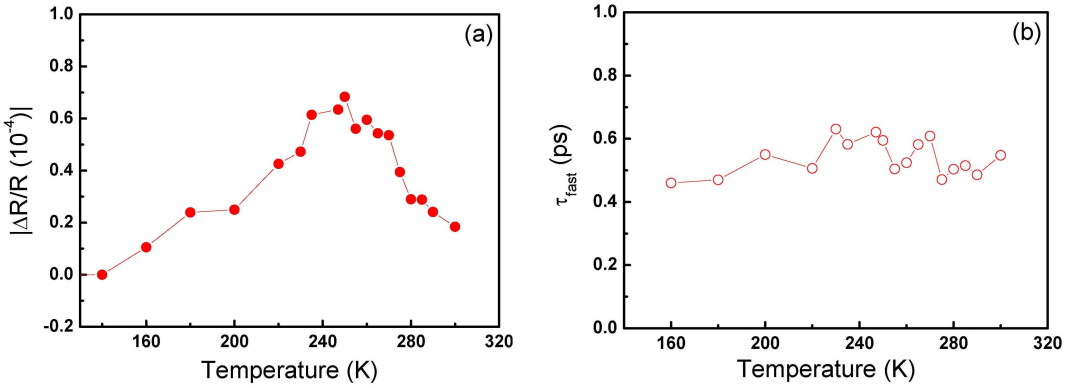

Figure 3. The temperature dependence of (a) amplitude A_{fast}(T) and (b) relaxation time τ_{fast}(T) of the fast component.

4. Summary

In summary, we used ultrafast optical pump-optical probe (OPOP) spectroscopy to probe polaron dynamics in $La_{0.7}Ca_{0.3}MnO_3$ thin films. The temperature dependence of A_{fast} and τ_{fast} of the fast component in $\Delta R/R$ reveals the electronic inhomogenity and percolative nature of the PM-FM/ metal- insulator transition in LCMO maganites. The present study demonstrates that ultrafast OPOP spectroscopy can serve as an alterative experimental technique to investigate the correlated J-T polaron dynamics in the PM and FM phases, which is crucial for understanding the role of polaron in the exotic physical properties of manganites.

Acknowledgements

This project is financially sponsored by National Science Council (grand no. NSC 95-2112-M-009-037-MY3) and Ministry of Eduation (2007 MOE ATU plan).

References

[1] Kiryukhin V, 2004 *New Journal of Physics* **6**, 155
[2] Adams C P, Lynn J W, Mukovskii Y M, Arsenov A A, and Shulyatev D A, 2000 *Phy. Rev. Letts.* **85**, 3954
[3] Shenoy V B, Sarma D D, and Rao C N R, 2006 *ChemPhysChem* **7**, 2053-2059
[4] Averitt R D and Taylor A J, 2002 *J. Phys.: Condens. Matter* **14**, R1357
[5] Hirobe Y, Kubo Y, Kouyama K, Kunugita H, Ema K, Kuwahara H, 2005 *Solid State Communications* **133**, 449
[6] Ren Y H, Zhang X H, Lüpke G, Schneider M, Onellion M, Perakis I E, Hu Y F, and Li Q, 2001 *Phy. Rev. B* **64**, 144401
[7] Ogasawara T, Matsubara M, Tomioka Y, Kuwata-Gonokami M, Okamoto H, and Tokura Y, 2003 *Phys. Rev. B* **68**, 180407(R)
[8] Luo C W, Chen M H, Chen S P, Wu K H, Juang J Y, Lin J-Y, Uen T M, and Gou Y S, 2003 *Phys. Rev. B* **68**, 220508(R)