Abstract

We report a study of the infrared properties of a series of \((\text{La}_{1-y}\text{Pr}_y)_{0.67}\text{Ca}_{0.33}\text{MnO}_3\) \((y = 0, 0.5, 0.6)\) thin films. The samples, grown on NdGaO\(_3\) substrates, showed optically active phonons corresponding to external, bending and stretching modes with no significant temperature shifts. The effective spectral weight, given by \(N_{\text{eff}}(\omega = 0.5 \text{ eV})\) of these films is seen to follow scaling behavior similar to polycrystalline samples.

Published by Elsevier B.V.

PACS: 75.47.Lx; 78.20.-e; 78.30.-j

Keywords: LPCMO; Infrared; Phonons

1. Introduction

The rare earth manganites, \(\text{R}_{1-x}\text{A}_x\text{MnO}_3\) (where \(\text{R}\) is a trivalent rare earth ion and \(\text{A}\) is divalent alkaline-earth ion) have been attracting considerable attention in recent years both because of their versatile physical phenomena and on account of potential device applications. Of special interest are the Insulator to Metal (I–M) transition near the ferromagnetic transition temperature \(T_c\), charge ordering, and colossal magnetoresistance (CMR) (a phenomenon where magnetic fields induce large changes in the resistivity).

The complex interplay between charge, spin, lattice and orbital degrees of freedom precludes a description of these phenomena based only on double exchange (DE). Indeed, dynamic Jahn–Teller coupling [1], polaron absorption [2] and phase separation [3] have been invoked. In this paper we present conductivity analyses of a series of LPCMO thin films to elucidate the nature of electron-phonon coupling mechanisms.

2. Experimental and results

Thin films of \((\text{La}_{1-y}\text{Pr}_y)_{0.67}\text{Ca}_{0.33}\text{MnO}_3\) (LPCMO) \((y = 0, 0.5, 0.6)\) were grown on NdGaO\(_3\) (NGO) substrates by pulsed laser deposition (PLD) in a oxygen atmosphere [4]. Optical reflectance was measured between 30 and 5000 cm\(^{-1}\) over a wide range of temperatures with a Bruker 113v Fourier Transform infrared (FTIR) spectrometer. A continuous-flow cryostat was used to regulate the temperature to within 0.2 K. At all measured temperatures, the absolute reflectance of the substrate was also measured to clarify the effect of the substrate on the observed properties.

The measured reflectance was modeled by using a Drude-Lorentz dielectric function and a least squares minimization to the reflectance was performed, including multilayer reflectance of a film-substrate system, yielding the optical constants.

The extracted conductivity (Fig. 1) spectra show three major transverse optic (TO) phonon modes. These correspond to the three IR active \(F_{1u}\) modes of a cubic perovskite. The external mode, located around 160 cm\(^{-1}\), represents a vibrating motion of the La (Ca,Pr) ions against the MnO\(_6\) octahedra. The bending mode which is associated with Mn–O–Mn bond angle is located around 340 cm\(^{-1}\). This reflects an internal motion of the Mn and O ions along a particular direction against the other oxygen ions in a perpendicular plane. The stretching modes around 570 cm\(^{-1}\) are associated with internal motion of the Mn ion against the oxygen octahedron and is sensitive to the Mn–O bond length.
stretching modes are observed at all temperatures in all samples.

One can also observe a broad mid infrared absorption which leads to an increase in spectral weight at temperatures below $T_c$. This absorption in has been attributed by many authors to a polaron absorption mechanisms \cite{2,6}.

Fig. 2 shows the temperature evolution of the prominent phonon modes in these films. Due to the proximity of the substrate phonon modes to the film phonon modes, the error bars on the frequencies obtained precludes observing significant temperature shifts with temperature as seen in polycrystalline samples \cite{5}.

Fig. 3 shows $N_{\text{eff}}(\omega = 0.5 \text{eV})$ as a function of doping. Similar to polycrystalline samples, there is a systematic decrease in spectral weight as Pr doping is increased. Following Kim et al. \cite{6}, the $N_{\text{eff}}(\omega = 0.5 \text{eV})/T_c$ is plotted.
against $T/T_c$ in the inset of Fig. 3. The $y=0.0$ and 0.5 sample are seen to fall on the same scaling curve. This effect was explained by Roder et al. [7], as the result of coupling between double exchange and Jahn–Teller distortions. In their theory, lattice distortion effects make both $T_c$ and effective electron density depend on the dopant concentration $y$, making $N_{\text{eff}}/T_c$ be directly proportional to the temperature-dependent DE bandwidth. Note that $y=0.6$, which has a phase separated ground state [8] does not fall on the scaling curve. This observed discrepancies may be related to charge ordering or enhanced spin fluctuations near $T_c$ [6].

3. Summary

In conclusion, the temperature evolution of the optical conductivity of LPCMO thin films was investigated as a function of doping. The effective electron density analyzes shows that $y=0.0$ and 0.5 can be described qualitatively by a model of double exchange with coupling to lattice distortions whereas $y=0.6$ may need a different mechanism, such as phase separation, to explain its behavior.

Acknowledgments

NM and DBT acknowledge support from the NSF and DOE through Grants DMR-0305043 and DE-AI02-03ER46070.

References