THz spectroscopic evidence for a charge-density-wave formation in a charge-ordered manganite $Pr_{0.7}Ca_{0.3}MnO_3$

Noriaki Kida [a,b,1], Masayoshi Tonouchi [a,b]

[a] *Research Center for Superconductor Photonics, Osaka University, 2-1 Yamadaoka, Suita, Osaka 565-0871, Japan*
[b] *CREST, Japan Science & Technology Corporation (JST), 2-1 Yamadaoka, Suita, Osaka 565-0871, Japan*

Abstract

Low-energy (0.5–4 meV) charge dynamics of the typical charge-ordered manganite $Pr_{0.7}Ca_{0.3}MnO_3$ have been investigated directly by using THz time-domain spectroscopy. We observed a finite peak structure in the complex optical spectrum around 2–3 meV well below the charge gap $\sim$ 300 meV and attributed it to the collective excitation mode arising from the charge-density-wave condensate.

Key words: THz time-domain spectroscopy; $Pr_{0.7}Ca_{0.3}MnO_3$; charge-density-wave; collective excitation mode

Optical spectroscopy is a powerful tool to reveal the electronic structure in strongly correlated electron system such as colossal magnetoresistive (CMR) manganites [1]. Various kinds of optical studies in the far-infrared to ultraviolet regions have been reported for CMR manganites and clarified that the large electronic construction in the order of eV occurs with temperature and magnetic field [1]. However, the low-energy ($\sim$ meV) charge dynamics in CMR manganites have not examined in detail owing to the difficulty of obtaining the probing light. In this work, we show the results of the low-energy (0.5–4 meV) optical spectra of the typical charge-ordered manganite $Pr_{0.7}Ca_{0.3}MnO_3$ by using THz time-domain spectroscopy (TDS). THz-TDS is a suitable method to capture the low-energy charge dynamics in CMR manganites [2–4] and does not need the Kramers-Kronig transformation, which is an indispensable relation to estimate the complex optical spectrum in conventional optical spectroscopies.

$Pr_{0.7}Ca_{0.3}MnO_3$ thin films on MgO(100) substrates were prepared by a pulsed laser deposition technique. Detailed electrical, magnetic, and THz characteristics of our samples are published elsewhere [4,5]. In THz-TDS experiments, we employed the photoconducting sampling technique in a transmission configuration. THz radiation as a light source is obtained from a dipole-type photoconducting antenna made on low-temperature (LT) GaAs excited by femtosecond optical pulses. Transmitted THz pulse through the sample is detected by the bow-tie-type LT-GaAs photoconducting antenna. Detailed transformation procedures from the data in the time-domain to that in the frequency-domain can be found in Refs. [2–4].

Figure 1 shows the transmission spectra on a logarithmic energy scale for $Pr_{0.7}Ca_{0.3}MnO_3$ thin film on MgO(100) substrate, measured at 4 K. In the measured ω range (0.5–4 meV), optical constants of MgO substrate have negligible variation with ω. As can be clearly seen, we observed a finite peak structure (a strong absorption) centered around 2 meV. The peak frequency is well below the charge gap $\sim$ 300 meV [6,7] and the external optical phonon mode ($\sim$ 20 meV) originated from a simple perovskite structure [7]. To the best of the knowledge, the existence of the finite peak structure in the order of meV has not been reported in CMR manganites so far.

In the following section, we focus our attention on the assignment of this structure. First, we consider the phase separation (PS) picture, in which the volume

[1] Corresponding author. Fax: +81-6-6879-7984. E-mail address: kida@rcsuper.osaka-u.ac.jp (N. Kida)

fraction of the ferromagnetic metallic domains embedded in a charge-ordered insulating phase is thought to determine the anomalous behavior of $Pr_{0.7}Ca_{0.3}MnO_3$ [8]. Typical example of the PS in charge-ordered manganites is a slow relaxation effect [8,9], in which the physical quantities (i.e., resistance and magnetization) are strongly subject to the history of the external perturbations. Very recently, Mathieu *et al.* reported that the magnetization under 20 Oe at 35 K in $Nd_{0.5}Ca_{0.5}MnO_3$ increases slowly in the time range of 10^2 second (slow relaxation effect) after the sample is rapidly cooled down. Moreover, they found that the relaxation curve considerably differs by the cooling rate [9]. In our sample, we observed the ferromagnetic order at 30 K with a coercive force of $\sim$ 500 Oe according to the magnetization measurements. In the framework of this PS picture, the finite peak structure appears as a result of the shifting of the Drude peak centered at $\omega = 0$ as in the case of the impurity-doping at Cu site in copper oxide superconductors: Basov *et al.* reported that the finite peak structure emerges around 10 meV by Zn-doping in $YBa_2Cu_4O_8$ as a consequence of the destruction of superconductivity [10]. Based on these studies, we performed following THz-TDS experiments. The sample is rapidly cooled from room temperature to 4 K with a cooling rate of $\sim$ 8 K per minute and measured the lapsed time dependence of the transmission spectra while keeping 4 K constant. The transmission spectra at selected lapsed time (0, 90, and 250 minute) are shown in Fig. 1. The data are normalized by the value at the absorption peak at 0 minute for comparison. We observed no slow relaxation effect as far as the finite peak structure is concerned; both the peak frequency $\sim$ 2 meV and the spectral shape show no variation per $\sim$ 250 minute in our experimental accuracy. This result provides the clear evidence that the observed structure does not originate from the shifting of the Drude peak even if the PS occurs in our sample.

The other potential candidate for the observed finite peak structure is the collective excitation mode arising from the charge-density-wave (CDW) condensate [11]. This consideration originates from following our experimental findings. (i) the energy scale of the observed structure is nearly same as that of the collective excitation mode observed in low-dimensional materials showing CDW condensate like $K_{0.3}MoO_3$ [11]. (ii) the spectral weight of the observed structure is 2 orders of magnitude smaller than that of the single-particle excitation [4,6,7] (iii) the scattering rate (full width at half maximum) of the observed structure follows the square temperature dependence [4], which is a typical characteristics of the collective excitation mode [11].

In summary, we briefly reported the existence of the finite peak structure around $\sim$ meV in the typical charge-ordered manganite $Pr_{0.7}Ca_{0.3}MnO_3$ by using THz time-domain spectroscopy. By measuring the lapsed time dependence of the observed structure at 4 K after the sample is cooled down with a cooling rate of $\sim$ 8 K per minute, we exclude the possibility that the observed structure originates from the shifting of the Drude peak due to the presence of the ferromagnetic metallic phase in our sample. In analogy to the optical properties of the low-dimensional materials, we assigned the observed structure to the collective excitation mode arising from the CDW condensate. Further detailed measurements are now in progress.

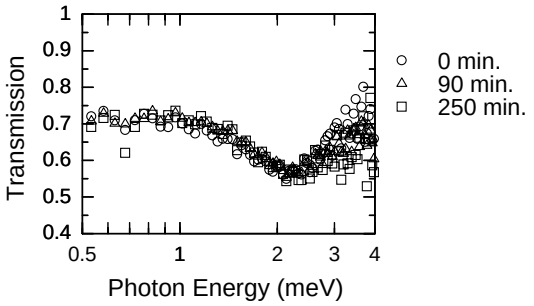

Fig. 1. Logarithmic energy plot of the transmission spectra of $Pr_{0.7}Ca_{0.3}MnO_3$ thin film on MgO(100) substrate as a function of lapsed time [0 (circles), 90 (triangles), and 250 (squares) minute]. The measurements are performed at 4 K after the sample is rapidly cooled down from room temperature with a cooling rate of $\sim$ 8 K per minute.

We thank M. Misra for reading the manuscript.

References

[1] For a review of the optical properties of the manganites, see, Y. Okimoto, Y. Tokura, J. Supercond. **13** (2000) 271.

[2] N. Kida, M. Hangyo, M. Tonouchi, Phys. Rev. B **62** (2000) R11965.

[3] N. Kida, M. Tonouchi, in *Physics in Local Lattice Distortions*, edited by H. Oyanagi, A. Bianconi (American Institute of Physics, New York, 2001), pp366–370.

[4] N. Kida, M. Tonouchi, Phys. Rev. B **66** (2002) 0244XX (in press).

[5] N. Kida, M. Tonouchi, Appl. Phys. Lett. **78** (2001) 4115.

[6] Y. Okimoto, Y. Tomioka, Y. Onose, Y. Otsuka, Y. Tokura, Phys. Rev. B **59** (1998) R9377.

[7] Y. Okimoto, Y. Tomioka, Y. Onose, Y. Otsuka, Y. Tokura, Phys. Rev. B **59** (1999) 7401.

[8] A. Anane, J.-P. Renard, L. Reversat, C. Dupas, P. Veillet, M. Viret, L. Pinsard, A. Revcolevschi, Phys. Rev. B **59** (1999) 77.

[9] R. Mathieu, P. Nordblad, A. R. Raju, C. N. R. Rao, Phys. Rev. B **65** (2002) 132416.

[10] D. N. Basov, B. Dabrowski, T. Timusk, Phys. Rev. Lett. **81** (1998) 2132.

[11] For a review of the CDW dynamics, see, G. Grüner, Rev. Mod. Phys. **60** (1988) 1129.