Magnetoelectrical Transport Properties of $\text{La}_{0.95}\text{Tb}_{0.05}\text{MnO}_3$ Perovskite Materials

KAITUO ZHANG and SHU ZHENG*

Henan Mechnaical and Electrical Engineering College, Electrical Engineering College, Xinxiang 453000, P.R. China

*Corresponding author: E-mail: shuer45@163.com

Received: 24 April 2013;

Accepted: 4 September 2013;

Published online: 30 January 2014;

AJC-14647

Polycrystalline bulk perovskite $\text{La}_{0.95}\text{Tb}_{0.05}\text{MnO}_3$ (LTMO) were synthesized by conventional solid-state reaction. Electric transport and magnetic and dielectric properties were examined. $\text{La}_{0.95}\text{Tb}_{0.05}\text{MnO}_3$ exhibits semiconductor and its magnetic properties were spin-glass like behavior. It possesses the dielectric relaxation at lower 100 K. Furthermore, the dielectric properties remain almost unchanged at higher 100 K.

Keywords: Solid-state reaction, $\text{La}_{0.95}\text{Tb}_{0.05}\text{MnO}_3$, Electrical properties, Perovskite.

INTRODUCTION

In recent years, the structure and oxide films of perovskite manganese are concerned much, because they exhibit unusual electrical transport, magnetic, dielectric and optical properties¹⁻⁴. When Sr^{2+} and Ca^{2+} ions partially substitute LaMnO_3 (LMO) material, it can be found metal transformed into insulator and giant magneto resistance effect at the vicinity of the transition point⁵⁻⁷. In addition, because TbMnO_3 exhibits magneto-electric coupling characteristics, causing a wide-spread interest^{8,9}. The temperature of TbMnO_3 's magneto-electric coupling is very low, which restricts the research and application of the TbMnO_3 system, so workers have been trying to improve the temperature of magneto-electric coupling¹⁰. In this paper, we used equivalence Tb^{3+} to replace the La^{3+} of LaMnO_3 , to study the properties of the electrical transport and dielectric.

EXPERIMENTAL

Preparation of $\text{La}_{0.95}\text{Tb}_{0.05}\text{MnO}_3$ (LTMO): Conventional solid-state reaction was used to prepare $\text{La}_{0.95}\text{Tb}_{0.05}\text{MnO}_3$ ceramic material, we prepared out successfully the single phase LTMO material samples, specific process for preparing LTMO sample is as follows:

Characterization of LTMO: X-ray powder diffraction analysis was performed on a D/max-2200PC X-ray diffractometer.

CuK_α radiation was used with power setting at 30 kV and 20 mA. For crystal phase identification, the typical operation parameters were set to a scan rate of 8° min^{-1} with 0.08 data interval and the scan range from 10° to 80° .

The four-point method was used to measure the electrical transport performance of the samples. First, the surface of the samples were polished and washed with alcohol, so the surface of samples remained clean and smooth. using indium to produce four electrodes on the sample surface. Using liquid nitrogen to cool the samples to the temperature of liquid nitrogen, then measured the curve of resistance - temperature in the slowly warming process.

Bying band field cooling (FC) and zero field cooling (ZFC) measurement modes to measure magnetic susceptibility curve of the samples in the superconducting quantum interference device (SQUID) system.

Using pulsed laser deposition (PLD) process, both the upper and lower surfaces of the sample were coated by a rule layer of gold and made electrode, then measured the dielectric temperature frequency spectrum of the electrode by using dielectric spectrometer Angilent4294A. Measuring temperatures is from 80 K to room temperature, measuring frequency is 2, 5, 10, 50 and 100 kHz, respectively.

RESULTS AND DISCUSSION

Crystal structure: XRD patterns of LTMO samples were shown in Fig. 1. The phase of LTMO sample is pure and its crystal structure belonging to the typical cubic crystal. The corresponding diffraction peaks of XRD spectrum of undoped LaMnO_3 who has the same preparation condition with LTMO moves to the left, when compared with that of LTMO¹¹, which

showed the lattice constant of LTMO was smaller than that of LaMnO_3 . This mainly due to the ionic radius of Tb^{3+} was smaller than that of La^{3+} , further illustrated the Tb^{3+} replaced partially La^{3+} indeed.

Fig. 1. XRD patterns of LTMO and LMO sample

Electrical transport performance: Hong *et al.*¹² first considered the magnetic semiconductor lattice polaron—a magneto-polaron who had the property of lattice distortion. When the size of the lattice distortion associated with polarization was comparable to the sizes of the lattice parameters and unit cell parameters, known as the small-polaron Holstein polaron. Since there were a lot of the same energy positions in the lattice, which could form a localized state band. The band width was very narrow, its migration played a leading role at very low temperatures only, manganese oxide system's polaron transport was generally considered to be adiabatic, in this case, the conductivity of the system as:

$$\sigma = \frac{\sigma_0 T_0}{T} \exp\left(-\frac{E_\sigma}{k_B T}\right)$$

The research results showed the adiabatic small polaron hopping behavior¹³. Later, Blasco *et al.*¹⁴ reported the resistivity of LCMO sample film showed the non-adiabatic small polaron hopping conductivity, its conductivity was:

$$\sigma(T) = \sigma_0 T^\alpha \exp\left(\frac{E_a}{k_B T}\right)$$

Taking the role of the disorder-induced localized into account, Coey *et al.*^{15,16} the high temperature resistance behaviour of CMR manganese oxide was due to Mott's variable-range transition mechanism. In this condition, the resistivity of the sample could be expressed as:

$$\sigma(T) = \sigma_\infty \exp\left[\left(\frac{T_0}{T}\right)^{1/4}\right]$$

For polycrystalline perovskite manganese oxides, because the system there was a complex association, it was difficult to explain the strange electronic transport properties only by the ideal disordered theory and the small polaron theory.

In the study of manganese oxide resistance behaviour, it is found that the type of semiconductor resistance behaviour showed small polaron hopping behaviour. However, in the actual material system, localization caused by the disorder always had a role in the resistance behaviour of the system.

Fig. 2 was $\ln \rho - T^{-1/4}$ curves of LTMO sample under zero and 0.3 T field. Linear fitting result of Mott variable range hopping theory. LTMO exhibited semiconducting behaviour in the temperature range studied. Moreover, the Mott variable-range theory well explained its electrical transport behaviour in the area of high-temperature.

Fig. 2. $\ln \rho - T^{-1/4}$ curves of LTMO sample under zero and 0.3 T field

In studying the electrical transport properties of the manganese oxide, it is found that the resistance behaviour of this type of semiconductor exhibited mainly small polaron hopping behaviour. However, in the actual material system, localization caused by the disorder always worked on the electrical transport properties of the system. In addition, external magnetic field had a huge impact on the electrical transport properties (Fig. 2). LTMO had magneto-resistance effect below the transition temperature.

With Tb^{3+} whose ionic radius was smaller than substituted La^{3+} of LTMO sample, which could cause large distortions of the Mn-O-Mn bond, thus weakened the exchange interaction and reduced electronic exchange integral, so that the transition probability of a carrier dropped. Below the transition temperature, the external magnetic field improved the order degree of the spin-glass state of the magnetic, which made the disorder degree of the magnetic space weakened, thus contributing to the delocalization of the electrons and causing resistance significantly reduce.

Dielectric properties: Fig. 3 consists ϵ -T curves of LTMO, it could be seen from the curve that the dielectric spectrum of the sample, regardless of the real part (ϵ') or the imaginary part (ϵ'') of the dielectric constant had a great change in the vicinity of 100 K. The significant relaxation peaks of ϵ'' appeared in the vicinity of 100 K, while the value of ϵ' was almost no change exceed the transition temperature. In addition, the dielectric relaxation occurs in the vicinity of the magnetic transition temperature. The transition temperature shifted to high temperature gradually as the frequency increasing (Fig. 3),

which were very similar with typical dielectric relaxation and the characteristics of the AC magnetic susceptibility¹². These results indicated that the dielectric relaxation and magnetic relaxation of the sample LTMO might exist some contact, in addition, LTMO sample had a high-temperature dielectric relaxation characteristics. The temperature of relaxation peak and the temperature of magnetic phase transition was same, which showed the dielectric of LTMO might be related to magnetic ordering to some extent near the transition temperature.

Fig. 3. ϵ -T curves of LTMO

In this regard, our group is building a measurement platform to delve into the existence of magnetic coupling in its system.

Conclusion

In this work, the LTMO sample was prepared successfully and analyzed the structure, transport and dielectric properties. The results showed that the crystal structure of the sample was the distortion cubic crystal system. The electrical transport properties exhibited semiconductor characteristics and magnetic ordering structure showed the spin-glass behaviour. In addition, LTMO sample had a high-temperature dielectric relaxation characteristics. The temperature of relaxation peak and the temperature of magnetic phase transition was same, which showed the dielectric of LTMO might be related to magnetic ordering to some extent near the transition temperature. However, it needed to do further research to understand the magnetic structure and properties of LTMO.

REFERENCES

1. W. Kinase and K. Harada, *J. Ferroelectric*, **333**, 21 (2006).
2. A. Asamitsu, Y. Moritomo, R. Kumai, Y. Tomioka and Y. Tokura, *J. Phys. Rev. B*, **54**, 1716 (1996).
3. Y. Tomioka, A. Asamitsu, H. Kuwahara, Y. Moritomo and Y. Tokura, *J. Phys. Rev. B*, **53**, 1689 (1996).
4. G.-M. Gao, C.-L. Chen and X.-L. Deng, *J. Inorg. Mater.*, **22**, 1011 (2007) (in Chinese).
5. T. Shimura, T. Hayashi, Y. Inaguma and M. Itoh, *J. Solid State Chem.*, **124**, 250 (1996).
6. O.Y. Gorbenco, A.R. Kaul, N.A. Babushkina and L.M. Belova, *J. Mater. Chem.*, **7**, 747 (1997).
7. Z.-R. Cai, W. Tong, B. Zhang and Y.-H. Zhang, *J. Inorg. Mater.*, **23**, 8 (2008).
8. T. Kimura, T. Goto, H. Shintani, K. Ishizaka, T. Arima and Y. Tokura, *Nature*, **426**, 55 (2003).
9. T. Goto, T. Kimura, G. Lawes, A.P. Ramirez and Y. Tokura, *Phys. Rev. Lett.*, **92**, 257201 (2004).
10. K. Noda, S. Nakamura, J. Nagayama and H. Kuwahara, *J. Appl. Phys.*, **97**, 10 C (2005).
11. K.H. Ahn and A.J. Millis, *J. Appl. Phys.*, **87**, 5013 (2000).
12. Y.T. Zhang, C.C. Wang and W.B. Liu, *J. Sci. China Ser. G-Phys. Mech. Astron.*, **52**, 1 (2009).
13. H. Zhang, Y.-C. Liu and Z. Huang, *Chinese J. Low Temp. Phys.*, **30**, 217 (2008).
14. J. Blasco, C. Ritter, J. García, J. de Teresa, J. Pérez-Cacho and M. Ibarra, *Phys. Rev. B*, **62**, 5609 (2000).
15. J.M.D. Coey, M. Viret, L. Ranno and K. Ounadjela, *Phys. Rev. Lett.*, **75**, 3910 (1995).
16. M. Viret, L. Ranno and J.M.D. Coey, *Phys. Rev. B*, **55**, 8067 (1997).