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Anomalous magnetostrictive effects in $(La_{1-x}Tb_x)_{2/3}Sr_{1/3}MnO_3$*

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A series of $(La_{1-x}Tb_x)_{2/3}Sr_{1/3}MnO_3$ polycrystalline samples has been studied by means of x-ray diffraction, magnetostriction, and thermal expansion measurements. It has been found that this series undergoes a phase transition from a rhombohedral to an orthorhombic form at the doping level $x \approx 0.20$ at room temperature accompanied by an anisotropic magnetostriction up to -50×10^{-6} under a magnetic field of 1T. The linear and volume magnetostrictions vary with chemical composition, even change sign. At T=80K, the magnetostrictions for the samples of x=0.20 and 0.40 exhibit different behaviours. The sample of x=0.20 has positive volume and linear magnetostrictions and a negative anisotropic magnetostriction, while the sample of x=0.40 has an opposite behaviour. The magnitude of volume magnetostriction for both the samples is essential ($\sim 10^{-4}$) at T=80K under a magnetic field of 4T. We conclude that these anomalous effects are due to the charge delocalization and the structural phase transition between orthorhombic and rhombohedral forms induced by the applied magnetic field.

Keywords: magnetostriction, phase transition
PACC: 7580, 7570

1. Introduction

The perovskite-type manganites $R_{1-x}A_xMnO_3$ (where R= rare earth, A= Ca, Sr, Ba, etc.) exhibit many spectacular properties. The structure and magnetic mechanism have been extensively studied,[1−5] and the colossal magnetoresistance (CMR) in these compounds has been widely reported[6,7]. The understanding of the relevant mechanisms is of great importance due to the promising technological application in CMR devices. Attempts were made to relate this observed CMR to the Zener double exchange. However, Millis *et al*[8] have shown that double exchange alone cannot account for the very large resistivity of the paramagnetic insulator (PMI) phase. Moritomo *et al*[9] investigated the dependence of resistivity and Curie temperature (T_C) of $La_{1-x}Sr_xMnO_3$ (LSMO) on pressure. Their results showed a good similarity between the effects of the magnetic field and the pressure on the temperature-dependent resistivity, which implied a possible large magnetostriction effect near T_C of the material. Afterwards, a large magnetostriction was found in $(Nd_{0.062}Sm_{0.938})_{1/2}Sr_{1/2}MnO_3$[10] and also in $La_{0.60}Y_{0.07}Ca_{0.33}MnO_3$,[11], showing the magnetotransport and magnetovolume properties are intimately related to each other. The magnetostriction technique, therefore, has become an invaluable experimental tool to investigate the electron-lattice interaction in manganites.

In this paper, we report the magnetoelastic properties of $(La_{1-x}Tb_x)_{2/3}Sr_{1/3}MnO_3$ (LTSMO). There are two principal reasons why the study is important: (1) The Tb^{3+} ion has a large orbital moment and gives rise to a giant anisotropic magnetostriction (AMS). (2) The terbium-doped LSMO undergoes a structural phase transition from a rhombohedral to an orthorhombic form with increasing Tb content, which is different from the samples $(La_{1-x}Tb_x)_{2/3}Ca_{1/3}MnO_3$ with the same orthorhombic structure for various compositions[12]. Thus, the structural phase transition in LTSMO compounds may show up new physical phenomena, which is proved by our experimental results.

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2. Experiment

A series of polycrystalline LTSMO ($0 \leq x \leq 0.5$) bulk samples were prepared by conventional solid-state reaction method. Stoichiometric amounts of La_2O_3, Tb_4O_7, $SrCO_3$, and Mn_3O_4 with purities not less than 99.9%, were mixed together and pre-sintered in air at 1000°C for two days. The mixtures were then pressed into pellets and sintered at 1200°C for 15h. x-ray diffraction (XRD) was employed to investigate the structure of the samples. Both linear thermal expansion ($\Delta L/L$) and magnetostriction (MS) were measured using the strain gauge technique. The parallel ($\lambda_{\parallel}$) and perpendicular ($\lambda_{\perp}$) magnetostrictions were measured under the condition of the strain gauge parallel and perpendicular to the magnetic field, respectively. Volume magnetostriction (VMS or ω) and AMS (λ_t) are directly calculated from the formulae $\omega = \lambda_{\parallel} + 2\lambda_{\perp}$ and $\lambda_t = \lambda_{\parallel} - \lambda_{\perp}$, respectively.

3. Results and discussion

Our samples have been characterized by different techniques. These results have been reported elsewhere.[13] The measurement of resistivity versus temperature curve showed that the compound goes through PMI into ferromagnetic-metal (FMM) state near the Curie point (T_C) with decreasing temperature. When a magnetic field H is applied, the resistivity drops sharply, giving rise to a MR value up to 10^3% near T_C at $\mu_0 H$=7T for the sample of x=0.40, the MR is defined as $\Delta\rho/\rho_H = \dfrac{\rho_H - \rho_0}{\rho_H}$, where ρ_0 and ρ_H is the resistivity in the zero and applied field H, respectively. The magnetization measurements showed that the T_C for x=0, 0.05, 0.15, 0.20, 0.25, 0.33, 0.40, 0.50 are 386, 376, 321, 302, 282, 220, 194 and 172K, respectively.

Figure 1 shows typical x-ray diffraction patterns for selected samples. For the sample with x=0, the crystal structure is typical of rhombohedral phase ($R\bar{3}c$ space group), which agrees with an early report.[14] At x ~0.20 the sample undergoes a structural phase transition between a rhombohedral phase and an orthorhombic phase ($Pbnm$ space group) with increasing Tb content. Typical orthorhombic structure was observed in the sample of x=0.40. The XRD patterns show that the lattice constant decreases with increasing x, and the diffraction peak shifts to higher 2θ angle, which has been clearly exhibited in the inset of Fig.1.

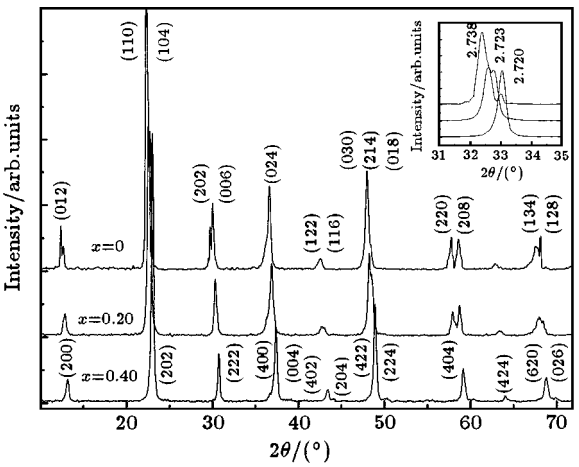


Fig.1. x-ray diffraction patterns for selected $(La_{1-x}Tb_x)_{2/3}Sr_{1/3}MnO_3$ samples (x=0, 0.20, 0.40). The inset shows the amplified XRD patterns of (110).

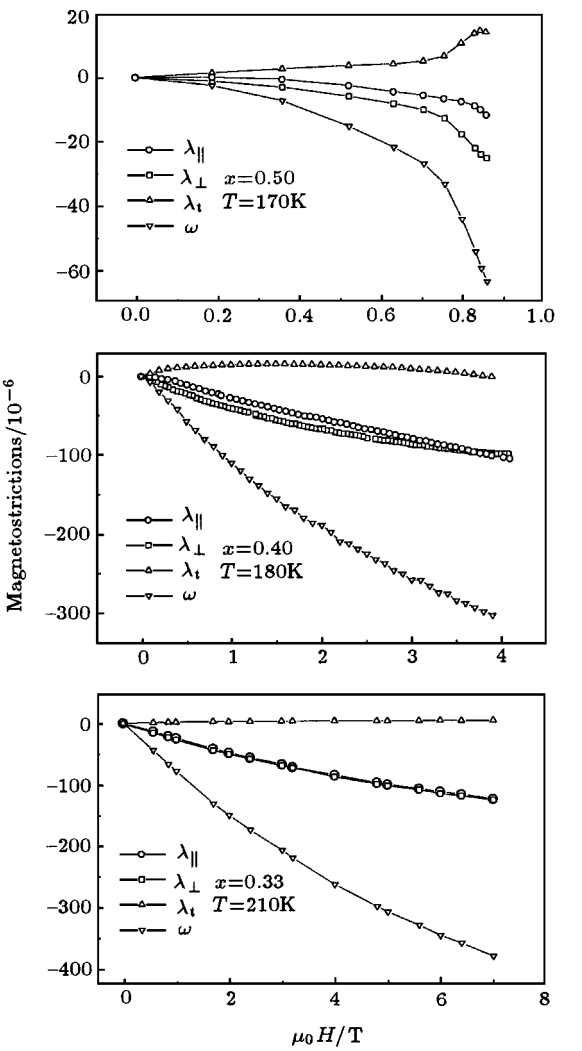


Fig.2. Temperature dependence of magnetostrictions for x=0.33, 0.40 and 0.50 near T_C.

Figure 2 shows the MS (x=0.33, 0.40 and 0.50) as a function of magnetic field at the temperature just

below T_C. Two characteristics can be seen from the figure: first, the MS shows a completely isotropic behaviour ($\lambda_{\parallel} = \lambda_{\perp}$), giving rise to a large VMS; and second, the magnitude of $\lambda_{\parallel}$ and $\lambda_{\perp}$ increases steadily with the magnetic field, having not reached saturation even under a strong magnetic field of 7T.

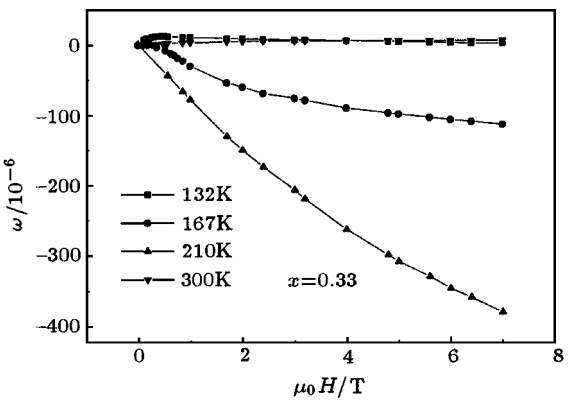


Fig.3. Volume magnetostriction for $(La_{0.67}\ Tb_{0.33})_{2/3}\ Sr_{1/3}\ MnO_3$ versus magnetic field at.

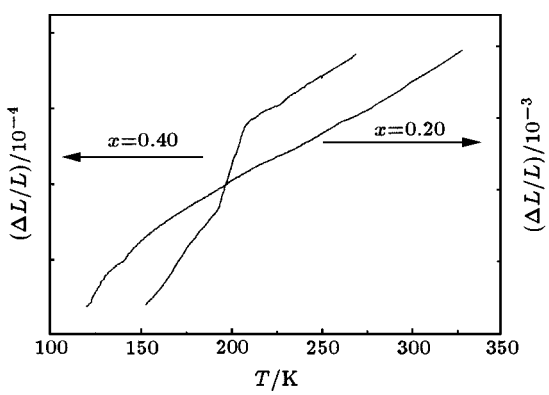


Fig.4. Linear thermal expansion for x=0.20 and 0.40.

The isotherms of VMS for x=0.33 at selected temperatures are shown in Fig.3. As T_C is approached, the magnitude of the VMS increases sharply with the magnetic field and reaches about 400×10^{-6} at $\mu_0 H$=7T and T=210K. The VMSs at the other temperatures are smaller than that near T_C. This behaviour may be attributed to a strong charge-lattice coupling. Because the anomalous VMS and CMR take place around T_C, we consider that this may originate from a carrier localization process accompanied by lattice distortion, as has been observed in $La_{2/3}Ca_{1/3}MnO_3$(LCMO) by De Teresa *et al.*[15] They pointed out that the localization is gradual and produces a local distortion as temperature decreases. Below T_C the establishment of the long-range ferromagnetic order brings the system into a FMM state. The sharp increase of the magnitude of VMS at T_C can be explained as the consequence of the charge delocalization introduced by the applied magnetic field. Thus, the charge localization and the local distortion at zero field are released by applying a magnetic field, causing the CMR and VMS effects near T_C.

Linear thermal expansion curves for x=0.40 and 0.20, as shown in Fig.4, exhibit different behaviours. The thermal expansion curve for x=0.40 exhibits a discontinuity in the vicinity of T_C, while it is continuous for x=0.20. In addition, it is emphasized that the magnitude of the thermal expansion for x=0.20 are five times larger than that of the sample of x=0.40. The difference may be due to the different crystallographic structures between the two specimens. As mentioned above, near T_C the charge delocalization causing an extra contribution to the thermal expansion, so we can observe the anomaly of the thermal expansion curve for x=0.40. While for x=0.20, the specimen is located in the structural phase-transition region near T_C, and the lattice undergoes a structural phase transition between the orthorhombic form (low-temperature phase) and the rhombohedral form (high-temperature phase) accompanied by lattice contraction.[16] This effect cancels the extra contribution to the thermal expansion caused by charge delocalization. So the thermal expansion curve for x=0.20 is continuous. The structural phase-transition effect can be confirmed by AMS results of LTSMO at room temperature, shown in Fig.5. We can see from the figure that the AMS values are smaller except that of x=0.20, which is due to a structural phase transition induced by an external magnetic field, resulting in a large AMS for x=0.20.[16]

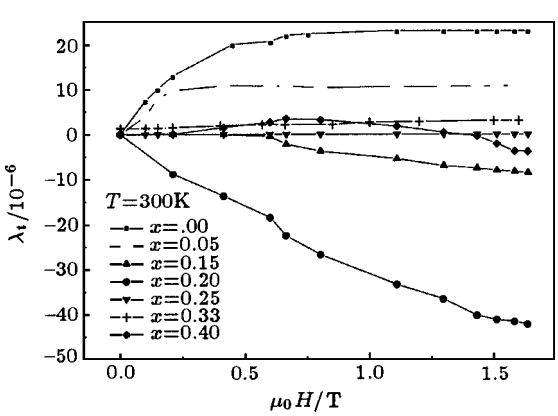


Fig.5. Anisotropic magnetostriction as a function of magnetic field at room temperature.

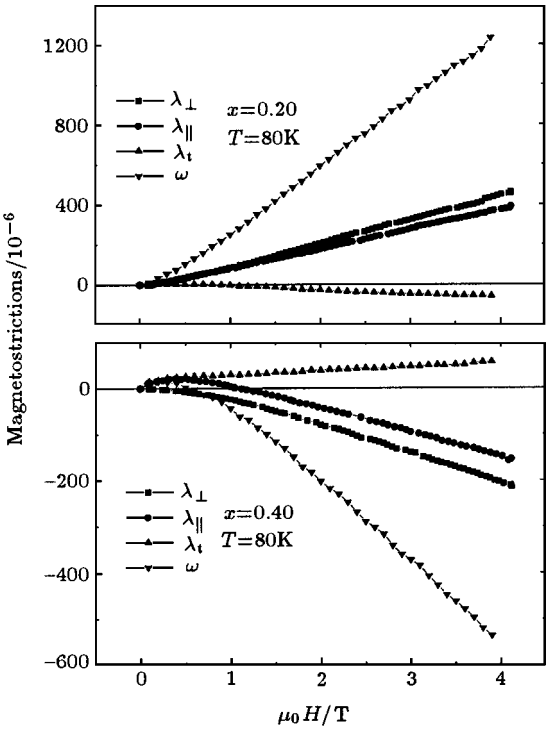


Fig.6. Magnetostrictions (x=0.20 and 0.40) as a function of magnetic field at T=80K.

The MSs (x=0.20 and 0.40) versus the applied magnetic fields at 80K are shown in Fig.6. LTSMO exhibits a large VMS and a medium AMS value at low temperatures, which is different from LCMO.[15] For LCMO, the VMS can be neglected at low temperatures. It is due to the fact that the co-operative Jahn–Teller effect, the exchange interaction and change in the magnetic moment play an important role in MS for LCMO. Whereas, the total MS of LTSMO comes not only from Mn^{3+} and Mn^{4+} ions but also from the trivalent rare-earth ion Tb^{3+}. The MS caused by Mn^{3+} and Mn^{4+} results from anisotropic exchange interaction and the crystalline electric field, the Tb^{3+} ion has a large anisotropic energy and large MS.[17] Therefore, we observe not only a medium MS but also a large VMS for LTSMO at low temperatures. The corresponding signs of MSs for x=0.20 and 0.40 are contrary, and the reason for this needs to be investigated further.

In summary, the anomalous MSs are ascribed to the charge delocalization. At x ~0.20 the specimen undergoes a structural phase transition from an orthorhombic to a rhombohedral form with increasing temperature and/or induced by an external magnetic field, resulting in the unusual thermal expansion and AMS at room temperature. The two structural phases existing in the series of LTSMO may lead to different MSs for x=0.20 and 0.40 samples at low temperatures.

References

[1] Kusters R M, Singleton D A, Mefreevy R and Hayes W 1989 *Physica* B **155** 362

[2] Jonker G H and Van Santen J H 1950 *Physica* **16** 337
Jonker G H 1956 *Physica* **22** 707

[3] Zhang N, Ding W P, Tan X L, Xu Y L, Xing D Y and Du Y W 1998 *Chin. Phys.* **7** 361

[4] Cao Q Q, Wu J, Gu K M, Tan T, Zhang S Y and Du Y W 1999 *Acta Phys. Sin.* **48** S268 (in Chinese)

[5] Zhou Y Q, Lian G J, Zhang Y, Xiong G C, Wang K Q and Ruan K Q 1998 *Chin. Phys.* **7** 919

[6] Okuda T, Asamitsu A, Tomioka Y, Kimura T, Taguchi Y and Tokura Y 1998 *Phys. Rev. Lett.* **81** 3203

[7] Jin S, O'Bryan H M, Tiefel T H, McCormack M and Rhodes W W 1995 *Appl. Phys. Lett.* **66** 382

[8] Millis A J, Littlewood P B and Shraiman B I 1995 *Phys. Rev. Lett.* **74** 5144

[9] Moritomo Y, Asamitsu A and Tokura Y 1995 *Phys. Rev.* B **51** 16491

[10] Kuwahara H, Tomioka Y, Moritomo Y, Asamitsu A, Kasai M, Kumai R and Tokura Y 1996 *Science* **272** 80

[11] Ibarra M R, Algarabel P A, Marquina C, Blasco J and Garcia J 1995 *Phys. Rev. Lett.* **75** 3541

[12] Blasco J, Garcia J, de Teresa J M, Ibarra M R, Algarabel P A and Marquina C 1996 *J. Phys.: Condens. Matter.* **8** 7427

[13] Wu J, Cao Q Q, Gu K M, Li Q L, Wang L, Zhang H C, Du Y W and Zhang S Y 1999 *Acta Phys. Sin.* **48** 370 (in Chinese)

[14] Urushbara A, Moritomo Y, Arima T, Asamitsu A, Kido G and Tokura Y 1995 *Phys. Rev.* B **51** 14103

[15] De Teresa J M, Ibarra M R, Blasco J, Garcia J, Marquina C and Algarabel P A 1996 *Phys. Rev.* B **54** 1187

[16] Asamitsu A, Maritono Y, Tomioka Y, Arima T and Tokura Y 1995 *Nature* **373** 407

[17] Rhyne J J and Legvold S 1965 *Phys. Rev.* **138** A507