Vol 17 No 7, July 2008
1674-1056/2008/17(07)/2717-04

Chinese Physics B

© 2008 Chin. Phys. Soc.
and IOP Publishing Ltd

Spontaneous magnetization and resistivity jumps in bilayered manganite $(La_{0.8}Eu_{0.2})_{4/3}Sr_{5/3}Mn_2O_7$ single crystals*

Zhao Jian-Jun(赵建军)[a)b)†], Lu Yi(鲁 毅)[a)], Haosibayar(浩斯巴雅尔)[a)], Xing Ru(邢 茹)[a)], Yang Ren-Fu(杨仁福)[b)], Li Qing-An(李庆安)[b)], Sun Young(孙 阳)[b)], and Cheng Zhao-Hua(成昭华)[b)]

[a)] *Department of Physics, Baotou Normal University, Baotou* 014030, *China*
[b)] *State Key Laboratory of Magnetism and Beijing National Laboratory for Condensed Matter Physics, Institute of Physics, Chinese Academy of Sciences, Beijing* 100190, *China*

(Received 7 September 2007; revised manuscript received 14 January 2008)

Owing to the inhomogeneous state resulting from the doping of a small number of Eu ions into $La_{4/3}Sr_{5/3}Mn_2O_7$, from the resulting single crystal $(La_{0.8}Eu_{0.2})_{4/3}Sr_{5/3}Mn_2O_7$ we have observed the magnetization jump, the resistivity jump, as well as the relaxation phenomena. For $(La_{0.8}Eu_{0.2})_{4/3}Sr_{5/3}Mn_2O_7$, it has a very delicate ground state due to the interplays among spin, charge, orbital, lattice degrees of freedom. Consequently, the magnetization state is sensitive to temperature, magnetic field, as well as time. Meanwhile, the evolution of the magnetization with time shows a spontaneous jump when both the temperature and the magnetic field are constant. Similar step-like behaviours are also observed in resistivity. All these results suggest that Eu doping can greatly modulate the physical properties of $La_{4/3}Sr_{5/3}Mn_2O_7$ and cause such interesting behaviours.

Keywords: manganite, magnetization jump, resistivity jump, relaxation
PACC: 7530G, 7600, 7530K

1. Introduction

Recently, a great deal of attention has been paid to the bilayered manganites $Ln_{2-2x}Sr_{1+2x}Mn_2O_7$ (Ln=La, Pr, Nd and so on).[1] The bilayered manganites have been proved to be an attractive system for understanding the colossal magnetoresistance (CMR) effect, which are constructed by alternately stacking a nonmagnetic insulating $(La,Sr)_2O_2$ block layer and a ferromagnetic-metallic MnO_2 bilayer sheet along the c axis. The crystalline anisotropy exhibits a strong coupling among magnetism, resistivity and lattice.[2−4] Magnetic and transport measurements have suggested that the properties of bilayered manganites are dependent on both the hole-doping level x and the size of the average A-site ionic $< r_A >$.[5] Especially, for $La_{2-2x}Sr_{1+2x}Mn_2O_7$ with x having a value of about 0.3, the magnetic structure is extremely sensitive to the hole concentration, even accompanied with spatial inhomogeneity.[4]

Some researchers have found an interesting phenomenon, magnetization jump, in some $R_{1-x}A_xMnO_3$ manganites such as $Pr_{0.6}Ca_{0.4}Mn_{0.96}Ga_{0.04}O_3$,[6] $Pr_{0.5}Ca_{0.5}Mn_{0.95}Ga_{0.05}O_3$,[7] and $Pr_{0.5}Ca_{0.5}Mn_{0.97}Ga_{0.03}O_3$.[8] In some bilayered manganites such as $(La_{0.4}Pr_{0.6})_{1.2}Sr_{1.8}Mn_2O_7$ and $(La_{0.5}Nd_{0.5})_{1.2}Sr_{1.8}Mn_2O_7$ crystals, magnetization jumps have also been reported.[9,10] The inhomogeneous structures play an important role in causing the phenomenon. Although the inhomogeneous structure is a generic property of strongly correlated systems, the phenomena (magnetization jump and relaxation) in bilayered manganites are greatly different from their parent compounds.[11] The work about this aspect needs carrying out further. We conjecture that magnetization jumps may be observed in other bilayered manganites. In this letter, we present a study of magnetization and resistivity in a bilayered single crystal $(La_{0.8}Eu_{0.2})_{4/3}Sr_{5/3}Mn_2O_7$ at a low temperature (T=2 K). The step-like behaviours are observed in magnetization and resistivity. In ad-

*Project supported by the National Natural Science Foundation of China (Grant No 50672126) and the Inner Mongolia Natural Science Foundation, China (Grant No 200607010102).
†E-mail: zhaojianjun197174@163.com

dition, a spontaneous magnetization jump is observed in relaxation experiments when both the temperature and the magnetic field are constant. These results indicate that the system has a very fragile ground state due to the competition among spin, orbital, charge and lattice degrees of freedom.

2. Experimental procedures

Single crystals of $(La_{0.8}Eu_{0.2})_{4/3}Sr_{5/3}Mn_2O_7$ were melt-grown in flowing air in a floating-zone optical image furnace. A large crystal with two shiny surfaces was cleaved from one of these crystals. X-ray diffraction (XRD) and back-reflection Laue XRD experiments were carried out to check the single crystallinity and determine the crystallographic direction. A piece of single crystal was cut and chosen to measure its magnetization and resistivity in a commercial physical property measurement system (Quantum Design, PPMS-14). The resistivity was measured by the standard four-point technique.

3. Results and discussion

Figure 1 shows the magnetic-field-dependent magnetization $(M-H)$ curves of the $(La_{0.8}Eu_{0.2})_{4/3}Sr_{5/3}Mn_2O_7$ at 2 K. In these measurements, we observe an abrupt magnetization jump in the single crystal $(La_{0.8}Eu_{0.2})_{4/3}Sr_{5/3}Mn_2O_7$ with $H//ab$-plane. $M-H$ curve shows a jump of nearly 40emu/g at about 7 T, and the in-plane resistivity shows also a downward step at the same field (Fig.2). These data indicate that the entire sample is involved in the step transition, rather than in the isolated regions. In order to reveal the mechanism of the jumps, we pay much attention to the low temperature properties of the crystal. The $M-H$ curves begin with a slow increase in low magnetic fields at 2 K. Then, there occurs a sharp step in the magnetization between 6 and 7 T, offering an evidence of metamagnetic transition. At above 7 T, the magnetization smoothly tends to saturation. At the same time, Fig.1 shows that the field-induced transformation is irreversible as clearly demonstrated by the descending branch of the $M-H$ curve, which is almost flat down to $\sim$1 T and then follows a rapid decrease as H tends to zero. This step-like feature in magnetization is similar to that observed in $Pr_{0.5}Ca_{0.5}Mn_{0.97}Ga_{0.03}O_3$ at 5K.[8]

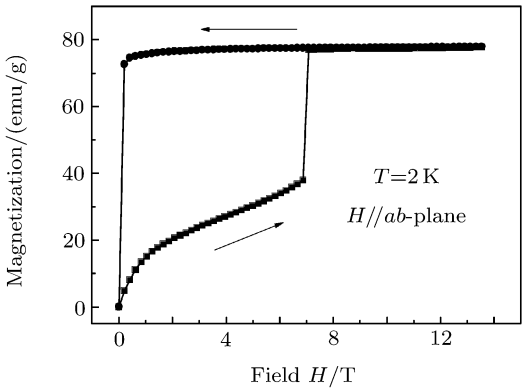


Fig.1. Magnetic-field-dependent magnetization $(M-H)$ curves of single crystal $(La_{0.8}Eu_{0.2})_{4/3}Sr_{5/3}Mn_2O_7$ at 2 K with $H//ab$ plane. For the loop, the magnetic field increases from 0 to 13T first, and then decreases to 0. The arrows indicate the directions of field variation.

Figure 2 shows the magnetic-field-dependent resistivity $(R-H)$ curves at 2 K with $H//ab$-plane. A sudden jump of resistivity is observed at a critical field of 6.7 T which corresponds to a transition field in the magnetization step. The step indicates that there exists a jerky growth of the ferromagnetic fraction in a phase-separation state during martensitic-like transformation.[12,13]

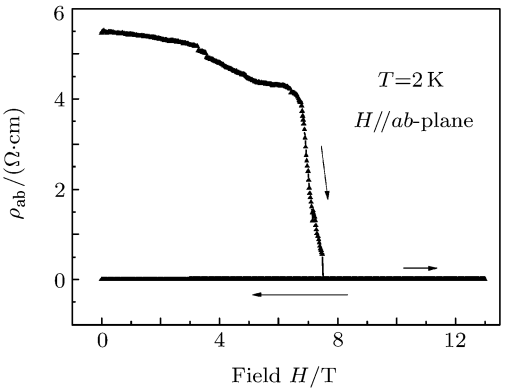


Fig.2. The zero field cooling (ZFC) magnetic-field-dependent resistivity $(R-H)$ curves of $(La_{0.8}Eu_{0.2})_{4/3}Sr_{5/3}Mn_2O_7$ with $H//ab$-plane at 2 K.

In order to further investigate the dynamics of the magnetization jumps, we also measured the time-dependent magnetization and resistivity of the sample with $H//ab$ plane (shown in Figs.3 and 4). The relaxation experiment was done in magnetic fields around 6.2 T at 2 K. The sample was first zero-field cooled (ZFC) from room temperature down to 2 K. After the temperature had been stable, a field of 6.2 T was applied and the magnetization was recorded as a

function of time. There occurred a spectacular magnetization jump as a function of time. Similar result was observed in $(La_{0.5}Nd_{0.5})_{1.2}Sr_{1.8}Mn_2O_7$ and $(La_{0.4}Pr_{0.6})_{1.2}Sr_{1.8}Mn_2O_7$.[10,14] The interesting phenomenon is that this kind of bilayered manganites can display a spontaneous jump in magnetization when both the temperature and magnetic field are constant. The temperature remained at 2.000 ± 0.003K during the relaxation experiments and the magnetic field was very stable during measuring. All the time-dependent magnetization ($M-t$) curves indicate that the magnetization increases with time under a magnetic field at low temperatures. This implies that the crystal tends to translate into a ferromagnetic (FM) state under a magnetic field.[15−17] Meanwhile, the magnetization step field is different from a real critical field for metamagnetic transition, which is possibly attributed to the influence of the average magnetic field sweep rate and field cooling changes.[7,10,12]

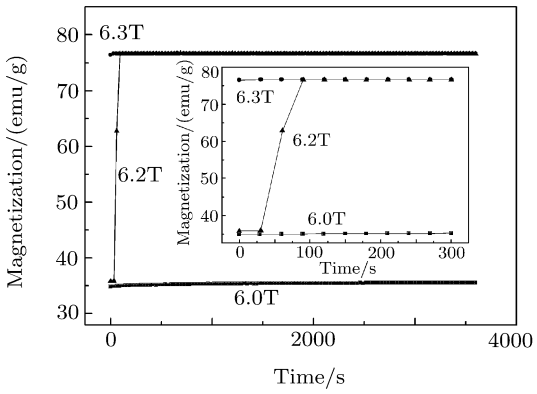


Fig.3. Curves of spontaneous magnetization jump as a function of time recorded at 2 K and different applied fields near the critical field for $(La_{0.8}Eu_{0.2})_{4/3}Sr_{5/3}Mn_2O_7$ with $H//ab$-plane. The inset shows an enlargement of the spontaneous magnetization jump.

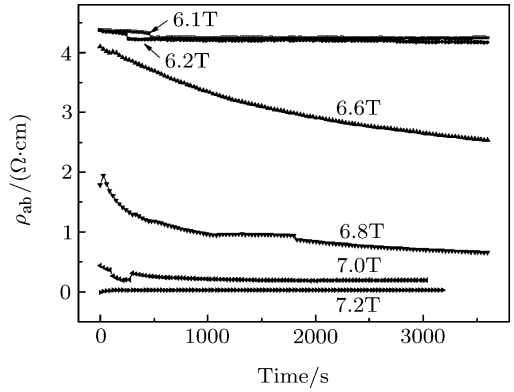


Fig.4. Resistivity as a function of time for $(La_{0.8}Eu_{0.2})_{4/3}Sr_{5/3}Mn_2O_7$ in the ab-plane at 2 K and different applied fields.

In perovskite manganites, their resistivity is closely correlated to magnetization. With the temperature fixed at 2 K, we measured the time-dependent resistivities under several magnetic fields (Fig.4). A spontaneous resistive jump happened in the fields ranging from 6.6 to 6.8 T at 2 K.

Now we turn to the origin of the step-like behaviours. A scenario based on phase separation is proposed. Under zero-field, the ferromagnetic zones without Eu and the antiferromagnetic zones around Eu are separated by spin-glass-like regions.[18−20] However, the coupling between ferromagnetic regions is very weak. Hence the initial magnetization is very small. Because the antiferromagnetic regions have strong distortions induced by Eu, there must be high elastic energy associated with the strains at the antiferromagnetic/spin-glass-like interfaces. The elastic energy tends to prevent the ferromagnetic transition. Upon increasing the magnetic field, the magnetic moments in the ferromagnetic regions are aligned. On the other hand, the boundary of AFM/FM, spin-glass-like regions, is shifting towards the antiferromagnetic regions step by step. In other words, the ferromagnetic regions are enlarged at the cost of the antiferromagnetic component, and the driving force is the applied magnetic field. However, the elastic energy near the surface of the antiferromagnetic regions also increases. Finally, at a critical magnetic field, all the spins in the antiferromagnetic regions realize a sudden reorientation along the direction of applied field. This process generates a magnetization jump. It can be predicted that before the jump, all the magnetic moments in the ferromagnetic regions have directed to the external field. So after the spin reorientation of the antiferromagnetic regions, a fully long-range ferromagnetic state is reached. In the spin reorientation process, the local stress on the surface of the antiferromagnetic regions is released. After the spin reorientation, the Zeeman energy of the system is also lowered. So the fully ferromagnetic state is energetically favourable. As the magnetic field decreases, the long-range ferromagnetic metallic state comes into being.

4. Conclusion

We have realized peculiar step-like magnetization and resistivity jumps in a bilayered single crystal $(La_{0.8}Eu_{0.2})_{4/3}Sr_{5/3}Mn_2O_7$ at a low temperature. In a magnetic field slightly below a critical field, the

magnetic and the resistive relaxations exhibit a spontaneous step after a long incubation time when both the temperature and magnetic field are constant. The phenomena indicate that a small number of Eu ions give rise to strong local lattice distortions, which results in the inhomogeneous state of the system. Our experimental results imply that the local distortions play an important role in changing the properties of bilayer manganites. The reproducibility of the observed phenomena in a range of compounds makes it clear that these jump transitions in the manganites present a challenge to the theoretical circle working on these materials.

References

[1] Moritomo Y, Asamitsu A, Kuwahara H and Tokura Y 1996 *Nature* **380** 141

[2] Argyriou D N, Mitchell J F, Goodenough J B, Chmaissem O, Short S and Jorgensen J D 1997 *Phys. Rev. Lett.* **78** 1568

[3] Mitcell J F, Argyriou D N and Bader S D 1997 *Phys. Rev.* B **55** 63

[4] Konoto M, Kohashi T, Koike K, Arima T, Kaneko Y, Kimura T and Tokura Y 2005 *Phys. Rev.* B **71** 184441

[5] Lu Y, Li Q A, Di N L, Li R W, Ma X, Kou Zh Q and Cheng Zh H 2003 *Chin. Phys.* **12** 1301

[6] Dho Joonghoe, Kim W S and Hur N H 2002 *Phys. Rev.* B **65** 24404

[7] Mahendiran R, Maignan A, Hebert S, Martin C, Hervieu M, Raveau B, Mitchell J F and Schiffer P 2002 *Phys. Rev. Lett.* **89** 286602

[8] Hardy V, Maignan A, Hebert S, Yaicle C, Martin C, Hervieu M, Lees M R, Rowlands G, Paul D McK and Raveau B 2003 *Phys. Rev.* B **68** 220402

[9] Wang F, Gukasov A, Moussa F, Hennion M, Apostu M, Suryanarayanan R and Revcolevschi A 2003 *Phys. Rev. Lett.* **91** 47204

[10] Liao D Q, Sun Y, Yang R F, Li Q A and Cheng Zh H 2006 *Phys. Rev.* B **74** 174434

[11] Yang R F, Sun Y, Di N L, Li Q A and Cheng Zh H 2007 *J. Magn. Mang. Mater.* **309** 149

[12] Hardy V, Majumdar S, Crowe S J, Lees M R, Paul D McK, Herve L, Maignan A, Hebert S, Martin C, Yaicle C, Hervieu M and Raveau B 2004 *Phys. Rev.* B **69** 020407

[13] Xie Y W, Wang D J, Shen B G and Sun J R 2007 *Chin. Phys.* **16** 3120

[14] Gordon I, Wagner P, Moshchalkov V V and Bruynseraede Y 2001 *Phys. Rev.* B **64** 092408

[15] Kim K H, Uehara M, Hess C, Sharma P A and Cheong S-W 2000 *Phys. Rev. Lett.* **84** 2961

[16] Ito A, Aruga H, Torikai E, Kikuchi M, Syono Y and Takei H 1986 *Phys. Rev. Lett.* **57** 483

[17] Jonason K, Vincent E, Hammann J, Bouchaud J P and Nordblad P 1998 *Phys. Rev. Lett.* **81** 3243

[18] Chun S H, Lyanda-Geller Y, Salamon M B, Suryanarayanan R, Dhalenne G and Revcolevschi A 2001 *J. Appl. Phys.* **90** 6307

[19] Li R W, Sun J R, Wang Zh H, Chen X, Zhang Sh Y and Shen B G 2000 *Chin. Phys.* **9** 630

[20] Tan G T, Chen Z H and Zhang X Z 2005 *Acta Phys. Sin.* **54** 379 (in Chinese)