Anisotropic transport and magnetic properties of $Pr_{1/2}Sr_{1/2}MnO_3$ single crystal sample

Cao S.X., Wang X.Y., Zhang Y.F., Yu L.M., Jing C., Zhang J.C., and Shen X. C.

Department of Physics, Shanghai University, Shanghai 200436, China

Resistivity and magnetization for $Pr_{1/2}Sr_{1/2}MnO_3$ single crystal samples were measured in the directions of parallel (//) and perpendicular ($\perp$) to the ferromagnetic (FM) plane. The observed anisotropic transport and magnetic properties in antiferromagnetic (AFM) and FM states can be explained by the double exchange model with d_{x2-y2} orbital structure: with the anisotropic orbital structure, carriers in the FM state or AFM state move only in the FM layers and show two-dimensional metallic behavior. As the A-type AFM state undergoes a transition to a canted spin state below T_N, it also shows spin-flop transition at $T<T_N$.

INTRODUCTION

Perovskite manganites $R_{1-x}A_xMnO_3$ (R and A are trivalent and divalent ions, respectively) have attracted much attention since the rediscovery of the colossal magnetoresistance effect. Beyond the classic double-exchange (DE) mechanism, it has been recognized that the interplay of spin, charge, lattice and orbital is important in these compounds. Among the perovskite manganites that showing colossal magnetoresistance (CMR) properties, $Pr_{1/2}Sr_{1/2}MnO_3$ is of great interest because of its particular magnetic and transport properties. Previous studies showed that this compound exhibits two first-order transitions versus temperature, from paramagnetic (PM) state to ferromagnetic (FM) state at $T_C{\sim}265$ K and from FM to antiferromagnetic (AFM) state at about 140 K [1,2]. The FM state in manganite perovskites is attributed to the double exchange mechanism; localized t_{2g} spins are ferromagnetically coupled to itinerant e_g electrons. An interesting feature of $Pr_{1/2}Sr_{1/2}MnO_3$ compound is that the ground state in low temperature studied by neutron scattering is a layered A-type AFM magnetic structure [3,4], instead of the well known CE-type spin and orbital ordered state. Within the ferromagnetic layers in the A-type AFM structure, one can expect that the DE mechanism is in effect, and that it enhances the conductivity within the FM layers. In addition, the crystal structure of the A-type system favors the d_{x2-y2} orbits, giving rise to two-dimensional band for the e_g electrons. Thus, it can be expected the two-dimensional anisotropic transport and magnetic properties for the A-type AFM structure. In this paper, we reported evident anisotropy of transport and magnetic properties for $Pr_{1/2}Sr_{1/2}MnO_3$ single crystal.

EXPERIMENTAL PROCEDURES

For growing the single crystal samples, the precursor is $Pr_{1/2}Sr_{1/2}MnO_3$ polycrystalline prepared using the conventional solid-state reaction technique. High purity Pr_6O_{11}, $SrCO_3$ and MnO_2 were used as starting materials. A prescribed amount of these powers were thoroughly grinded and mixed homogeneously, then precalcined in air at 1100^oC, 1250^oC for 12 hours respectively, including midst grinding and pressing into pellets. At last, it was sintered at 1300^oC for 24 hours. All the processes were performed in air and cooled with the furnace. The purity and structure of the polycrystalline $Pr_{1/2}Sr_{1/2}MnO_3$ were checked by powder X-ray diffraction method. Then the obtained samples were crushed, grinded to powder and pressed into rod shape under high pressure of about 150 MPa. Single crystal samples of $Pr_{1/2}Sr_{1/2}MnO_3$ were melt-grown using an four-mirror optical floating zone furnace in flowing air with travelling speed of 1~3 mm/h. The feed and seed rods were well-formed $Pr_{1/2}Sr_{1/2}MnO_3$ polycrystalline, rotating in opposite directions at 30

rpm. To release the thermal strain, the as-grown crystal was annealed in flowing oxygen gas at 1200°C for 24 hrs and was slowly cooled to room temperature. The preferential growth direction of the single crystal sample is along the **b** axis, and the **b** axis is known in the FM plane from the studies of structure and orbital schematic of $Pr_{1/2}Sr_{1/2}MnO_3$ compound [3,4]. Thus the boule can be easily divided into pieces along the growth direction and the section has good metallic luster. For all experiments, we use the small sample cut with the edges parallel (//) or perpendicular ($\perp$) to the growth direction (i.e., FM plane) to investigate the anisotropy. The quality of the single crystal samples was characterized by XRD method. Using the Quantum design Physical Property Measurement system (PPMS-9T), Magnetization was measured by means of an induction method using a couple of coaxial pickup coils and resistivity was measured by the standard four-probe method with a dc current.

RESULTS AND DISCUSSION

The crystal structures of the polycrystalline $Pr_{1/2}Sr_{1/2}MnO_3$ at room temperature belong to the monoclinic $P21/n$ structure, consisting of alternating bucking of MnO_6 octahedral along the b-axis. The result of the bulk single crystal sample's XRD diffraction in figure 1 shows only one diffraction peak in the two directions indicates the good quality of the single crystal sample.

In figure 2, temperature dependence of the resistivity at $H=0$ for a current in plane ($\rho_{//}$) and for a current perpendicular to FM plane ($\rho_{\perp}$) are shown. It shows very different transport properties in the two directions, that is, $\rho_{//}$ shows metallic-type transport behavior and insulating-type behavior for $\rho_{\perp}$ in the measured temperature region (4.2~350K), even without any insulator-metal transition like the results reported in $Pr_{1/2}Sr_{1/2}MnO_3$ single crystal [1,2,5].

Figure 1 XRD diffraction patterns of $Pr_{0.5}Sr_{0.5}MnO_3$ single crystal sample in different directions measured at room temperature.

Corresponding to the ferromagnetic transition near T_C~275 K, $\rho_{//}$ decreases dramatically while no visible change for $\rho_{\perp}$. Around antiferromagnetic transition, $\rho_{\perp}$ is distinctly enhanced while $\rho_{//}$ continuously decreases; as a result, the transport anisotropy enhanced by the AFM ordering. It shows that in
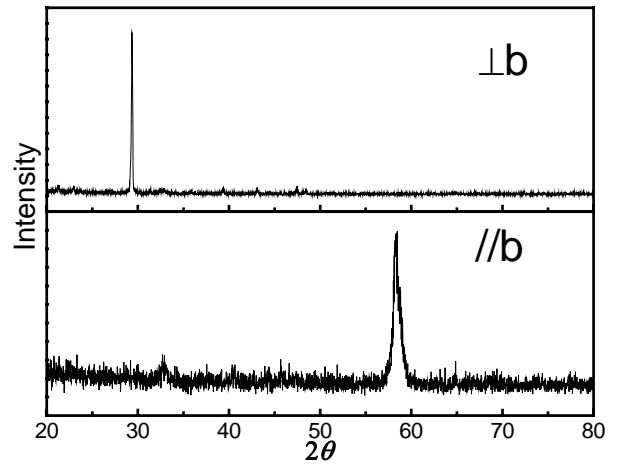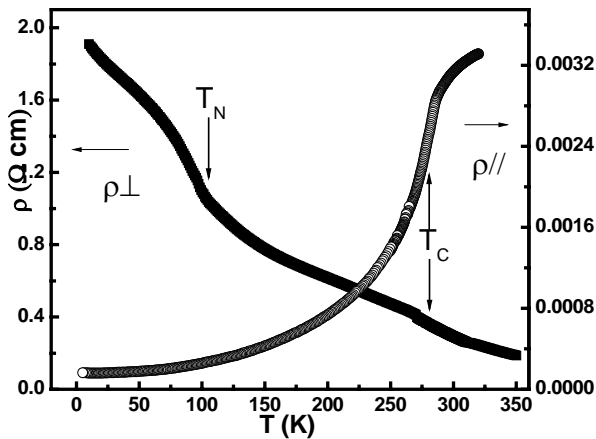

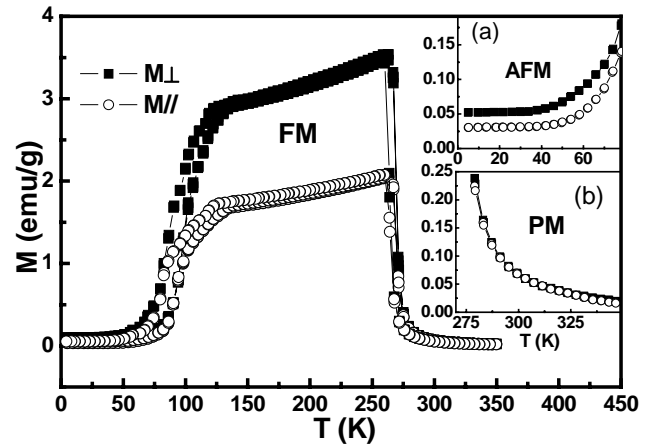Figure 2 Temperature-dependence of resistivity at $H=0$ field. The arrows denote transition temperature.

Figure 3 M-T curves of $Pr_{1/2}Sr_{1/2}MnO_3$ at $H=100$ Oe. The inset (a), (b) shows magnetic properties in AFM state and PM state respectively.

$Pr_{1/2}Sr_{1/2}MnO_3$ system two-dimensional anisotropy of the transport properties exists in the measurement temperature region 4.2~350K.

Temperature dependences of magnetization of the $Pr_{1/2}Sr_{1/2}MnO_3$ single crystal sample under an applied field parallel ($M_{//}$) or perpendicular ($M_\perp$) to the FM plane are shown in figure 3. It is clearly shown that there are two transitions, from paramagnetic (PM) state to ferromagnetic (FM) one at $T\sim275$ K and from FM to antiferromagnetic (AFM) one at about 135 K. The thermal hysteresis between heating and cooling process indicate the first order magnetic transition. The PM–FM transition is followed by a slowly decrease of the magnetization indicating an FM canted spin state at low temperature [6]. In figure 3, noticeable different values of magnetization $M_{//}$ and $M_\perp$ indicate the strong anisotropic magnetic properties in the FM state; there is also anisotropy exist in the AFM state (inset of Fig. 3(a)), while there is not anisotropy in the PM state (inset Fig. 3(b)).

For the two-dimensional anisotropy of the transport and magnetic properties at low temperatures, it can explained based on the A-type AFM ground state with the d_{x2-y2} orbital ordering: DE mechanism within FM layers enhances the hopping probability of the itinerant e_g electrons and decrease the resistivity. Due to the couple of orbital and spin, magnetization shows different values in the two directions.

From a viewpoint of the DE mechanism for the CMR effects, one may expect that the system in the pure ferromagnetic metallic state has no orbital ordering and shows an isotropic behavior by gaining a maximum kinetic energy. If the systems show the d_{x2-y2}-type orbital order, their physical properties should exhibit different directional dependences. Indeed, isotropic spin wave excitations were observed in the FM state of $La_{1-x}Sr_xMnO_3$ with $x\leqslant 0.1$[7]. These results seem to indicate that a transfer integral in these systems is isotropic, and there is no orbital ordering. However, Maezono and Nagaosa [8] suggested theoretically that the orbital ordering could be even a dynamical resonant ("orbital liquid") state in the FM metallic state. By using the Monte Carlo simulation, Yunoki et al.[9] also claimed the existence of an orbital ordered state in the FM phase. Quasielastic and dynamical diffuse scattering study by Kawano-Furukawa et al. [4] showed the spins between the planes exhibit AFM correlations even though the system is still in the FM phase at 220 K in the $Pr_{1/2}Sr_{1/2}MnO_3$ compound. The two-dimensional FM spin correlations develop within the FM planes and A-type AFM correlations develop in the perpendicular direction. These are consistent with the idea that the d_{x2-y2}-type orbital order is formed within FM planes in the $Pr_{1/2}Sr_{1/2}MnO_3$ compound. The strong anisotropic magnetic properties in the FM state shown in Fig. 3 provide a further advantage to identify the effects of the orbital ordering experimentally.

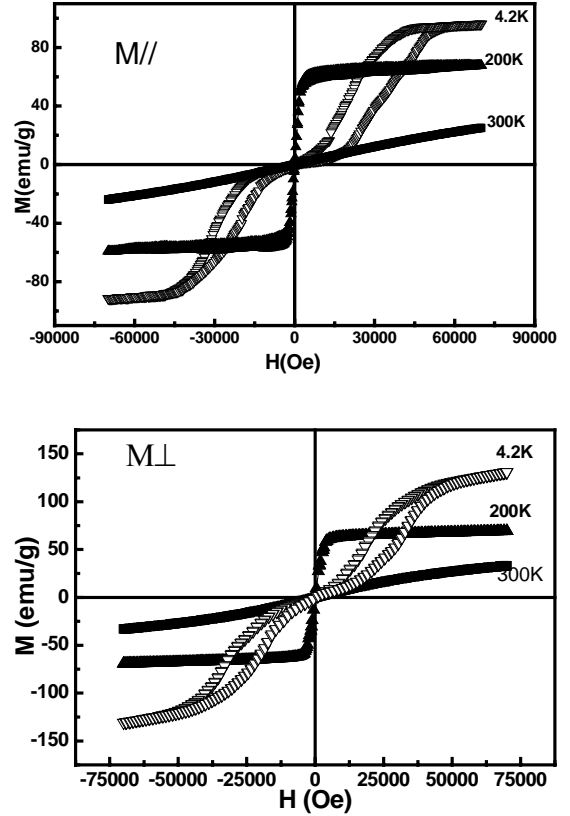Kawano-Furukawa et al. also reported even in the PM phase, the dynamical spin fluctuations are

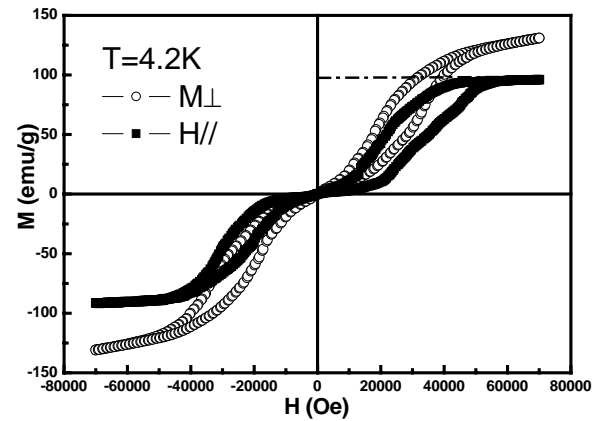Figure 4 M-H Curves of $Pr_{1/2}Sr_{1/2}MnO_3$ at different temperatures.

Figure 5 The anisotropy magnetic properties at T=4.2 K

anisotropy due to the polarization of the d_{x2-y2}–type orbital, but the isotropy in PM state of the present experiment result (as can be seen in Fig. 3(b)) is contrary to their explanation.

In Figure 4, field-dependence of magnetization at fixed temperatures for $M_{//}$ and $M_\perp$ are shown. Compare to the saturation magnetization of $M_{//}$ and $M_\perp$ at 4.2 K, an interesting phenomenon is that $M_\perp$ is larger than $M_{//}$, instead of gradually changing to equality at enough large field as usual. This phenomenon is not found at 200 K and 300 K. The magnetization energy ($E = \int_0^{Ms} HdM$) in the FM plane is larger than that of perpendicular direction (see Fig. 5), i.e., the spins perpendicular to the FM plane can align along the applied field easier than those parallel to the FM plane. As the A-type AFM state (AFM1) undergoes a transition to a canted spin state (AFM2) below T_N [6], it also shows spin–flop transition in M-H curve at $T<T_N$.

SUMMARY

We have measured the resistivity and magnetization for $Pr_{1/2}Sr_{1/2}MnO_3$ single crystal samples in the directions of parallel (//) or perpendicular ($\perp$) to the FM plane. The observed anisotropy of transport and magnetic properties in AFM and FM states can be explained by the DE model with d_{x2-y2} orbital structure: with the nearly anisotropy orbital structure, carriers in the FM state or AFM state can move only in the FM layers and form two-dimension metallic behavior. In such a classical DE model the presence of spin-canted phase is predicted, which is consistent with the magnetization results. As the A-type AFM state undergoes a transition to a canted spin state below T_N, it also shows a spin –flop transition at $T<T_N$.

ACKNOWLEDGMENT

REFERENCES

1. Pollert E., Jirak Z., Hejtmanek J., Strejc A., Kuzel R., and Hardy V., Detailed study of the structural and magnetic transitions in $Pr_{1-x}Sr_xMnO_3$ single crystals ($0.48\leq x \leq0.57$), Journal of Magnetism and Magnetic Materials (2002) 246 290–296
2. Tomioka Y., Asamitsu A., Moritomo Y., Kuwahara H., and Tokura Y., Collapse of a Charge-Ordered State under a Magnetic Field in $Pr_{1/2}Sr_{1/2}MnO_3$, Physical Review Letters (1995) 74 5108-5111
3. Kawano H., Kajimoto R., Yoshizawa., Fernandez-Baca J. A., Tomioka Y., KuwaharaH., and Tokura Y., Two-dimensional anisotropy in a layered metallic antiferromagnet $RE_{1-x}Sr_xMnO_3$ with $x\sim1/2$, Physica B (1998), 241-243 289-294
4. Kawano-Furukara H., Kajimoto R., Yoshizawa H., Tomioka Y., Kuwahara H., and Yokura Y., Orbital order and a canted phase in the paramagnetic and ferromagnetic states of 50% hole-doped colossal magnetoresistance manganites, Physical Review B (2003) 67 174422
5. Argyriou D. N., Hinks D. G., Mitchell J. F., Potter C. D., Schultz A. J., Young D. M., Jorgensen J. D., and Bader S. D., The Room Temperature Crystal Structure of the Perovskite $Pr_{0.5}Sr_{0.5}MnO_3$, Journal of Solid State Chemistry (1996) 124 381-384
6. Hayashi T., Miura N., Tomioka Y., and Tokura Y., Metamagnetic transition and recovery of isotropic transport phenomena in A-type antiferromagnetic $Pr_{0.45}Sr_{0.55}MnO_3$, Journal of Physics and Chemistry of Solids (2002) 63 925-928
7. Martin M. C., Shirane G., Endoh Y., Hirota K., Moritomo Y., and Tokura Y., Magnetism and structural distortion in the $La_{0.7}Sr_{0.3}MnO_3$ metallic ferromagnet, Phys. Rev. B (1996) 53 14285
8. Maezono R., and Nagaosa N., Theory of spin-wave excitation in manganites, Phys. Rev. B (2000) 61 1189-1192
9. Yunoki S., Moreo A., and Dagotto E., Phase Separation Induced by Orbital Degrees of Freedom in Models for Manganites with Jahn-Teller Phonons, Physical Review Letters (1998) 81 5612-5615