Pressure induced canted ferromagnetic order in $La_{1.32}Sr_{1.68}Mn_2O_7$ layered manganite

2010 J. Phys.: Conf. Ser. 200 052013

(http://iopscience.iop.org/1742-6596/200/5/052013)

Pressure induced canted ferromagnetic order in $La_{1.32}Sr_{1.68}Mn_2O_7$ layered manganite

M. Kumaresavanji[1], M. S. Reis[2], Y. T. Xing[1] and M. B. Fontes[1]

[1]Centro Brasileiro de Pesquisas Fisicas, Rua Dr. Xavier Sigaud 150, Urca, RJ, Brazil.
[2]CICECO, University of Aveiro, Aveiro, Portugal.

E-mail: `vanji.hplt@gmail.com`

Abstract. The electrical resistance, magnetization and magnetoresistance of $La_{1.32}Sr_{1.68}Mn_2O_7$ layered manganite have been studied under hydrostatic pressure and magnetic field. At ambient pressure, the temperature dependence of the magnetization shows a ferromagnetic to paramagnetic transition at 118 K. Close to this transition, we have also observed a metal-insulator transition at $T = T^1_{MI}$ in the temperature dependence of the resistivity $\rho(T)$ measurement. The increasing pressure enhances a second metal-insulator transition T^2_{MI} at a critical pressure $6 \leq P_C \leq 7$ kbar. With further increasing pressure, both the T^1_{MI} and T^2_{MI} shift to high temperatures and the peak resistivity suppresses abruptly. We could not observe any transition correspond to T^2_{MI} in the temperature dependence of the magnetization up to 10.2 kbar. A large negative tunneling magnetoresistance was observed around T_C in the field dependence of the $\rho(T)$ curves and the pressure reduces magnetoresistance ratio significantly. From our results, we argue that the increasing pressure induces a canted ferromagnetic order which leads to a ferromagnetic insulating phase at low temperature.

1. Introduction

Recently, interesting results have been reported on the n=2 compounds of the Ruddelsden-Popper series $La_{2-2x}Sr_{1+2x}Mn_2O_7$ in which MnO_2 bilayers and $(La, Sr)_2O_2$ insulating rock-salt layers are stacked alternately [1]. In contrast with the cubic perovskite manganites, layered manganites possess high anisotropic transport and magnetic properties due to the low dimensionality. The crystal structure, electrical transport and magnetic phases of $La_{2-2x}Sr_{1+2x}Mn_2O_7$ have been investigated using X-ray and neutron diffraction by many research groups [2, 3]. Among them, Kubota et al.[2] found that the $La_{2-2x}Sr_{1+2x}Mn_2O_7$ system is a ferromagnetic metal in a narrow doping range $0.30 \leq x \leq 0.48$. The transport and magnetic properties of layered manganites are very sensitive to applied pressure due to the anisotropic compressibility of these materials. Most of the pressure studies have carried out in $La_{2-2x}Sr_{1+2x}Mn_2O_7$ $(0.30 \leq x \leq 0.32)$ layered manganites due to its interesting behaviors at low temperature [4, 5]. Generally, the applied pressure leads to a steep drop of the resistivity accompanying shifts of transition temperature to high temperature values. The change in Mn - O - Mn bond angle and Mn - O bond length and a buckling of the Mn - O(3) - Mn linkage in the ab-plane accompanying a compressibility along the c-axis by the inter-bilayer spacing were also reported on the application of external pressure [6, 7].

In this work, we investigate the hydrostatic pressure and magnetic field effects on electrical transport, magnetization and magnetoresistance at low temperature in $La_{1.32}Sr_{1.68}Mn_2O_7$

layered manganite. Some previous studies reported that $La_{1.32}Sr_{1.68}Mn_2O_7$ undergoes a ferromagnetic to paramagnetic transition at ~ 118 K [1, 2]. At temperatures below ~ 118 K, the magnetic moments on the Mn site are ferromagnetically coupled both between and with in the bilayers.

2. Experimental methods

The $La_{1.32}Sr_{1.68}Mn_2O_7$ polycrystalline sample was prepared by standard high temperature solid-state reaction method. Room temperature powder XRD patterns of the compound were recorded with CuK_α radiation using a Phillips diffractometer in the 2θ range $20\,^\circ$ to $80\,^\circ$. The patterns were analyzed with the Rietveld method using the GSAS program and show a tetragonal system with the unit cell parameters a = 3.8661(3) Å and c = 20.1759(5) Å. The electrical resistance measurements were performed in a Be-Cu self clamp type hydrostatic pressure cell with Fluorinert 75 as a pressure transmitting medium, up to 25 kbar at temperatures from 4.2 K to 300 K. We have used the Pb and Manganin as pressure monometers at low temperature and at room temperature, respectively. Another miniature hydrostatic pressure cell made of nonmagnetic Be-Cu alloy was used for the magnetization measurements in a SQUID magnetometer (Quantum Design Ltd.) up to 10 kbar at temperatures from 5 K to 300 K.

3. Results

3.1. Magnetization under pressure

The magnetic phases of the prepared sample $La_{1.32}Sr_{1.68}Mn_2O_7$ have been verified from M(H) curves, which show a ferromagnetic state at 5 K and a paramagnetic state at room temperature. In figure.1 temperature dependence of the magnetization (field cooled with 100 Oe) is shown,

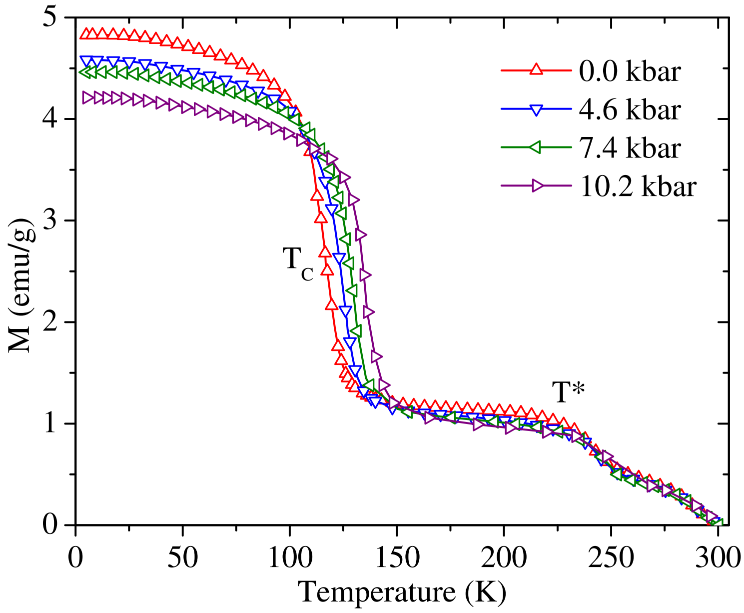

Figure 1. Temperature dependence of the magnetization at different pressures.

which results a sharp ferromagnetic transition (T_C) at 118 K. Even above the T_C, we have observed another transition at T*, which was reported earlier [8]. There is a 2D short range ferromagnetic order between T_C and T* temperatures. We have measured temperature dependence of the magnetization under pressure up to 10.2 kbar. The pressure shifts the T_C to high temperature values at a rate of 1.7 K/kbar and reduces the magnetic moments significantly at low temperature ferromagnetic regime. However, we could not observe any change in T* temperature. Though the pressure changes the 3D ferromagnetic order (T_C) at low temperature, it does not change the 2D ferromagnetic order (T*) existed in MnO_2 sheets.

3.2. Electrical resistivity under pressure and external field

The temperature dependence of the resistivity shows a sharp metal to insulator transition (T^1_{MI}) at 118 K as shown in figure.2, which coincides with T_C. The upturn below T^1_{MI} can be attributed

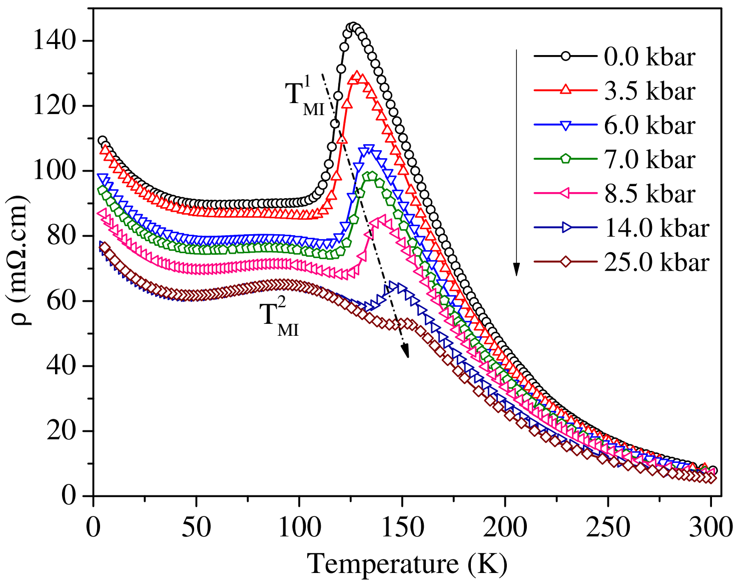

Figure 2. Temperature dependence of the resistivity under pressure.

to the two dimensional confinement of the e_g carriers. The increasing pressure significantly affects the transport behavior, especially at low temperature. The pressure shifts the transition temperature (T^1_{MI}) towards the high temperature values and suppresses the resistance values at T^1_{MI}. At a critical pressure $6 \leq P_C \leq 7$ kbar, a second metal-insulator transition T^2_{MI} appears around 90 K. The T^1_{MI} and T^2_{MI} are shifted with pressure towards the high temperatures at a rate of 1.7 and 1.3 K/kbar, respectively. The amplitude of the peak of the resistivity curves decrease at a rate of -3.63 mΩ/kbar for T^1_{MI} and -1.04 mΩ/kbar for T^2_{MI}. The insulating behavior is fairly suppressed at around T^1_{MI}, reflecting the long-range ferromagnetic spin ordering in the respective MnO_2 sheets.

The field dependence of the resistivity ρ(H) at 4.2 K was measured for 0 kbar, 5.9 kbar, 9.4 kbar and 25 kbar and plotted in figure.3. In the measurement, we have observed a large negative magnetoresistance on the application of field up to 5 T. The magnetoresistance ratio MRR $= \frac{\rho_0 - \rho_H}{\rho_H} \times 100$ decreased from 128% (ambient pressure) to 95% (25 kbar) at 5 T. In figure.4

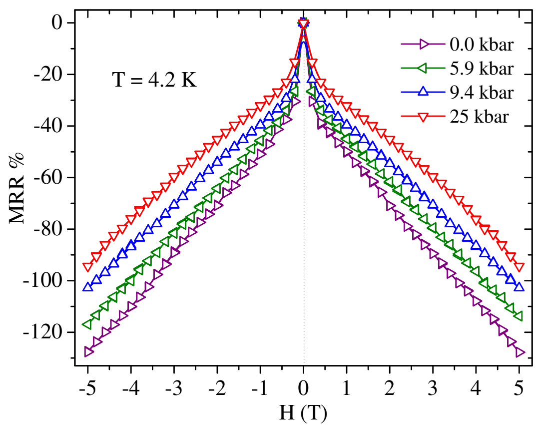

Figure 3. Field dependence of the magnetoresistance ratio at different pressures.

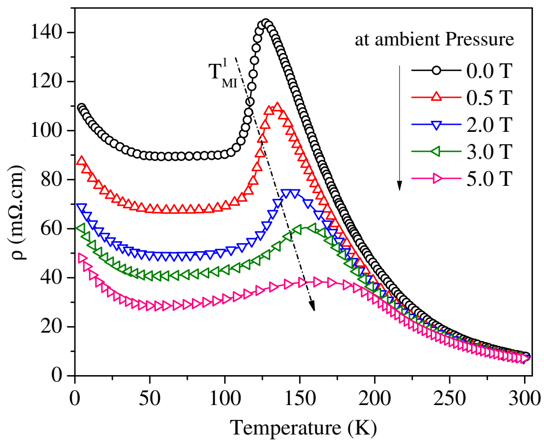

Figure 4. Temperature dependence of the resistivity at different values of magnetic field.

we show the temperature dependence of the resistivity under magnetic field. The measurement does not show any transition at 90 K upon the application of magnetic field up to 5 T at ambient pressure. Also we have measured ρ(T) under magnetic field for the pressures 0 kbar, 5.9 kbar, 9.4 kbar and 25 kbar, which are not plotted here. A large negative tunneling magnetoresistance was observed at T^1_{MI} in all the pressure ranges. The MRR of about 300% was observed at T^1_{MI} for ambient pressure, which is larger than observed at 4.2 K. The ρ(T) curves for all the pressure range have shown a suppression of T^1_{MI} at 5 T field and the increasing pressure reduces the MRR significantly.

4. Discussion

Low temperature ferromagnetic metallic (FMM) phase is explained by the double exchange (DE) mechanism [9] and the high temperature paramagnetic insulating (PMI) phase is explained by the formation of Jahn-Teller polarons due to the distortion of MnO_6 octahedra above T_C [10]. The conductivity in the high temperature paramagnetic region is dominated by the polaronic conduction, arising from the localized charge carriers due to Jahn-Teller distortion. At high pressure the low temperature phase is nearly ferromagnetic (Figure.1), but the ρ(T) curves (Figure.2) show that it becomes an insulator. Many groups reported pressure induced antiferromagnetic (AFM) transition in $La_{2-2x}Sr_{1+2x}Mn_2O_7$ (x = 0.3 and x = 0.32) [5, 11]. In the present work (x = 0.34), we could not observe any AFM transition induced by pressure. The $La_{1.32}Sr_{1.68}Mn_2O_7$ compound is a simple ferromagnetic coupled bilayer for all the temperature range below T_C. It is obvious that the pressure changes Mn - O bond length and Mn - O - Mn bond angle significantly, which will modify the spin ordering in the MnO_2 layers. The DE is possible when t_{2g} spins are aligned in a parallel manner. The hopping amplitude $t = t_0 \cos(\theta/2)$ is maximum when the spins are aligned parallel, where θ is the angle between spins placed at the neighboring t_{2g} states [12]. If the spins are aligned antiparallel (t = 0), then there is no hopping of electron for DE. In our case, the tilted MnO_6 octahedra do not participate effectively in DE, due to its canted spin configuration ($\theta \neq 0$). Thus, we have observed a canted ferromagnetic insulating phase (CFMI) induced by the pressure. Even though the DE model predicts that a canted ferromagnetic state may evolve from an antiferromagnetic state [13], we are unable to claim the existence of antiferromagnetic state from the pressure dependence of M(T) curves

(Figure.1). The reduced magnetic moments by pressure is also an evidence of weakening the ferromagnetic DE interaction. Also, the ρ(T) upon application of field (Figure. 4) elucidate our results obtained previously.

Since, the bilayer manganites consist of the ferromagnetic metal - insulator - ferromagnetic metal junction, the large negative magnetoresistance is explained in terms of the inter bilayer tunneling magnetoresistance effect (TMR) because of the high spin polarization of conduction electrons [14]. The change in tunneling resistance is closely related to the spin polarization of conduction electrons in each FM layer. The ferromagnetically coupled interlayer parts act as a leaky current path along the c-axis. By applying a magnetic field, the magnetization process removes carrier blocking $[(\mathrm{La},\mathrm{Sr})_2\mathrm{O}_2]$ boundaries and allows the inter-bilayer tunneling of spin polarized electrons. However, we observed a decrease in MRR with increasing pressure, this reduction is of 30% compared with ambient pressure. The tunneling of conduction electrons are highly depends on the orientation of the FM layers. Few studies have been reported on the relative change in spin polarized tunneling with the intra layer orientations [15]. The tunneling of electrons are higher when the intra layers are ferromagnetically coupled and lower for the anti-parallel orientation. Since, the pressure induces a canted ferromagnetic spin state in our compound, the tunneling is not taking place efficiently at inter-layer. Therefore, the MRR decreases with pressure.

5. Conclusion

We have synthesized a bilayer manganite and measured the electrical transport and magnetic properties under pressure and magnetic field. The pressure induces a second metal-insulator transition in ρ(T) curves. Further, we have measured the magnetization under pressure, in which we could not observe any second transition even up to 10.2 kbar. It reveals that at high pressure the low temperature phase is nearly ferromagnetic, but the ρ(T) curves show that it becomes an insulator. The applied pressure tilted MnO_6 octahedra and established a canted ferromagnetic order which weakens the double exchange interaction of electrons. By this way, the pressure induced a canted ferromagnetic state that leads to an electron localized ferromagnetic insulating behavior. A spin polarized tunneling magnetoresistance has been observed at $\mathrm{T_C}$ on the external field up to 5 T. Pressure reduces the MRR significantly due to the induced canted ferromagnetic spin state.

Acknowledgements

We gratefully acknowledge Prof. Elisa B. Saitovitch, for the help extended in this work. M. Kumaresavanji acknowledges the Conselho Nacional de Desenvolvimento Cientifico e Tecnologico (CNPq), Brazil and the Third World Academy of Sciences (TWAS), Italy for the award of CNPq-TWAS fellowship.

References

[1] T. Kimura and Y. Tokura 2000 *Annu. Rev. Mater. Sci.* **30** 451 .
[2] M. Kubota, H. Fujioka, K. Hirota, K. Ohoyama, Y. Moritomo, H. Yoshizawa and Y. Endoh 2000 *J. Phys. Soc. Jap* **69** 1606.
[3] J. F. Mitchell, C.D. Ling, J.E. Millburn, D.N. Argyriou, A. Berger, M. Medarde, D. Miller and Z.P. Luo 2002 *Appl. Phys. A* **74** S1776.
[4] Y. Moritomo, K. Hirota, H. Nakao, T. Kiyama, Y. Murakami, S. Okamoto, S. Ishihara, S. Maekawa, M. Kubota and H. Yoshizawa 2000 *Phys. Rev. B* **62** 17.
[5] K. V. Kamenev, G.J. McIntyre, Z. Arnold, J. Kamarad, M.R. Lees, G. Balakrishnan, E.M.L. Chung and D. McK 2001 *Paul, Phys. Rev. Lett.* **87** 167203.
[6] Ravhi S. Kumar, D. Prabhakaran, A. Boothroyd, M. F. Nicol and A. Cornelius 2006 *J. Phys. Chem. Solids* **67** 2046.
[7] K. V. Kamenev, M.R. Lees, G. Balakrishnan, D. McK.Paul, W.G. Marshall, V.G. Tissen and M.V. Nefedova 2000 *Phys. Rev. Lett.* **84** 2710.

[8] Aihwa Wang, G. Cao Y. Liu, Yi Long, Y. Li, Z. Feng and Joseph H. Ross 2005 *J. Appl. Phys.* **97** 103906.
[9] C. Zener 1951 *Phys. Rev.* **82** 403.
[10] A.J. Millis, P.B. Littlewood and B.I. Shraiman 1995 *Phys. Rev. Lett.* **74** 5144.
[11] J. S. Zhou, J.B. Goodenough and J.F. Mitchell 2000 *Phys. Rev. B* **61** R9217.
[12] D. S. Dessau and Z. X. Shen 1998 in *Colossal magnetoresistive oxides*, edited by Y. Tokura, (Gordon: Breach publishers).
[13] P.G. De Gennes 1960 *Phys. Rev.* **118** 141.
[14] Yu Lu, X.W. Li, G.Q. Gong, G. Xiao, A. Gupta, P. Lecoeur, J.Z. Sun, Y.Y. Wang and V.P. Dravid 1996 *Phys. Rev. B* **54** R8357.
[15] M. Julliere 1975 *Phys. Lett. A* **54** 225.