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Effect of pressure and magnetic field on bilayer $La_{1.25}Sr_{1.75}Mn_2O_7$ single crystal

S. Arumugam[a,*], K. Mydeen[a], Magda Fontes[b], N. Manivannan[a], M. Kumaresa Vanji[a], K.U. RamaTulasi[a], S.M. Ramos[c], Elisa Baggio Saitovitch[b], D. Prabhakaran[d], A.T. Boothroyd[d]

[a]*High Pressure Low Temperature Lab, School of Physics, Bharathidasan University, Tiruchirappalli-620 024, India*
[b]*Centro Brasileiro de Pesquisas Fisicas, Dr Xavier Sigaud 150, Urca, Rio de Janeiro, RJ 22290-180, Brazil*
[c]*Instituto de Fisica, Universidade Federal Fluminense, av. Litorânea sn, Boa Viagen, Niterói-RJ 24210-340, Brazil*
[d]*Department of Physics, Clarendon Laboratory, University of Oxford, Oxford OX1, UK*

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Abstract

We report the temperature dependence of susceptibility for various pressures, magnetic fields and constant magnetic field of 5 T with various pressures on $La_{2-2x}Sr_{1+2x}Mn_2O_7$ single crystal to understand the effectiveness of pressure and magnetic field in altering the magnetic properties. We find that the Curie temperature, T_c, increases under pressure ($dT_c/dP = 10.9$ K/GPa) and it indicates the enhancement of ferromagnetic phase under pressure up to 2 GPa. The magnetic field dependence of T_c is about 26 K for 3 T. The combined effect of pressure and constant magnetic field (5 T) shows $dT_c/dP = 11.3$ K/GPa and the peak structure is suppressed and broadened. The application of magnetic field of 5 T realizes 3D spin ordered state below T_c at atmospheric pressure. Both peak structure in χ_c and 3D spin ordered state are suppressed, and changes to 2D-like spin ordered state by increase of pressure. These results reveal that the pressure and the magnetic field are more competitive in altering the magnetic properties of bilayer manganite $La_{1.25}Sr_{1.75}Mn_2O_7$ single crystal.

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1. Introduction

Doped manganite exhibits colossal magnetoresistance (cmr), which was discovered in the cubic perovskite manganese oxides [1] and later extended to other manganites from the same family, containing one or more MnO_2 layers [2,3]. These groups of compounds are known as Ruddlesden-Popper series. The double layered ($n=2$) compounds $La_{2-2x}Sr_{1+2x}Mn_2O_7$ (LSMO) have received serious attention as the cmr effect is very much stronger than in the 3D perovskite ($n=\alpha$) compounds. In the LSMO perovskite in which MnO_2 bilayers and $(La_{2-x}Sr_x)O_2$ blocks are stacked along the *c*-axis direction [2–4]. It shows quasi-two dimensional structure, enhanced cmr effect, reduced ferromagnetic transition temperature, variety of magnetic structure as a function of doping and large anisotropy in resistivity between *ab*-plane and *c*-axis [2–16]. Hence, exotic electronic, magnetic and transport properties

* Corresponding author. Tel.: +91 431 2407057; fax: +91 431 2407045.
E-mail address: sarumugam1963@yahoo.com (S. Arumugam).

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are expected in these manganite materials. The physics behind this group of materials could not be explained by the double exchange theory alone, since one has to consider the complex interplay of spin, charge, structural and orbital degrees of freedom.

The long-range magnetic order in the doped manganites is due to the interplay between the double exchange and superexchange interaction, and the doping level or external pressure controls it. The pressure effect increases the overlap between cation and anion orbitals in the Mn–O network and the magnetic effect increases the alignment of magnetic moments on the manganese sites and it is expected to increase the Curie temperature (T_c) and the metal–insulator transition (T_{MI}) [17,18]. Also, the pressure is not expected to make any changes in the number of charge carriers and it does not introduce a preferred orientation of magnetic moments in the system. Recent pressure experiments on cmr materials show an interesting features like: (1) simultaneous increase of T_{MI} and decrease of resistivity (2) increase of T_c and Neel temperature (3) evolution of ferromagnetic (FM) phase (4) reduction in Jahn–Teller distortion of MnO_6 octahedra (5) decrease of magnetic anisotropy and coercive field (6) increase of spontaneous magnetization (7) increase of Mn–O–Mn bond angle and decrease of Mn–O bond length, etc. [1–18].

However, application of magnetic field is expected to do the parallel alignment of magnetic moments on the manganese sites, destruction of charge ordering and increase of T_c. The doping of divalent atoms in the La-site leads to the suppression of Jahn–Teller distortion, increase of T_c and the FM exchange interaction [19,20]. Hence, it is quite interesting to investigate the magnetic properties of cmr materials under the high pressure and the high magnetic field. Here, we report the temperature dependence of susceptibility for various pressures, magnetic fields and constant magnetic field of 5 T with various pressures on LSMO single crystal to understand the effectiveness of both pressure and magnetic field in altering the magnetic properties.

2. Experiment

The crystals of LSMO were grown from sintered rods of the same nominal composition by the travelling-solvent floating-zone method using mirror furnace and the details are reported elsewhere [21]. Laue X-ray diffraction and Electron-probe microstructure analysis (EPMA) were used for checking the quality of single crystals. The DC resistivity under atmospheric pressure was carried out on single crystal of size $0.75\times1\times1.2\ mm^3$ with silver paint as an electrode using four-probe method to estimate the metal–insulator transition. The temperature was measured using silicon diode sensor. The DC magnetization was measured by superconducting quantum interference device magnetometer (quantum design) and the measurements were taken in both zero field-cooled (ZFC) and field-cooled (FC) conditions under applied field of 0.1 G. The AC-susceptibility was measured in a Cu–Be hydrostatic pressure cell using fluorinate #77 as a pressure medium. The pressure was determined by calibrated manganin gauge at room temperature. The actual pressure at low temperature was calculated from the mid point of the superconducting transition temperature of Pb. The Pb was kept very close to the sample to avoid error in the measurement of pressure. The magnetic field was applied using Oxford-7 T superconducting magnet.

3. Results and discussion

The temperature dependence of the resistivity of LSMO single crystal at atmospheric pressure is present in Fig. 1. The T_{MI} where we observed very high resistivity, is 129 K. The inset shows the DC magnetization of LSMO125 single crystal under ambient pressure at 0.1 G with field-cooled (FC) and ZFC measurements using MPMS SQUID magnetometer. The Curie temperature (T_c) is defined as the mid point temperature of the transition curve, corresponding to an average transition temperature. We found that the T_c is coincides with the T_{MI} value (129 K).

Fig. 2 shows the normalized AC-susceptibility as a function of temperature, measured at various pressures. The inset shows the pressure dependence of T_c up to 2 GPa in the absence of magnetic field. A remarkable increase of T_c as a function of pressure was observed ($dT_c/dP = 10.9$ K/GPa), which indicates enhancement of FM phase. The applied pressure mainly weakens the interlayer coupling leads to realization of 2D FM state down to low temperature, and it

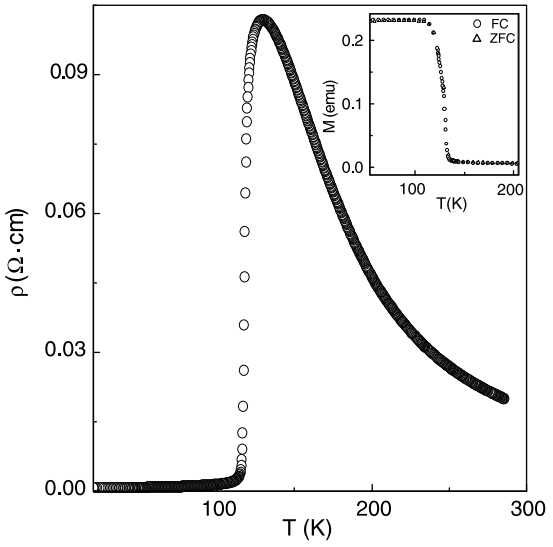

Fig. 1. The temperature dependence of the resistivity for LSMO125 single crystal under ambient pressure [inset: Field cooled magnetization and zero-field cooled magnetization at $H = 0.1$ G].

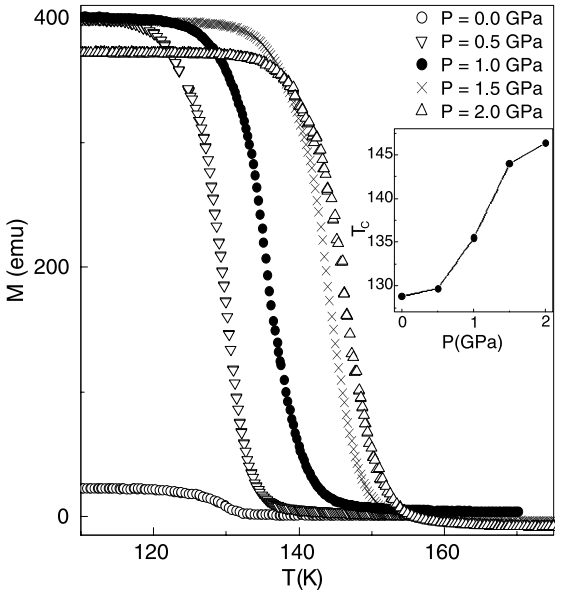

Fig. 2. Normalized AC-susceptibility as a function of temperature for LSMO125 single crystal under various pressures [inset: T_c of LSMO125 single crystal as a function of pressure].

drastically increases as a function of pressure. The observed positive pressure coefficient seems to be less than the value reported for $La_{0.79}Ca_{0.21}MnO_3$ ($dT_c/dP = 37$ K/GPa) and $La_{0.6}Ca_{0.4}MnO_3$ ($dT_c/dP = 16$ K/GPa). The present result reveals that pressure enhanced two-dimensional (2D) character in the charge transport in the bilayer manganites.

Application of high magnetic field is expected to increase the coupling between the bilayers and it leads to realization of 3D-spin ordered state at atmospheric pressure. Fig. 3 shows temperature dependence of DC magnetization of LSMO125 single crystal at various applied magnetic fields up to 3.0 T at atmospheric pressure and the inset shows the magnetic field dependence of T_c of the same crystal. We found that the T_c increases to 26 K for the magnetic field of 3 T without any pressure and the application of external magnetic field is increases the coupling between the bilayers, and induces the 3D FM spin arrangement irrespective of any pressure. The independent effects of hydrostatic pressure and the magnetic field on LSMO125 single crystal show the reasonable variation on T_c and revels that the pressure and the magnetic field are more competitive in altering magnetic properties of bilayer manganites.

We have also investigated the effect of AC-susceptibility of LSMO125 system with constant magnetic field of 5 T ($\chi_c \| c$-axis) under the various hydrostatic pressures and the results are plotted in Fig. 4. A sharp maximum of χ_c is observed at $T_c \sim 165$ K at atmospheric pressure with 5 T as a magnetic field and peak structure is an indication of realization 3D-spin arrangement. The peak structure is suppressed and broadened, and the T_c is shifted to higher temperature ($dT_c/dP = 11.3$ K/GPa) when the pressure increases to 2 GPa with constant magnetic field of 5 T. This trend shows that the crossover of charge transport from 3D to 2D and the evolution of 2D FM character increase when pressure increases. The combination of hydrostatic pressure of more than 2 GPa and the magnetic field of >5 T may be needed for completely suppressing the 3D character in the bilayer manganites. These results suggest that the pressure (magnetic) effect likely suppresses (promotes) the

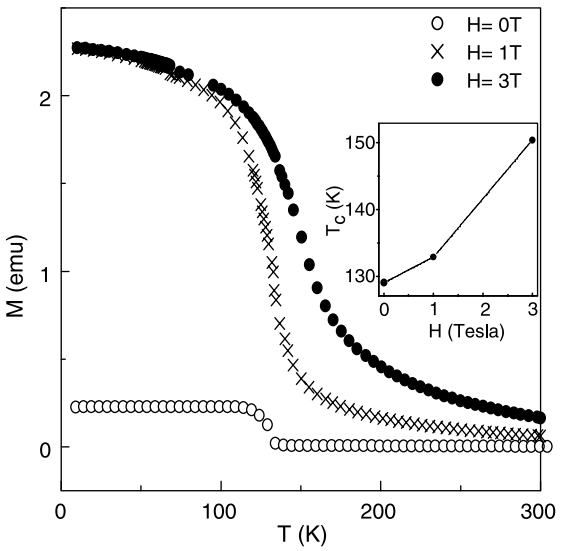

Fig. 3. DC magnetization as a function of temperature at various magnetic fields for LSMO125 single crystals [inset: T_c of LSMO125 single crystal as a function of magnetic field].

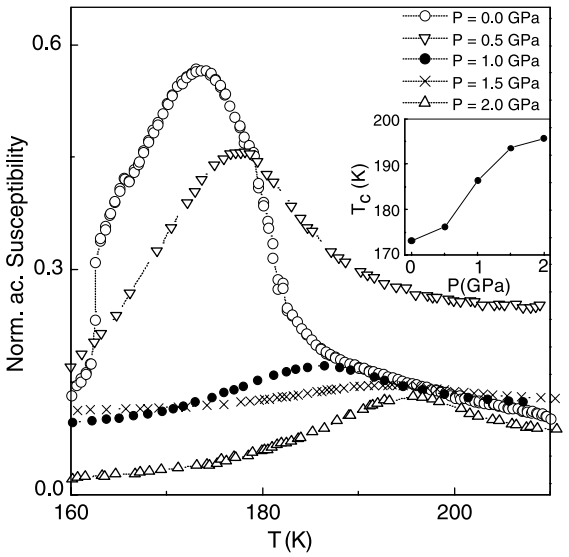

Fig. 4. The temperature dependence of normalized AC-susceptibility at different pressures with 5 T as a constant magnetic field [inset: T_c vs. pressure with constant magnetic field as a 5 T].

FM coupling between bilayers and promotes for 2D (3D) charge transport. The consequence of the combined effect of pressure and magnetic field is unpredictable for perovskite type bilayer manganites with anisotropic compressibility and accurate knowledge of spin structure must be obtained from neutron scattering measurements under high pressure and magnetic field for a complete understanding.

The results of the present study agree well with the results of effects of pressure and magnetic field on $La_{1.4}Sr_{1.5}Mn_2O_7$ (LSMO140) system as reported by Kimura et al. [16]. They have shown that the magnitude of the magnetic coupling between the adjacent bilayers weakened as the pressure increases, which makes the charge dynamics more 2D. Also, they reported that the application of magnetic field induced 2D–3D crossover with high ratio of tunneling magnetoresistance. However, T_N and the amplitude of peak susceptibility (χ_{cmax}) decrease with pressure and broadened at 1.1 GPa in LSMO140 system [16].

Also, our result is comparable with the results of pressure effects of $(La_{0.6}Nd_{0.4})_{1.2}Sr_{1.8}Mn_2O_7$ (LNSMO120) system in the low pressure region in which the amplitude of χ_{cmax} as well as Neel temperature (T_N) increases as a function of pressure ($dT_N/dP = 5$ K/GPa) up to 1.9 GPa and then χ_{cmax} decreases and T_N increases up to 7.1 GPa [22,23]. The structural analysis under high pressure of LNSMO140 bilayer manganites shows that Nd doping results into longer Mn–O(1) and shorter Mn–O(2) bond lengths, while Mn–O(3) bond length remains the same. Further increase in Nd content >0.4 would result in the interchange of Mn–O(1) and Mn–O(2) bond lengths, which is similar to the changes induced by the application of external pressure as reported by Kamenev et al. [22]. The T_N increases as amplitude of peak susceptibility χ_c decreases as the pressure increases >1.9 GPa, which is an indication of inter change of Mn–O(1) and Mn–O(2) bond length, i.e. Mn–O(2) bond length is longer than Mn–O(1) bond length at >1.9 GPa. Similar ratio of bond length is expected in LSMO125 system. Argyriou et al. [11] reported from the structural analysis of LSMO120 system with the increase of Mn moment and positive dT_c/dP in the FM state is consistent with the present results. Hence, the effect of pressure and the magnetic field variations observed in the present system is agreed in comparison with the results of reported in LNSMO120 and LSMO140 systems.

The outcome of our previous investigation of resistivity measurements under uniaxial pressure on the same single crystals gives supporting evidence for the enhancement of 2D character in the charge transport with pressure parallel to *ab*-plane, and enhancement of 3D character with pressure parallel to *c*-axis [24]. The results of uniaxial pressure parallel to *c*-axis reveal that the amplitude of peak resistivity decreases and the T_{MI} increases while measuring the resistivity parallel to *ab*-plane, which favors 3D-charge transport. The results of uniaxial pressure parallel to *ab*-plane reveal that the amplitude of peak resistivity increases and the T_{MI} decreases while measuring the resistivity parallel to *c*-axis, which favors 2D-charge transport [24].

Hence, we conclude that the pressure and magnetic field effects on LSMO125 single crystal show the enhancement of 2D and 3D-spin arrangement respectively. The 3D spin structure change over to 2D like spin structure by increasing the pressure at constant magnetic field of 5 T. Also, the pressure and the magnetic field are competitive in controlling the magnetic properties of bilayer manganites.

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References

[1] R.M. Kusters, J. Singleton, D.A. Keen, R. Mcgreevy, W. Hayes, Physica B 155 (1989) 362.
[2] Y. Morimoto, A. Asamitsu, H. Kuwahara, Y. Tokura, Nature 380 (1996) 141.
[3] R. Mahesh, R. Mahendran, A.K. Raychaudhuri, C.N.R. Rao, J. Solid State Chem. 122 (1996) 4481.
[4] M.D. Coey, M. Viret, S. von Molnar, Adv. Phys. 48 (1999) 167.
[5] T.G. Perring, G. Aeppli, T. Kimura, Y. Tokura, M.A. Adams, Phys. Rev. B 58 (1998) R14693.
[6] J.F. Mitchell, J.E. Millburn, M. Medarde, D.N. Argyriou, J.D. Jorgensen, J. Appl. Phys. 85 (1999) 4352.
[7] M. Imada, A. Fujimori, Y. Tokura, Rev. Mod. Phys. 70 (1998) 1039.
[8] U. Welp, A. Berger, V.K. Vlasko-Vlasov, Q. Li, K.E. Grey, J.F. Mitchell, Phys. Rev. B 62 (2000) 8615.
[9] K. Hirota, Y. Morimoto, H. Fujioka, M. Kubota, H. Yoshizawa, Y. Endo, J. Phys. Soc. Jpn. 67 (1998) 3380.
[10] D.N. Argyriou, J.F. Mitchell, P.G. Radaelli, H.N. Bordallo, D.E. Cox, M. Medarde, J.D. Jorgensen, Phys. Rev. B 59 (1999) 8695.
[11] D.N. Argyriou, J.F. Mitchell, J.B. Goodenough, O. Chmaissen, S. Short, J.D. Jorgensen, Phys. Rev. Lett. 78 (1997) 1568.
[12] T. Akimoto, Y. Morimoto, K. Ohoyama, S. Okamoto, S. Ishihara, S. Makekawa, A. Nakamura, Phys. Rev. B 59 (1999) R14153.
[13] Y.-H. Choi, E.-O. Chi, Y.-U. Kwon, H.C. Kim, H.-C. Ri, C.-H. Lee, J.-S. Lee, H.-S. Shim, Phys. Rev. B 63 (2001) 054437.
[14] D.B. Romero, Y. Morimoto, J.F. Mitchell, H.D. Drew, Phys. Rev. B 63 (2001) 132404.
[15] Y. Morimoto, K. Ohoyama, M. Ohashi, Phys. Rev. B 59 (1999) 157.
[16] T. Kimura, A. Asamitsu, Y. Tomioka, Y. Tokura, Phys. Rev. Lett. 79 (1997) 3720.
[17] H.Y. Hwang, T.T.M. Palstra, S.-W. Cheong, B. Batlog, Phys. Rev. B 52 (1995) 15046.

[18] J.J. Neumeier, M.F. Hundley, J.D. Thompson, R.H. Heffner, Phys. Rev. B 52 (1995) R7006.
[19] H.Y. Hwang, S.-W. Cheong, P.G. Radaelli, M. Marezio, B. Batlog, Phys. Rev. Lett. 75 (1995) 914.
[20] Y. Morimoto, H. Kuwahara, Y. Tokura, Physica B 237–238 (1997) 26.
[21] D. Prabhakaran, A.T. Boothroyd, J. Mater. Sci. Mater. Electron. 14 (2003) 587.
[22] K.V. Kamenev, M.R. Lees, G. Balakrishnan, C.D. Dewhurst, D.McK. Paul, Physica B 265 (1999) 191.
[23] K.V. Kamenev, M.R. Lees, G. Balakrishnan, D.McK. Paul, W.G. Marshall, V.G. Tissen, M.V. Nefedova, Phys. Rev. Lett. 84 (2000) 2710.
[24] K. Arumugam, M. Mydeen, M. Kumerasa, Vanji, D. Prabhakaran, A.T. Boothroyd, R.K. Sharma, J. Solid State Chem. (submitted).