Lattice coupled first order magnetoresistance transition in an A-type antiferromagnet: Pr$_{0.46}$Sr$_{0.54}$MnO$_3$

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Abstract

We investigated magnetic, magnetotransport and magnetostriction properties of the A-type antiferromagnet Pr$_{0.46}$Sr$_{0.54}$MnO$_3$ which undergoes a first order paramagnetic-antiferromagnetic transition below $T_N = 210$ K while cooling and $T_N = 215$ K while warming. The zero field ($\mu_0 H = 0$ T) resistivity shows a sudden jump at $T_N$ and a small bump around $T_{\text{max}} = 220$ K ($> T_N$). $T_N$ shifts down and $T_{\text{max}}$ shifts up with increasing $\mu_0 H$. Magnetoresistance as high as -45-57 % at 7 T is found in the temperature range 180 K-230 K. Isothermal measurements indicate that the field induced antiferromagnetic to ferromagnetic transition below $T_N$ is accompanied by a rapid decrease of the resistivity but increase of volume ($\Delta V/V = +0.25 \%$ at 180 K and 13.7 T). This lattice coupled magnetoresistance transition is suggested due to the field induced structural transition from the low volume orthorhombic to the high volume tetragonal structure.
Extensive studies following the discovery of a magnetic field induced destruction of the antiferromagnetic insulating phase in Pr$_{0.5}$Sr$_{0.5}$MnO$_3$ revealed that the low temperature insulating phase is actually a two dimensional ferromagnetic metal but with successive ferromagnetic planes coupled antiferromagnetically. This peculiar type of antiferromagnetism known as A-type antiferromagnetism is caused by two dimensional ordering of the Jahn-Teller split $e_g$ - $d_{x^2-y^2}$ orbitals of Mn$^{3+}$ ions. When heated from the low temperature, the interplanar antiferromagnetic coupling becomes weak and Pr$_{0.5}$Sr$_{0.5}$MnO$_3$ transforms into a ferromagnetic metal in the temperature range 145-270 K before changing into a paramagnetic insulator. There are indications that ferromagnetic domains from nano to micron size coexist with the antiferromagnetic phase even far below the Neel temperature in Pr$_{0.5}$Sr$_{0.5}$MnO$_3$ by NMR and magnetic studies. With increasing Sr content the strength of A-type antiferromagnetic interaction increases at the expense of the ferromagnetic interaction and finally A-type antiferromagnetism gives away to the C-type antiferromagnetism in Pr$_{0.3}$Sr$_{0.7}$MnO$_3$. The A-type antiferromagnetism is so far not observed for $x < 0.5$ to in any of the RE$_{1-x}$AE$_x$MnO$_3$ (RE is a trivalent rare earth ion and AE is a divalent alkaline earth ion) series. Although there are few electrical transport studies in the A-type antiferromagnetic compounds away $x = 0.5$, there is hardly any study on magnetotransport in the A-type antiferromagnetic compounds away from half doping. Kuwahara et al found that the out plane resistivity in Nd$_{0.45}$Sr$_{0.55}$MnO$_3$ is 4 orders of magnitude higher than the in plane value at 45 mK and the in plane resistivity at $\mu_0H = 10$ T and 45 mK is an order of magnitude lower than the out of plane resistivity. However, it is not known whether lattice distortion plays any role in the magnetoresistance of the A-type antiferromagnet. In order to investigate this aspect we have chosen Pr$_{0.40}$Sr$_{0.54}$MnO$_3$ which is an A-type antiferromagnet and shows a large magnetoresistance close to the Neel temperature. To clarify this aspect we carried out magnetostriction measurement on Pr$_{1-x}$Sr$_x$MnO$_3$ ($x = 0.54, 0.55$). We also investigated Pr$_{0.45}$Sr$_{0.55}$MnO$_3$ but a detailed study was discarded because a magnetic field more for $\mu_0H = 14$ T which is beyond our experimental limit was needed to induce an antiferromagnetic to ferromagnetic transition.
We measured four probe resistivity of the polycrystalline $Pr_{0.46}Sr_{0.54}MnO_3$ over a wide temperature (10 K- 320 K) and field (0 T $\leq H \leq$ 7 T) range using a Quantum Design Physical Properties Measuring System. Magnetic measurements were carried out using a Quantum Design SQUID magnetometer. In the ac susceptibility measurement the amplitude of ac signal was 3 Oe and the frequency was 100 Hz. Magnetostriiction measurements using the strain gauge method in a pulsed field up to 13.7 T was measured at University of Zaragoza, Spain. We measured both parallel ($\lambda_{\text{par}}$) and perpendicular ($\lambda_{\text{per}}$) magnetostriictions with field parallel and perpendicular to the measuring directions respectively. The volume magnetostriction of randomly oriented polycrystallites is determined through the relation $\omega = \lambda_{\text{par}} + 2\lambda_{\text{per}}$

Fig. 1 (a) shows the real ($\chi'$) and imaginary ($\chi''$) parts of the ac susceptibility while warming from 10 K. Both $\chi'$ and $\chi''$ exhibit a peak around $T_N = 215$ K due to A-type antiferromagnetic to paramagnetic transition. The paramagnetic to antiferromagnetic transition while cooling occurs at $T_N = 210$ K but the data is not shown for clarity. The antiferro-paramagnetic transition is accompanied by orthorhombic (Fmmm) to tetragonal (I4/mcm) structural transition as revealed by neutron diffraction study.\(^9\) The inverse susceptibility is shown on the right scale. The low temperature anomaly in the inverse susceptibility ($1/\chi'$) around 50 K is possibly related to ordering of the Pr moments and above $T_N$, $1/\chi'$obeys the Curie-Weiss law with a positive Weiss temperature $\theta_p = 230$ K and an effective moment of $P_{\text{eff}} = 7.1837 \mu_B$. The experimentally observed value of $P_{\text{eff}}$ is higher than the theoretically calculated value of $P_{\text{eff}} = \sqrt{0.46P_{\text{Pr}^{3+}}^2 + 0.46P_{\text{Mn}^{3+}}^2 + 0.54P_{\text{Mn}^{4+}}^2} = 5.004 \mu_B$ where $P_{\text{Pr}^{3+}}^2 = 12.816$, $P_{\text{Mn}^{3+}}^2 = 24$, $P_{\text{Mn}^{4+}}^2 = 15$. Judging from the earlier magnetization and resistivity data in $Pr_{1-x}Sr_xMnO_3$ series\(^7\) a short range ferromagnetic ordering with $T_C \approx 220$ K is not an unlikely possibility in $x = 0.54$. The closeness of $T_C$ to $T_N$ makes it difficult to distinguish in $x = 0.54$ from the susceptibility data alone. A strong ferromagnetic correlations exist much above the $T_C$ as suggested by the high effective magnetic moment value. When the strength of the external magnetic field increases, ferromagnetic correlations becomes much
stronger and $T_C$ shifts up in temperature. This is reflected in Fig. 1(b) which shows the inverse dc susceptibility ($H/M$) while field cooling for different $H$ values over 320K-160 K. The curves for different $H$ do not match in the paramagnetic region over a wide temperature range (200 K- 300 K). The short range ferromagnetic ordering is also reflected in the $M(H)$ isotherms in Fig. 1(c) at $T = 215$ K, 220 K, and 225 K. As $T$ is reduced below 210 K, antiferromagnetic correlations overcome ferromagnetic contributions. At 210 K, $M(H)$ increases initially increases linearly with the field but then raises rapidly around $\mu_0 H_c = 1.2$ T. The rapid increase of $M(H)$ signals antiferromagnetic to ferromagnetic transition. The transition is first order as suggested by the hysteresis behavior. As $T$ decreases further $\mu_0 H_c$ increases to rapidly to 4.0 T at 200 K.

Fig. 2(a) shows temperature dependence of the resistivity $\rho(T)$ for $H = 0$ T and 7 T. $\rho(T)$ increases gradually as $T$ is lowered below 320 K and exhibits a bump around $T_{max} = 220$ K as shown in the inset. Then, $\rho(T)$ jumps up around $T_N = 210$ K and then continues to increase. The value of $\rho(T)$ at 4.2 K is very small ($\approx 30.0$ mΩ cm) which is slightly larger than the in plane resistivity at 4.2 K ($\approx 10$ mΩ cm) of the single crystal A-type antiferromagnet Nd$_{0.45}$Sr$_{0.55}$MnO$_3$. The weak bump at $T_{max}$ is caused by the short range ferromagnetic order. The $T_{max}$ is shifted up to more than 300 K and $T_N$ is shifted down to 184 K under $\mu_0 H = 7$ T. Fig. 2(b) shows $\rho(T)$ curves while cooling and warming under a field for different values of $H$ over a limited temperature range. The $\rho(0T)$ curve exhibits hysteresis of 5 K width during the paramagnetic (tetragonal) to antiferromagnetic (orthorhombic) while cooling and the reverse transition upon warming. We take the middle point of the hysteresis region as the Neel temperature for convenience. When the field is increased $T_N$ systematically shifts down and $T_{max}$ shifts up as shown in Fig. 3. The widening of difference between $T_C$ and $T_N$ is reflected in the metallic like resistivity behavior in between $T_C$ and $T_N$. The applied magnetic field strongly reduces the resistivity in between $T_C$ and $T_N$.

Fig. 4 (a) shows the field dependence of the magnetoresistance $MR = [\rho(H)-\rho(0)]/\rho(0)$ at selected temperatures. The data were taken after zero field cooling to a given temperature
from \( T = 300 \) K. At 215 K, the magnitude of MR increases rapidly for \( H < 1 \) T and then gradually for higher fields. The data taken at 220 K, 225 K and 230 K (not shown here for clarity) are similar to the 215 K. The field dependence of these data are similar to the MR behavior of the spin dependent tunnelling between the ferromagnetic grains in polycrystalline manganites below the ferromagnetic Curie temperature. When \( T \) is decreased below 210 K, MR gradually decreases with \( H \) initially and then rapidly around \( \mu_0 H_C^+ \) before levelling off at high fields. At 205 K, \( \mu_0 H_C^+ = 2.2 \) T during field increasing part which closely correlates with \( \mu_0 H_C^+ \) found in the M(H) data in Fig. 1(c). In the A-type antiferromagnetic state, metallic conduction is two dimensional in absence of magnetic field. When the interplanar antiferromagnetic coupling is destroyed by a strong magnetic field, the charge transport changes into three dimensional in character which results in a rapid increase in the value of the MR. The field induced antiferromagnetic to ferromagnetic transition is first order with a constant hysteresis width of 1.5 T. The maximum magnetoresistance at 7 T is -50-55 % which is comparable to other manganites.

Is the MR due to changes in the spin sector alone? To understand this aspect, we measured magnetostriction isotherms which are shown in Fig. 4(b). We find a giant volume expansion with hysteresis in the temperature range 205 K-170 K. This behavior is opposite the volume contraction under a magnetic field found in most of the manganites investigated earlier. The volume change is rather small when the sample is in the AF state but shows a sudden jump during the field induced antiferromagnetic to ferromagnetic transition. The critical field \( \mu_0 H_C^+ = 2.25 \) T at 205 K is in close agreement with the MR and M(H) data. After the completion of the AF-FM transition \( \Delta V/V \) shows a gradual variation with further increase in H.is gradual. At 210 K (i.e., \( T \approx T_N \)) \( \Delta V/V \) is small and it attains the maximum value of 0.25 % at 180 K and then decreases to 0.16 % at 170 K. This is because the maximum available field of 13.7 T is insufficient to complete the AF-FM transition. What is the origin of the observed volume expansion under the magnetic field? The A-type AF order is coupled to the \( d_{x^2-y^2} \) orbital ordering. Orbital ordering creates quadrupole moments which
can interact with the lattice through electron phonon coupling. The orbital ordering is also destroyed together with the AF ordering for $H >> \mu_0 H_C$. This triggers a field induced structural transition from the orthorhombic to possibly a tetragonal structure. This possibility is more likely given the fact the antiferromagnetic phase has lower volume than the paramagnetic phase as revealed by the volume thermal expansion data shown in Fig. 5. The antiferromagnetic phase has orthorhombic structure and the paramagnetic phase has tetragonal structure. The volume thermal expansion decreases by $\approx 0.18 \%$ in between 220 K and 200 K as the temperature is decreased and the structural transition shows hysteresis. If there were no magnetic and structural transition, $\Delta V/V$ would have followed the Grüneissan behavior shown as a fit to the high temperature data. The volume expansion under the magnetic field has contributions one from volume change due to magnetic field induced structural transition and another from volume change due to spontaneous magnetostriction of the field induced ferromagnetic phase. At 190 K the experimentally observed volume is 0.21 % lower than the Grüneissan value whereas the volume magnetostriction is 0.25 %. This small difference is due the contribution from spontaneous magnetostriction of the ferromagnetic phase. Thus, our study indicates that the magnetotransport in the A-type AF $Pr_{0.46}Sr_{0.54}MnO_3$ is strongly coupled to the structural transition under the field. It will be interesting to know whether a large first order magnetoresistance behavior can be ever observed in a manganite without a change in lattice distortion or structural transition.

In conclusion, we have shown the A-type AF $Pr_{0.46}Sr_{0.54}MnO_3$ exhibits a first order magnetoresistance behavior below the Neel temperature which involves not only spin and charge but also the lattice. We found that the field induced antiferromagnetic to ferromagnetic transition is accompanied by a giant volume expansion which is suggested to the field induced structural transition from the low volume orthorhombic to the high volume tetragonal structure. First order spin-charge-lattice coupled transition is also of great interest to study giant magnetocaloric effect and future research on our compound need to be focussed on this aspect.
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Figure captions

Fig. 1: Left scale; Temperature dependence of the real ($\chi'$) and imaginary ($\chi''$) parts of the ac susceptibility of Pr$_{0.46}$Sr$_{0.54}$MnO$_3$. Right scale $1/\chi'$ versus temperature. The Curie-Weiss fit is shown by the thick line. (b). The inverse dc magnetic susceptibility for different values of H in a limited temperature range while field cooling from 320 K (c). Isothermal magnetization data.

Fig. 2: Temperature dependence of the resistivity $\rho(T)$ at $\mu_0H = 0$ T and 7 T over 320 K-10 K range while field cooling. Inset shows the bump around 220 K ($>T_N$) in a enlarged scale. (b) $\rho(T)$ for different $\mu_0H$ values in a limited temperature range (160 K-320 K).

Fig. 3: Field dependence of the Neel temperature ($T_N$) and the temperaure $T_{max}$ corresponding to the bump in the resistivity above $T_N$.

Fig. 4: Field dependence of the isothermal (a) magnetoresistance and (b) volume magnetostriction.

Fig. 5: Volume thermal expansion of Pr$_{0.46}$Sr$_{0.54}$MnO$_3$. The structural determination is from the neutron diffraction study (Ref. 9) The hysteresis region is marked by the vertical dashed lines.
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