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Role of double exchange interaction on the magnetic and electrical properties of $Pr_{0.8}Sr_{0.2}MnO_3$ ferromagnetic insulating manganite

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In this work the ferromagnetic state in a ferromagnetic insulating manganite viz. $Pr_{0.8}Sr_{0.2}MnO_3$ was analyzed. The temperature variation of ac susceptibility is seen to show two transitions with a high temperature transition occurring at 158 K and a low temperature cusp (T_f) around 90 K. The high temperature transition (T_C) was found to be independent of frequency while the low temperature transition shifts with frequency indicating a frustrated magnetic state. The relaxation time observed from the critical slowing down spin analysis indicates that this state is that of a cluster glass. Temperature variation of resistivity shows an insulating behavior with two distinct humps corresponding to T_C and T_f. Temperature variation of thermopower exhibits a peak at T_C and decreases below T_C which is attributed to the sudden release of holes from traps, similar to a ferromagnetic metallic manganite. Magnetoresistance is seen to show a change in curvature upon cooling the sample across T_C which in conjunction with the peak in resistivity and thermopower suggests that this transition is mediated by a double exchange mechanism. The frustrated state is seen to arise because of a competition between double exchange and superexchange mechanisms. The effect of double exchange mechanism is seen to persist all the way down to 5 K as seen from the magnetoresistance analysis. These results clearly suggest that double exchange still plays an important role in dictating the magnetic and electrical properties in ferromagnetic insulating manganites. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2170590]

I. INTRODUCTION

In the low-doped state, manganites exhibit a ferromagnetic insulating ground state which cannot be completely explained by the Zener's double exchange (DE) mechanism.[1] Other interactions such as Jahn–Teller interaction, and superexchange interactions, are stronger in this doping regime and they compete with the double exchange mechanism resulting in a complex magnetic state in these systems. Various theories have been proposed to explain the origin of this ferromagnetic phase.[2–5] However, the exact role of the double exchange in determining the transport and magnetic properties in these low doped phases is still under debate. In this work we have analyzed the ferromagnetic state in $Pr_{0.8}Sr_{0.2}MnO_3$ using ac susceptibility, resistivity, magnetotransport, and thermopower measurements to study the extent of the influence of DE in this compound.

II. EXPERIMENT

A polycrystalline $Pr_{0.8}Sr_{0.2}MnO_3$ sample was prepared using the conventional solid-state method. The phase purity was checked using x-ray diffraction (Rich-Seifert, Germany). The temperature variation of dc magnetization was done using a superconducting quantum interference device magnetometer (MPMS XL, Quantum Design, USA) in the field-cooled mode. ac susceptibility measurements were performed as a function of temperature at three different frequencies using a commercial susceptometer (Sumitomo Corpn, Japan). Temperature and field variation of resistivity were measured using an Oxford flow cryostat from 350 to 5 K and the temperature variation of thermopower was measured using a closed cycle refrigerator.

III. RESULTS AND DISCUSSION

$Pr_{0.8}Sr_{0.2}MnO_3$ (PSMO2) sample was found to be single phase with an orthorhombic structure and the diffractogram is shown in Fig. 1(a). Temperature variation of magnetization (field cooled) at an applied field of 1000 Oe [Fig. 1(b)] shows a T_C of 158 K [obtained from differentiating the $M(T)$ curve with respect to temperature] while the real and imaginary parts of ac susceptibility [Fig. 2(a)] show two transitions, one at 158 K and a low temperature cusp around 90 K (T_f). To probe the presence of the two-transition behavior, we have performed ac susceptibility at different ac frequencies (313, 1000, and 10 000 Hz). ac susceptibility at different frequencies [Fig. 2(b)] shows that, while the high tempera-

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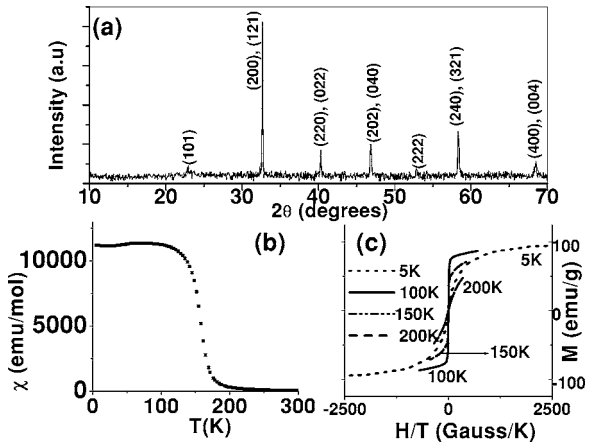

FIG. 1. (a) X-ray diffractogram for the sample which could be indexed to an orthorhombic structure with lattice constants, $a=5.47$ Å, $b=7.72$ Å, and $c=5.48$ Å. (b) Temperature variation of dc susceptibility in field cooled (FC) mode in an applied field of 1000 Oe. (c) M vs H/T for various temperatures above and below T_f.

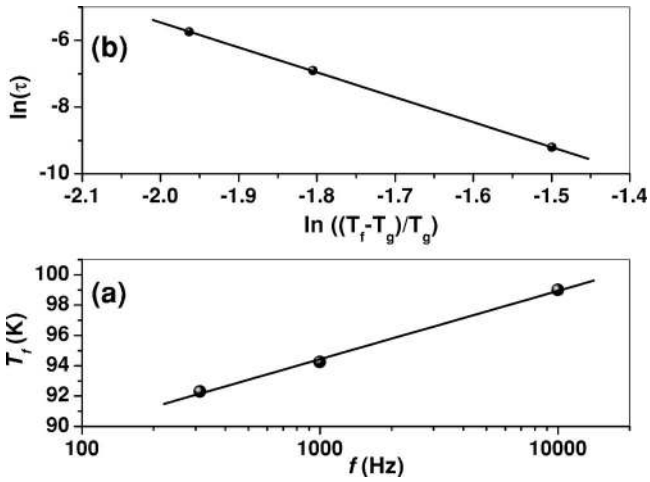

FIG. 3. (a) Variation of the T_f, the cusp temperature as a function of frequency. The symbols represent the data points while the line is a linear fit whose intercept gives the glass transition temperature T_g at zero frequency. (b) $\ln(\tau)$ vs $\ln[(T_f-T_g)/T_g]$. The symbols represent the data points while the line is a linear fit to the critical slowing down behavior.

ture transition is independent of frequency, the low temperature cusp shifts to higher temperatures with increase in frequency. The fact that this high temperature transition is seen at an applied magnetic field of 1000 Oe and is independent of frequency implies that it is a robust transition corresponding to the conventional paramagnetic to ferromagnetic transition. The shift in cusp temperature with frequency usually arises from either a superparamagnetic behavior or spin/cluster glass like behavior. M versus H/T curves do not overlap at all above T_f [Fig. 1(c)] as expected for a superparamagnet[6] indicating the absence of superparamagnetic behavior and suggests that the origin of the cusp could be due to a spin/cluster glass type state. The cusp temperature as a function of the applied frequency [Fig. 3(a)] yields a straight line which when interpolated to zero frequency gives T_g, the glass transition temperature at zero frequency. According to the critical slowing down behavior of the conventional spin glass model,[7] $\tau/\tau_0=[(T_f-T_g)/T_g)]^{-z\nu}$, where T_g is the cluster/spin glass transition temperature, T_f is the frequency dependent freezing temperature at which the maximum relaxation occurs, time τ corresponds to the measured frequency, τ_0 is the characteristic time constant, and $z\nu$ is the critical exponent. The fit to this model [Fig. 3(b)] yields a value of $\tau_0=1.4\times10^{-9}$ s and $z\nu=7.5$. The value of τ_0 suggests that this state is that of a cluster glass since in conventional spin glasses it is usually seen to be around 10^{-13} s.[7] Thus ac susceptibility measurement clearly suggests that the low temperature transition is to that of a magnetically frustrated state while the high temperature transition is from a paramagnetic to ferromagnetic state. To further study the influence of these two transitions on the transport (electrical and charge) we have performed temperature and field variation of electrical resistivity and temperature variation of thermopower.

Temperature variation of electrical resistivity in 0 and 8 T magnetic fields for the PSMO2 sample is shown in Fig. 4. Temperature variation of resistivity shows an insulating behavior throughout the measured temperature range (5–350 K) with two distinct humps seen near T_C and T_f. Below T_f resistivity rises sharply, indicating that the carriers are more localized in that state. It is seen that the sample

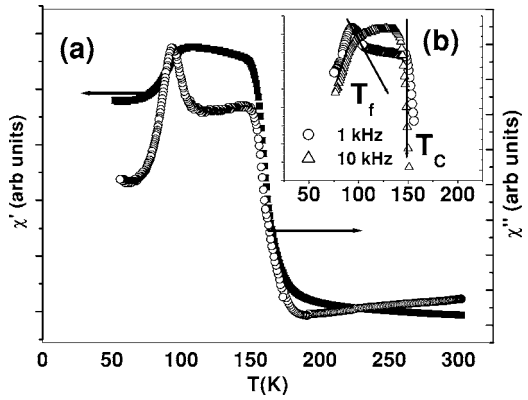

FIG. 2. (a) Temperature variation of real (χ') and imaginary (χ'') part of ac magnetic susceptibility in an applied frequency (f) of 313 Hz. (b) Temperature variation of the imaginary part of susceptibility at different frequencies (1 and 10 kHz). The cusp is seen to shift to higher temperatures with increase in frequency. Cusp observed for $f=313$ Hz is not shown for the sake of clarity.

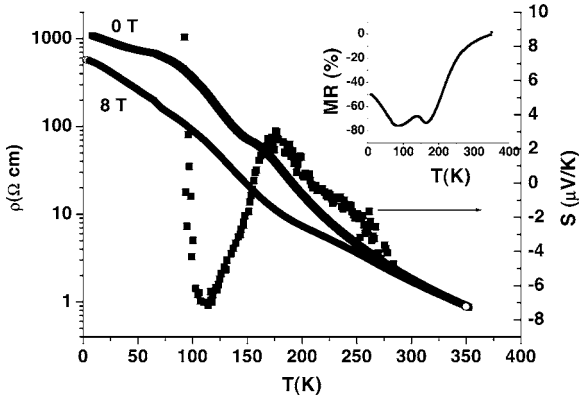

FIG. 4. Temperature variation of electrical resistivity in an applied field of 0 and 8 T and temperature variation of thermopower. (Inset) Temperature variation of magnetoresistance in an applied magnetic field of 8 T.

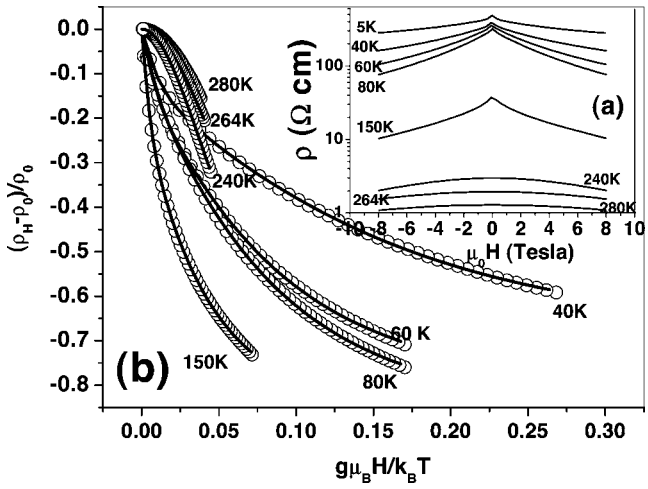

FIG. 5. (a) Field variation of resistivity at different temperatures above and below T_C. (b) Curve fits to the Brillouin and square of Brillouin function below and above T_C, respectively. Symbols represent data points while line is the curve fit to the model as reported in Ref. 10.

remains insulating even in a magnetic field of 8 T implying that the insulating state is very robust. The observed insulating behavior can be attributed to lack of carriers to bring about the metallicity in the compound since this composition ($x=0.2$) lies in the ferromagnetic insulating phase of the PSMO system.[8] Temperature variation of thermoelectric power S (T) is also shown in Fig. 4. Thermoelectric power is found to exhibit a peak as the sample is cooled toward T_C from above and then decreases sharply at T_C. This sharp fall in S (T) is attributed to the delocalization of carriers which condense from a random distribution of small polarons into a state that eliminates polaronic distinguishability between Mn^{3+} and Mn^{4+} for transitions that are mediated by double exchange.[9] Below T_f, thermopower like resistivity is also seen to increase rapidly. This is probably due to the onset of the magnetically frustrated state leading to carrier localization. Thus, both thermopower and resistivity measurements indicate that carriers get localized below the second transition (T_f). This clearly suggests that a competing mechanism to the double exchange is present and may arise from the superexchange interactions between the Mn^{3+}–O–Mn^{3+} linkages leading to the observed frustrated state. The field variation of resistivity at temperatures above and below T_C is shown in Fig. 5(a). It is seen that there is a change in curvature of the plots as the sample is cooled across T_C similar to what is observed in ferromagnetic metallic manganites near T_C. This, along with the thermopower peak points to the presence of itinerant carriers near T_C, suggesting that this transition is mediated by the double exchange mechanism. The field variation of resistivity was analyzed using the model proposed by Wagner *et al.*[10] where they assume that the activation energy of the hopping barrier is modified depending on whether it is a paramagnetic (hopping barrier increases) or a ferromagnetic (hopping barrier decreases) state. This leads the MR to scale as a Brillouin function in the ferromagnetic state and as a square of the Brillouin function in the paramagnetic state. This model was seen to fit the magnetoresistance (MR) just like in a ferromagnetic metallic manganite without any additional constraints [Fig. 5(b)] even below T_f down to 5 K. This implies that itinerant carriers are present even at 5 K suggesting that the effect of double exchange mechanism is present even at 5 K. These results point out that the double exchange interactions mediate the high temperature paramagnetic to ferromagnetic transition and in the ferromagnetic state below T_C, it competes with other interactions such as Jahn–Teller and superexchange resulting in a frustrated state. However, the effect of this interaction is seen to persist even up to low temperature leading to a finite magnetoresistance. These results are in agreement with the electronic phase separation theory in manganites where it is predicted from Monte Carlo simulations that the ferromagnetic state consists of coexisting ferromagnetic and antiferromagnetic clusters and this coexistence leads to the origin of the colossal magnetoresistance property of the manganites.[1]

We have thus shown that even for low doped ferromagnetic insulating manganites, double exchange interaction plays a vital role in determining the magnetic and electrical, magneto-, and charge transport properties.

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