



# Magnetic and electric transport properties of polycrystalline manganite $\text{Pr}_{0.6}\text{Na}_{0.4}\text{MnO}_3$

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## ABSTRACT

The magnetic and electric transport properties of the polycrystalline manganite  $\text{Pr}_{0.6}\text{Na}_{0.4}\text{MnO}_3$  were reported. In zero field, the sample shows an insulator behavior. Under applied fields higher than 10 kOe, a field-induced insulator–metal transition occurs, and at the same time the colossal magnetoresistance (CMR) effect is also observed. At the temperature of 5 K, the metamagnetic transition from charge ordering/antiferromagnetic (CO/AFM) to ferromagnetic (FM) state is observed. With increasing temperature, the metastable FM phases can transform back to the CO state, accompanied by a high field hysteresis. The CMR effect of the sample can be understood by a percolative regime and is related to the CO–FM transition under a large magnetic field.

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## 1. Introduction

The hole-doped manganites  $\text{R}_{1-x}\text{A}_x\text{MnO}_3$  ( $\text{R}$  = rare-earth ions,  $\text{A}$  = divalent ions), whose resistivity can change on the order of  $10^4$ – $10^6$  by applying external magnetic field  $H$  of a few tesla, continue attracting the attention of condensed matter physicists [1–3]. This colossal magnetoresistance (CMR) effect can be qualitatively understood in the framework of the double exchange (DE) interaction model [4]. When the rare-earth site is doped with a divalent ion, a proportional number of  $\text{Mn}^{3+}$  ions are converted into  $\text{Mn}^{4+}$  ions and mobile  $e_g$  electrons are introduced, mediating the ferromagnetic (FM) interaction between  $\text{Mn}^{3+}$  and  $\text{Mn}^{4+}$  according to the DE interaction. In these manganite systems, the hopping of  $e_g$  electrons between two partially filled d orbitals of neighboring  $\text{Mn}^{3+}$  and  $\text{Mn}^{4+}$  ions via the orbital overlap  $e_g$ – $\text{O}2p_\sigma$ – $e_g$ , and the strong Hund coupling between the  $t_{2g}$  core spins and the mobile  $e_g$  electrons' spins cause the FM interaction between  $\text{Mn}^{3+}$  and  $\text{Mn}^{4+}$ .

However, further studies suggest that the lattice distortion due to the Jahn–Teller effect [5,6] can also be crucial for explaining the magnetic and transport mechanisms in mixed-valence

manganites. Furthermore, a phase-separation (PS) scenario, which assumes the coexistence at different length scales of ferromagnetic metallic (FMM) and antiferromagnetic (AFM) charge and orbital ordered insulating phases, has recently been proposed to explain the CMR effect [2–7]. The competition between the interactions and/or orders inherent in manganites, such as between DE ferromagnetism and superexchange antiferromagnetism and between charge–orbital ordering (CO–OO) state and metallic state, will produce the multicritical state [8]. It has been accepted that, given a temperature  $T$  and magnetic field  $H$ , the electronic and magnetic ground state of manganites can be inhomogeneous due to the coexistence of the FMM phase and the CO/AFM insulating phase. The coexisting two phases originate from the electronic phase separation [2,7,9].

Among this class of compounds, Pr-doped manganites  $\text{Pr}_{1-x}\text{A}_x\text{MnO}_3$  ( $\text{A}$  =  $\text{Ca}^{2+}$ ,  $\text{Sr}^{2+}$ , etc.) have been the subject of several studies due to their diverse physical and structural properties with doping concentration at different temperatures [10–15]. For instance, the 30% Ca-doped sample undergoes a charge-ordering (CO) transition near 220 K ( $T_{\text{CO}}$ ), an AFM transition at about 150 K ( $T_{\text{N}}$ ), and a FM transition around 100 K ( $T_{\text{C}}$ ). Below the AFM transition temperature (TFM), interestingly, application of a high magnetic field leads to FM and metallic states. Although the nature of the induced FM and metallic phases is not yet completely understood, it has been suggested that the induced

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state might be due to the development of FM clusters in the CO and AFM matrix after the application of a magnetic field.

Presently, most studies focused on divalent alkaline earth-metal doping in  $R_{1-x}A_x\text{MnO}_3$  compounds. The studies on monovalent metal-doped samples have not been exhaustively reported [16–18]. In fact, the substitution of monovalent metal ions (such as  $\text{K}^+$ ,  $\text{Na}^+$ , etc.) for the rare-earth ions can also lead to the change of the valence of Mn ions, hence the DE interaction. In this paper, we present the investigation of the magnetic and electrical transport properties of a polycrystalline manganite sample  $\text{Pr}_{0.6}\text{Na}_{0.4}\text{MnO}_3$ .

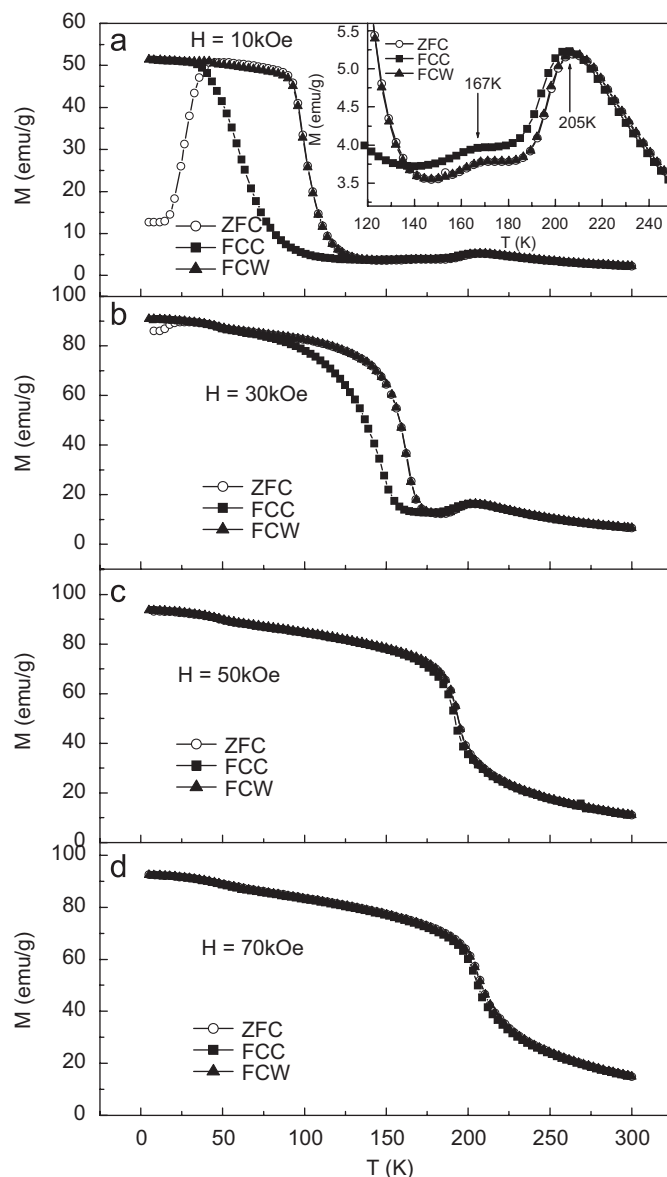
## 2. Experimental procedure

The sample was prepared by means of the conventional solid-state reaction method. Stoichiometric amounts of  $\text{Pr}_6\text{O}_{11}$ ,  $\text{Na}_2\text{CO}_3$ , and  $\text{MnCO}_3$  powders were thoroughly mixed, ground and then calcined for 24 h at 900 °C. The powder thus obtained was then reground, pressed into pellet, and sintered in the air at 1100 °C for 48 h. The structure of the samples was characterized by X-ray diffraction at room temperature using a D-max Rigaku system with  $\text{CuK}_\alpha$  radiation. The sample is of single phase and the diffraction spectra can be indexed based on an orthorhombic perovskite structure with the Pnma space group. The corresponding lattice parameters at room temperature, determined from Rietveld refinements, are  $a = 5.4523 \text{ \AA}$ ,  $b = 5.4556 \text{ \AA}$ , and  $c = 7.7021 \text{ \AA}$ .

The magnetic measurements were carried out with a quantum design superconducting quantum interference device magnetic property measurement system. The electric resistivities were measured by a standard four-probe method with a quantum design physical property measurement system.

## 3. Results and discussion

Fig. 1 shows the temperature dependence of the magnetization of the sample, measuring in a field of 10, 30, 50, and 70 kOe using different experimental procedures: zero-field cooled (ZFC) field-cooled cooling (FCC), and field-cooled warming (FCW). For the field of 10 kOe, all ZFC, FCC, and FCW curves exhibit a hump around 205 K, which indicates the forming of a CO state [19]. At 167 K, a small but discernible kink is also observed, which indicates the onset of the AFM ordering. For clarity, the enlarged curves are shown in the inset of Fig. 1(a). At lower temperature, all ZFC, FCC, and FCW curves exhibit a FM transition, characterized by a fast increase of the magnetization with decreasing temperature. The TFM, which is defined as the peak temperature of  $dM/dT$ , is  $\sim 102 \text{ K}$  for the FCC curve and  $\sim 64 \text{ K}$  for the ZFC and FCW curves. A thermal hysteresis is found in the FCC and FCW cycle, characterizing a first-order transition. It suggests the FM and CO/AFM phases coexist in the sample and the FM develops in the CO/AFM matrix. Below about 41 K, the ZFC and FCW curves separate from each other. The magnetization after ZFC has a smaller value than that after field cooling (FC). It suggests that, under FC, the applied field favors the formation of FM phase at the expense of the CO/AFM one; hence, a larger fraction of FM phase compared with that under ZFC. Furthermore, it should be noticed that, below about 13 K,  $M_{\text{ZFC}}$  is nearly independent of temperature. Above this temperature,  $M_{\text{ZFC}}$  increases rapidly with increasing temperature. It suggests that the sample is blocked in a metastable PS state after ZFC due to the quenched disorder [20,21] or the strains between the FM and CO/AFM states [22]. Increasing the temperature, the system becomes unblocked due to the thermal fluctuation, and the applied field promotes a growth of FM phase



**Fig. 1.** Temperature dependence of the magnetization of  $\text{Pr}_{0.6}\text{Na}_{0.4}\text{MnO}_3$ , measuring in a field of 10 kOe (a), 30 kOe (b), 50 kOe (c), and 70 kOe (d), using different experimental procedures: zero-field cooled (ZFC), field-cooled cooling (FCC), and field-cooled warming (FCW). Inset of Fig. 1(a) shows the enlargement of the  $M(T)$  curves under 10 kOe.

over the CO/AFM one, leading to the rapid increasing of the magnetization.

As shown in Fig. 1, the difference between ZFC and FCW curves becomes smaller with increasing field and the ZFC curve merges with the FCW one when the applied field is 70 kOe. Furthermore, the hysteresis also becomes smaller, and the FM transition shifts to higher temperature with increasing field. It suggests that, after ZFC, applying higher field will make more CO/AFM component transformed to FM phases. When the applied field is as high as 70 kOe, the sample is almost full FM. Therefore, no difference is observed among the ZFC, FCC, and FCW curves of 70 kOe.

Fig. 2 shows the temperature dependence of resistivity  $\rho(T)$  of  $\text{Pr}_{0.6}\text{Na}_{0.4}\text{MnO}_3$  in various fields (from 0 to 130 kOe). The resistivity was measured while the sample was cooled from 300 K in a fixed field. In zero field, the resistivity increases monotonically with the decreasing temperature by more than five orders of magnitude from 300 K to the lowest measuring temperature  $T_{\text{min}}$ , exhibiting an insulating behavior. Below  $T_{\text{min}}$ ,

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