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Magnetic and magneotransport properties of $Nd_{0.75}Na_{0.25}MnO_3$ manganite

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ABSTRACT

We report the magnetic and magnetotransport properties of the polycrystalline manganite $Nd_{0.75}Na_{0.25}MnO_3$. In zero field, the sample shows a ferromagnetic insulator behavior. Under applied fields higher than 30 kOe, a field-induced insulator-metal, followed by a metal-insulator transition at a lower temperature occur; 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 phase 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.

1. Introduction

The hole-doped manganites $R_{1-x}A_xMnO_3$ (R = rare-earth ions, 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 Mn^{3+} ions are converted into Mn^{4+} ions and mobile e_g electrons are introduced, mediating the ferromagnetic (FM) interaction between Mn^{3+} and 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 Mn^{3+} and Mn^{4+} ions via the orbital overlap e_g–O2p_σ–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 Mn^{3+} and Mn^{4+}.

However, further studies suggest that the lattice distortion due to the Jahn–Teller (JT) 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 that between double exchange 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 FMM phase and CO/AFM insulating phase. The coexisting two phases originate from the electronic phase separation [2,7,9].

Among this class of compounds, Nd-based hole-doped manganites $Nd_{1-x}A_xMnO_3$ are interesting in that the strength of the double exchange interactions is weaker than that of the La-based compounds, due to large lattice distortions provoked by the smaller Nd ions [10]. For example, $Nd_{1-x}Ca_xMnO_3$ with $0 \leq x \leq 0.25$ shows a FM semiconducting behavior, but that with $x > 0.80$ exhibits an AFM semiconducting ground state at low temperature. Furthermore, as $0.30 \leq x \leq 0.80$, $Nd_{1-x}Ca_xMnO_3$ shows a rich variety of phenomena [10,11], for example the manganite $Nd_{0.5}Ca_{0.5}MnO_3$ undergoes a charge ordering transition at $T_{CO} = 250$ K and the AFM ordering transition at $T_N = 160$ K.

Presently, most studies have focused on manganites in which the rare earth site is doped with divalent ions. However, the studies haven't been exhaustively reported on manganites in which the rare earth site is doped with monovalent ions [12–14]. In fact, the substitution of monovalent ions (such as K^+, Na^+ etc.) for rare-earth ions can also lead to the change of the valence of Mn ions, hence the DE interaction. In this paper, we investigate the magnetic and magnetotransport properties of a polycrystalline manganite sample $Nd_{0.75}Na_{0.25}MnO_3$.

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2. Experimental procedure

The sample was prepared by means of the conventional solid-state reaction method. Stoichiometric amounts of Nd_2O_3, Na_2CO_3, and $MnCO_3$ 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 air at 1200 °C for 24 h. The structure of the sample was characterized by X-ray diffraction (XRD) at room temperature using a D-max Rigaku system with Cu Kα 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 $a = 5.4416$ Å, $b = 7.6667$ Å, and $b = 5.4255$ Å.

The magnetic measurements were carried out with a Quantum Design superconducting quantum interference device (SQUID) magnetic property measurement system (MPMS). The resistivities of the sample were measured by a standard four-probe method with a Quantum Design physical property measurement system (PPMS).

3. Results and discussion

Fig. 1 shows the temperature dependence of the magnetization of the sample measured with applied field of $H = 100$ Oe, 10 kOe, and 70 kOe using different experimental procedures: zero-field cooled (ZFC), field-cooled cooling (FCC), and field-cooled warming (FCW). For $H = 100$ Oe, all ZFC, FCC, and FCW curves exhibit a FM transition, characterized by a fast increase of the magnetization with decreasing temperature. The Curie temperature T_c, defined as the peak temperature of dM/dT, is ~110 K. The inset of Fig. 1(a) shows the inverse susceptibility versus temperature for $Nd_{0.75}Na_{0.25}MnO_3$, calculated by the ZFC curve. It is found that the Curie–Weiss law for the experimental curve is not satisfactory throughout the whole temperature range above T_c. The curve deviates from the linearity below ~170 K and above ~240 K. The former is attributed to the onset of a short range FM correlation [2,15]. As to the latter, such an anomaly is usually found in half doped manganites at the CO transition [16–18]. Consequently, we identify this anomaly as a signal of this ordering taking place at $T_{CO} \sim 240$ K.

Although $M_{FCC}(T)$ coincides with $M_{FCW}(T)$ below T_c, there is a significant difference between $M_{FCW}(T)$ and $M_{ZFC}(T)$ in $H = 100$ Oe. For the phase separated manganite systems, two mechanisms have been proposed to interpret the difference between $M_{FCW}(T)$ and $M_{ZFC}(T)$. One is the cluster-glass behavior. The coexistence of FM and CO/AFM phases in manganites implies the frustration of magnetic interactions, which is attributed to either the cluster interaction within the phase separated state [19–21] or the competition between the double exchange and superexchange interactions [22,23]. For zero field cooling magnetization $M_{ZFC}(T)$, below T_c, the moments of the FM clusters are arranged randomly and frozen at low temperature, forming a spin-glass state. The appearance of the spin-glass state induces the decrease of the magnetization at a lower temperature. Cooling the sample in a magnetic field, the moments of clusters will arrange more ordering and cause the magnetization larger than that without magnetic field, which results in the difference between $M_{FCW}(T)$ and $M_{ZFC}(T)$.

The other mechanism is ascribed to the different relative volume fraction of the FM and CO/AFM phases after zero field cooling and field cooling. Under ZFC, the sample is blocked in a metastable state due to the quenched disorder [24,25] or the strains between the FM and CO/AFM states [26]. With increasing temperature, the applied field unblocks the system, promoting a growth of FM phase over the CO/AFM one and leading to the increase of the magnetization. Under field cooling, the field favors the formation of FM phase at the expense of the CO/AFM one, hence, a larger fraction of FM phase occurs compared with that under ZFC.

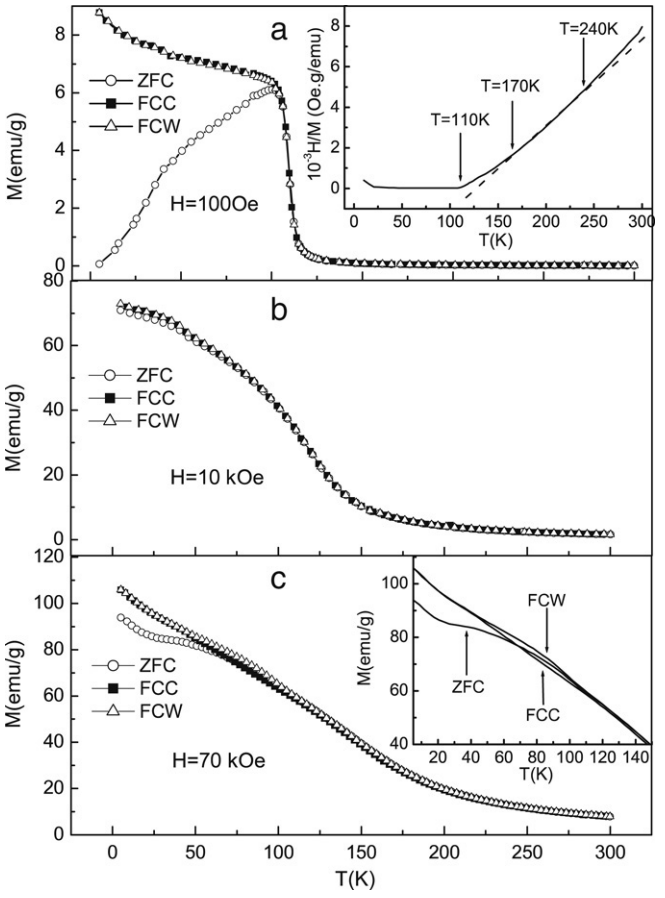

Fig. 1. The temperature dependence of the magnetization of $Nd_{0.75}Na_{0.25}MnO_3$, measure with $H = 100$ Oe (a), 10 kOe (b) and 70 kOe (c). Inset of Fig. 1(a) shows the inverse susceptibility versus temperature of the sample, calculated by the ZFC curve of 100 Oe. Inset of Fig. 1(c) shows the enlargement of the $M(T)$ curves under 70 kOe.

Since the field of 100 Oe is rather small, it should not change the relative fractions of the FM and CO/AFM too much. Nevertheless for our sample, $M_{FCW}(T)$ is considerably different from $M_{ZFC}(T)$ in 100 Oe. This difference, therefore, probably originates from a cluster-glass phase. In a larger field of 10 kOe, $M_{FCW}(T)$ is almost the same as $M_{ZFC}(T)$, as shown in Fig. 1(b), which suggests the cluster-glass state is melted in such a higher field. However, in $H = 70$ kOe, as shown in Fig. 1(c), the difference between $M_{FCW}(T)$ and $M_{ZFC}(T)$ does not reduce, or rather, $M_{FCW}(T)$ is significantly larger than $M_{ZFC}(T)$ at low temperature. To clarify this behavior, the enlargement of the curves is shown in the inset of Fig. 1(c). It suggests that, in a high field such as 70 kOe, the different relative fractions of FM and CO/AFM phases account for the difference between $M_{FCW}(T)$ and $M_{ZFC}(T)$. Furthermore, 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, which further supports our conjecture.

Fig. 2 shows the temperature dependence of resistivity $\rho(T)$ of $Nd_{0.75}Na_{0.25}MnO_3$ in various fields (from 0 to 130 kOe). The resistivity was measured when the sample was cooled from 300 K in a fixed field. Its zero-field resistivity increases monotonically with decreasing temperature by more than six orders of magnitude from 300 K to the lowest measuring temperature T_{min}, exhibiting