



Complex ferromagnetic state transformation in single crystal $\text{Nd}_{0.5}\text{Pb}_{0.5}\text{MnO}_3$

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ABSTRACT

The magnetic properties of $\text{Nd}_{0.5}\text{Pb}_{0.5}\text{MnO}_3$ single crystal have been studied by dc magnetization and magnetotransport data. The ferromagnetic (FM) to paramagnetic (PM) transition temperature T_C and the metal–insulator transition temperature are 177 and 194 K, respectively. The deviation of the inverse susceptibility from the Curie–Weiss law indicates the presence of a Griffiths-like phase in this system, viz., an inverse susceptibility characterized by $\chi^{-1} \propto (T - T_C^{\text{rand}})^{1-\lambda}$ with $\lambda = 0.81$ and $T_C^{\text{rand}} = 183$ K close to the T_C 177 K. The slopes of H/M vs. M^2 isotherms are negative between 300 and 194 K and positive above 300 K and below 194 K. The transition cannot be regarded as first or second order on the basis of the Banerjee criterion. The above interesting and complex magnetic transition is a manifestation of competition between the double exchange mechanism and correlations arising from coupled spin and lattice degrees of freedom present in studied system. To analyze the magnetism around T_C , the critical behavior of $\text{Nd}_{0.5}\text{Pb}_{0.5}\text{MnO}_3$ is investigated based on the data of static magnetization measurements. It is seen that the critical behavior of $\text{Nd}_{0.5}\text{Pb}_{0.5}\text{MnO}_3$ belongs to the mean-field model and shows a second-order like magnetic transition at high fields, however, the poor scaling is found toward low fields, which suggest that the critical behavior is related to the observed complicated magnetic transition. The possible origin of observed complex FM state has been discussed based on the competition between the charge/orbital order clusters and the A site disorder.

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1. Introduction

Doped transition-metal oxides with a perovskite structure have been extensively studied over the past several years, in which many of them display a colossal magnetoresistance (CMR)—viz., a very large change in resistance induced by a applied magnetic field [1–3]. Such behavior is associated with the occurrence of a metal–insulator (MI) transition, the temperature of which exhibits a marked dependence on magnetic fields. The close relation between transportation and magnetism in these materials has been explained by the double exchange (DE) mechanism, where the e_g electrons of Mn^{3+} hop between neighboring sites via the $2p$ orbital of O^{2-} [4], phase separation combined with percolation [5] and the occurrence of a Griffiths phase [5,6] among others. However, the physical mechanism of the CMR effect remains controversial. In more general terms, manganese perovskites represent an interesting class of materials with an unusual combination of coupling between charge, spin, orbital, and various degrees of freedom, making them an important class of strongly correlated electronic systems.

The mixed-valent manganese oxides are characterized by a general formula $\text{A}_{1-x}\text{B}_x\text{MnO}_3$ where A is a rare-earth ion and B is a divalent ion with x representing the doping level. Changes in x modulate the valence of the Mn ions to maintain charge neutrality. The accompanying mismatch in the ionic size of ions occupying the A site leads to a distortion in the crystal structure frequently characterized by the Goldschmidt “tolerance factor” t_f [3–5]. The t_f can be described by

$$\frac{\langle r_A + r_O \rangle}{\sqrt{2}} (\langle r_{\text{Mn}} \rangle + r_O),$$

where r_O is the radius of the oxygen ion, while $\langle r_A \rangle$ and $\langle r_{\text{Mn}} \rangle$ are the average radii of A and Mn ions, respectively. When t_f is close to 1, a cubic perovskite structure is realized; as t_f decreases, the lattice structure firstly transforms to rhombohedral ($0.96 < t < 1$) structure and then to orthorhombic ($t < 0.96$) one [3,7]. Both the T_f and the variance of A site ionic radii ($\sigma^2 = \sum_i x_i r_i^2 - \langle r_A \rangle^2$, where x_i and r_i are the atomic fraction and ionic radii of i -type ions at A site, respectively), provide quantitative measures of the disorder in manganites [3–5]. As we know, the $\text{Ln}_{0.5}\text{Re}_{0.5}\text{MnO}_3$ ($\text{Ln} = \text{La}, \text{Pr}$ and Sm , etc. and $\text{Re} = \text{Ca}, \text{Sr}$ and Ba) manganites also show the charge/orbital ordered (CO/OO) [8], however, the CO/OO dose not present for $\text{Re} = \text{Pb}$ manganites, which activates us to investigate the magnetic properties of these materials in details. In this paper, the nature of the ferromagnetic (FM) to

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paramagnetic (PM) transition in $\text{Nd}_{0.5}\text{Pb}_{0.5}\text{MnO}_3$ single crystal is further investigated by analyzing the dc magnetization data and the critical behavior near T_C at low and high fields.

2. Experiment

Single crystals of $\text{Nd}_{1-x}\text{Pb}_x\text{MnO}_3$ were grown by the flux-melt technique using the PbO-PbF_2 (1:1.15) flux [9–12]. Growth precursors Nd_2O_3 , PbO , MnO_2 , and PbF_2 were weighted in appropriate ratio and homogenized in an agate pestle. The homogenized mixture was transformed into a platinum crucible, and the growth was carried out in a resistive furnace. The furnace was first heated to the 1050°C and held at this temperature for 24 h to ensure the sufficient dissolution of the raw materials, and then cooled to 800°C at a rate of 1°C/h . After above steps the furnace was cooled rapidly to room temperature to avoid possible twinning. The single crystals can be separated mechanically for investigation. The structure and phase purity of the samples were examined by X-ray diffraction (XRD) using $\text{Cu } K\alpha$ radiation at room temperature. The compositions of the samples were examined by energy dispersive spectra (EDS) technique. The difference between the experimental and nominal content is about 5%. $\text{Nd}_{0.5}\text{Pb}_{0.5}\text{MnO}_3$ was chosen to for our investigation among these crystals. Extensive magnetization measurements $M(T, H)$ were performed on a well characterized $\text{Nd}_{0.5}\text{Pb}_{0.5}\text{MnO}_3$ single crystals in external static magnetic fields H up to 48 kOe in the temperature range $158\text{ K} \leq T \leq 320\text{ K}$ with a Quantum Design superconducting quantum interference device (SQUID) system ($1.8\text{ K} \leq T \leq 400\text{ K}$, $0\text{ T} \leq H \leq 5\text{ T}$). The resistivity was carried out in a commercial Quantum Design Physical Property Measurement System (PPMS, $1.8\text{ K} \leq T \leq 400\text{ K}$, $0\text{ T} \leq H \leq 9\text{ T}$) from 5 to 380 K. The magnetization data $M(T, H)$ below 196 K was used to analyze the critical behavior of the $\text{Nd}_{1-x}\text{Pb}_x\text{MnO}_3$ single crystal. The effect internal field used for the scaling analysis has been corrected for demagnetization, $H_{\text{eff}} = H_{\text{appl}} - D_a M$, where D_a is the demagnetization factor obtained from M vs. H measurements in the low-field linear-response regime at 2 K.

3. Result and discussion

The temperature dependence of magnetization $M(T)$ and resistivity $\rho(T)$ of $\text{Nd}_{0.5}\text{Pb}_{0.5}\text{MnO}_3$ single crystal are shown in Fig. 1. The data are collected in a warming process with a 100 Oe applied magnetic field using zero-field-cooling (ZFC) and field-cooling (FC) modes, respectively. It can be obtained from Fig. 1(a) that the transition of FM–PM temperature T_C is 177 K defined by the temperature corresponding to the peak of dM/dT vs. T curve. Fig. 1(b) shows the magnetization obtained in ZFC mode and the resistivities measured in zero applied magnetic field. The metal–insulator T_{MI} is about 194 K, which corresponds to the peak in $\rho(T)$ curve. The T_{MI} is slightly larger than T_C . Fig. 2(a) shows the temperature dependence of the inverse ZFC magnetic susceptibility $\chi^{-1}(T)$. It is well known that the relation between $\chi^{-1}(T)$ and T of PM materials should follow the Curie–Weiss (CW) law, i.e., $\chi(T) = C/(T - \Theta)$, where C and Θ are the Curie constant and Weiss temperature. The solid line in Fig. 2(a) is the calculated curve deduced from the CW equation. It can be seen that the experimental curve can not be described by the CW law in the whole PM region. The slope of line alters at the temperature T_S (about 300 K). In the middle temperature range ($300\text{ K} < T < 194\text{ K}$), the inverse susceptibility is near linear and abruptly down around 194 K. The Curie constant and the Weiss temperature are $2.75\text{ K cm}^3/\text{mol}$ and 70.33 K , respectively. The effective magnetic moment μ_{eff} obtained from the Curie constant

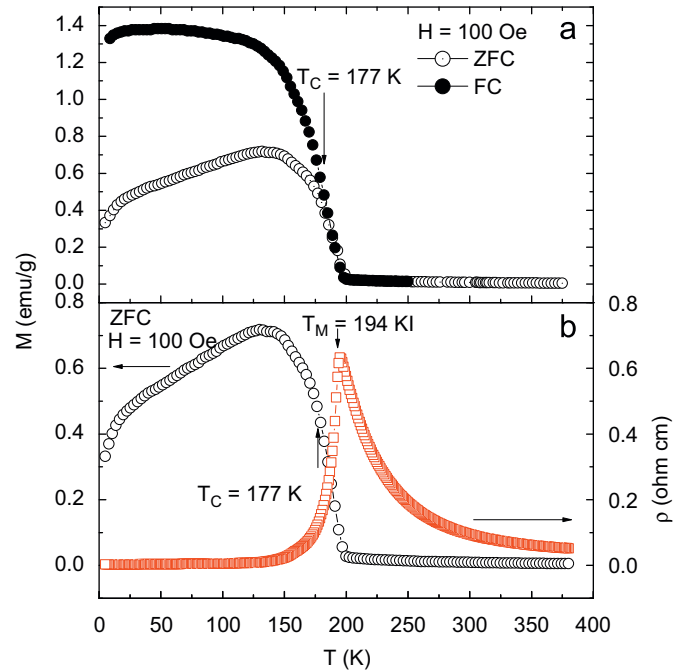


Fig. 1. (a) ZFC and FC magnetization measured as a function of temperature under a magnetic field $H = 100$ Oe and (b) temperature dependence of magnetization and resistivity measured at an applied field of 100 and 0 Oe, respectively, T_{MI} denotes the metal–insulator transition temperature.

is $4.69\mu_B/\text{f.u.}$. The theoretical effective PM moment per formula unit of $\text{Nd}_{0.5}\text{Pb}_{0.5}\text{MnO}_3$ can be obtained $5.10\mu_B/\text{f.u.}$ ($\mu_{\text{eff}}^{\text{th}} = \sqrt{0.5[\mu_{\text{eff}}^{\text{th}}(\text{Mn}^{3+})]^2 + 0.5[\mu_{\text{eff}}^{\text{th}}(\text{Mn}^{4+})]^2 + 0.5[\mu_{\text{eff}}^{\text{th}}(\text{Nd}^{3+})]^2}$). It is seen that the experimentally obtained μ_{eff} is slightly small than the theoretical value in the high temperature, which might be related to the charge/orbital ordered (COO) clusters present in the high temperature [23]. However, the local correlation between the COO clusters represent in the middle temperature might be responsible for change in the slope of inverse susceptibility at 300 K. The more detailed discussions will be given below.

In order to get a further insight into the kind of abnormal behavior of inverse susceptibility of $\text{Nd}_{0.5}\text{Pb}_{0.5}\text{MnO}_3$ in the middle temperature range, Fig. 2(b) shows the temperature dependence of the inverse ZFC magnetization susceptibility measured at $H = 10\text{ Oe}$ applied magnetic field from 150 to 220 K. The $\chi^{-1}(T)$ curve exhibits a Griffiths-like phase downturn below a certain temperature 194 K, which is close to the T_{MI} . The onset of this downturn is denoted as T_C (i.e. the temperature where $\chi^{-1}(T)$ deviates from the Curie–Weiss behavior) below which the FM clusters emerge in the PM matrix, as is described in a Griffiths phase (GP) system [13]. As we know, GP have been reported in doped Mn perovskites and other systems based on a variety of physical measurements and reflects the influence of disorder on the phase complexity in the manganites and related systems [13–15]. The GP is univocally characterized by a magnetic susceptibility exponent lower than unity, i.e.

$$\chi^{-1}(T) \propto (T - T_C^{\text{Rand}})^{1-\lambda}, \quad 0 \leq \lambda \leq 1 \quad (1)$$

where T_C^{Rand} is a fitting parameter [13–15]. The inset of Fig. 2(b) confirms the power law relation (Eq. (1)) and the fitting parameters are obtained: $\lambda = 0.81$ and $T_C^{\text{Rand}} = 183\text{ K}$. The estimated exponent λ lies in the expected range $0 \leq \lambda \leq 1$ and the value of T_C^{Rand} is close to the T_C . The evolution of this singularity with the applied magnetic fields is also investigated, the $\chi^{-1}(T)$ curves under different magnetic fields are shown in Fig. 3. It is seen that the shape of the curves do not give birth to considerable

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