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Comparative study of heterogeneous magnetic state above T_C in $La_{0.82}Sr_{0.18}CoO_3$ cobaltite and $La_{0.83}Sr_{0.17}MnO_3$ manganite

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ABSTRACT

The magnetic, transport and structural properties are studied for $La_{0.83}Sr_{0.17}MnO_3$ and $La_{0.82}Sr_{0.18}CoO_3$ single crystals with nearly the same doping and the metallic ground state. Their comparisons have shown that ferromagnetic clusters originate in the paramagnetic matrix below $T^* > T_C$ in both samples and exhibit similar properties. This suggests the possible universality of such phenomena in doped mixed-valence oxides of transition metals with the perovskite-type structure. The cluster density increases on cooling and plays an important role on the physical properties of these systems. The differences in cluster evolutions and scenarios of their insulator–metal transitions are related to different magnetic behaviors of the matrixes in these crystals that is mainly due to distinct spin states of the Mn^{3+} and Co^{3+} ions.

1. Introduction

The occurrence of nanoscale phase separation (PS) in hole doped manganites has been well established and is very important in determining their magnetic and transport properties [1]. However, the mechanisms of PS and properties of the phase-separated state remain the subject of much discussion. Recently formation of an inhomogeneous magnetic state at some T^* above the Curie temperature, T_C, caused by origination of the ferromagnetic (F) metallic (M) clusters in the paramagnetic matrix was revealed in the doped by different ways $La_{1-x}Ca_x MnO_3$, $Nd_{1-x}Ba_xMnO_3$ (NBMO) and $La_{0.88}MnO_x$ manganites with metallic [2–5] as well as insulating (I) [6,7] ground states. The properties of this state in different compounds appear to be quite similar that suggests a possible universality of this feature in manganites, exhibiting the F ordering. Study of this state in the doped cobaltites is the topic of the current research activity, which has been mainly investigated in $La_{1-x}Sr_xCoO_3$ (LSCO) [8]. The parent compound $LaCoO_3$ is a nonmagnetic insulator in the ground state since the Co^{3+} ion exhibits the low spin ($t_{2g}^6 e_g^0$, $S=0$) state. A finite spin state can be thermally induced or stabilized by the doping. Upon hole doping, magnetic Co^{4+} ions appear and the double exchange interaction between Co^{3+} and Co^{4+} ions, as it is generally accepted, leads to formation of the FM clusters. At low doping, the nanoscopic ($\sim$3 nm) FM droplets originate in a non-F semiconducting matrix, forming a magneto-electronic PS state [8]. In LSCO as doping increases, these clusters coalesce at $x \approx 0.18$, leading to a percolative type I–M transition at T_{IM}; the onset of long-range F order occurs at $T_C \geq T_{IM}$ [9,10]. The properties of the PS state were well studied at low T. The clustered regime above T_C occurring at $x \geq 0.18$ remains poorly understood. It is well known that LSCO reveals basic properties that are similar to those of the hole doped manganites: magnetic and insulator–metal (I–M) transitions and magnetoresistivity (MR). It is interesting to compare the features of clustered state above T_C in doped cobaltites and manganites. We examine these regimes in the single crystals LSCO ($x=0.18$) and $La_{0.83}Sr_{0.17}MnO_3$ (LSMO) with close levels of doping and metallic ground state. We found a similarity in cluster behavior above T_C in both systems in contrast to the conclusion of paper [8]. Considerable difference appears on approaching T_C, when the matrix comes into play in manganite. In $x=0.18$ LSCO, cooling leads to fast increase of the cluster density that is accompanied by their coalescence and percolation near I–M transition $T_C \approx T_{IM}$. In manganite the matrix crucially affects the behavior of clusters near T_C and leads to formation of the state that is close to a magnetically homogeneous one. The studies include the data on the transport and magnetic properties (the linear and nonlinear (second and third order) susceptibilities) for both samples as well as an analysis of the crystal structure (neutron diffraction) and small angle polarized neutron scattering (SAPNS) for the powdered cobaltite.

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2. Experimental details

The LSCO polycrystalline sample was fabricated by the standard solid-state reaction method. The single crystals were grown by the floating-zone method and were tested by described earlier ways, displaying a mosaicity of less than 1.5° and a single phase state [11]. The diffraction experiments were performed on the superpositional powder neutron diffractometer of PNPI at 2.3 K and 300 K. The resistance was measured by the standard four-probe method. The installation and measurements of the *ac* susceptibility were described in Ref. [6]. Measurements of the second harmonic of the longitudinal magnetization, M_2, in parallel *dc*- and *ac*-harmonic magnetic fields $H(t)=H+h\ \sin\ \omega t$ ($h \leq 35$ Oe, $\omega/2\pi=15.7$ MHz) allow one to determine reliably T^* and control T-evolution of the clustered state since this method is extremely sensitive to the appearance of a F fraction in a sample. It was demonstrated by our studies of this regime in the $Nd_{1-x}Ba_xMnO_3$ and $La_{0.88}MnO_x$ manganites [5–7]. The Re $M_2(H)$ and Im $M_2(H)$ parts of the M_2 were simultaneously recorded as functions of a steady magnetic field H for various sample temperatures (95–315 K). This field was scanned symmetrically relative to the point $H=0$ for detecting a field (H-) hysteresis of the signal with $M_2(H=0) \neq 0$. The amplitude of the H-scan was 300 Oe. According to the symmetry properties of M_2 the H-hysteresis appearance suggests the formation of a spontaneous ferromagnetic (F) moment in a sample. The polarization measurements were performed using a small-angle polarized neutron scattering (SAPNS) home made setup "Vector" with a wavelength $\lambda=9.2$ Å, $\Delta\lambda/\lambda=0.25$ (WWR-M reactor Gatchina).

3. Results and discussion

3.1. $La_{0.83}Sr_{0.17}MnO_3$

The $x \geq 0.15$ LaSrMnO series reveals the rhombohedral *R-3c* symmetry in high temperature region [12]. With decreasing T a transition is found to orthorhombic *Pbnm* structure at $T_S \approx 375$ K for $x=0.15$ that is accompanied by appearance of a small kink in resistivity. The doping increase leads to decrease of this temperature down to $T_S \sim 200$ K for $x=0.175$. Our sample exhibits such a transition at $T_S \approx 272$ K and shows isolator like behavior down to $T_{IM} \approx 252$ K where I–M transition occurs (insert 1 of Fig. 1a). Temperature dependence of the *ac* susceptibility χ ($\omega/2\pi=95$ kHz) and the amplitude of the third harmonic of magnetization $|M_3|$ ($\omega/2\pi=20$ kHz) are displayed in Fig. 1(a) and inset (2) of this Figure. According to $\chi'(T)$, T_C is near to 260 K. T_C can be determined more precisely from the $|M_3(T)|$ data as a position of the $|M_3(T)|$ maximum at the largest T [6]. It gives $T_C \approx 263\ \mathrm{K} > T_{IM}$ and this is accompanied by a kink in $R(T)$ (inset 1 of Fig. 1a.) The appearance of the F clusters causes a deviation of the $1/4\pi\chi'(T)$ curve down from the high T Curie–Weiss law [2] that occurs at $T^*_\chi \approx 280$ K (not shown).

Let us consider the data on the second harmonic of magnetization displayed for LSMO in Fig. 2. Unlike the NBMO series, in this compound the signal of the F clusters is clearly seen in the Re $M_2(H)$ component at high temperature $T^*=313$ K, whereas Im $M_2(H)$ has a not far from linear H-dependence due to a contribution of the matrix with a weakly expressed signal from the clusters near $H=0$ (Fig. 2, 313 K). On cooling from T^* down to $T^\# \approx 289$ K, the cluster response reveals an irregular character and relatively slowly increases (see panels 312.8 K, 289 K in Fig. 2). It becomes to dominate in the Re $M_2(H)$ part and is reliably detected in the Im $M_2(H)$ at $T^\#$. This is the same hysteretic response with a characteristic extremum in a weak $H \approx 25$ Oe (inset of Fig. 1b) as was found in other manganites and where it was attributed to the

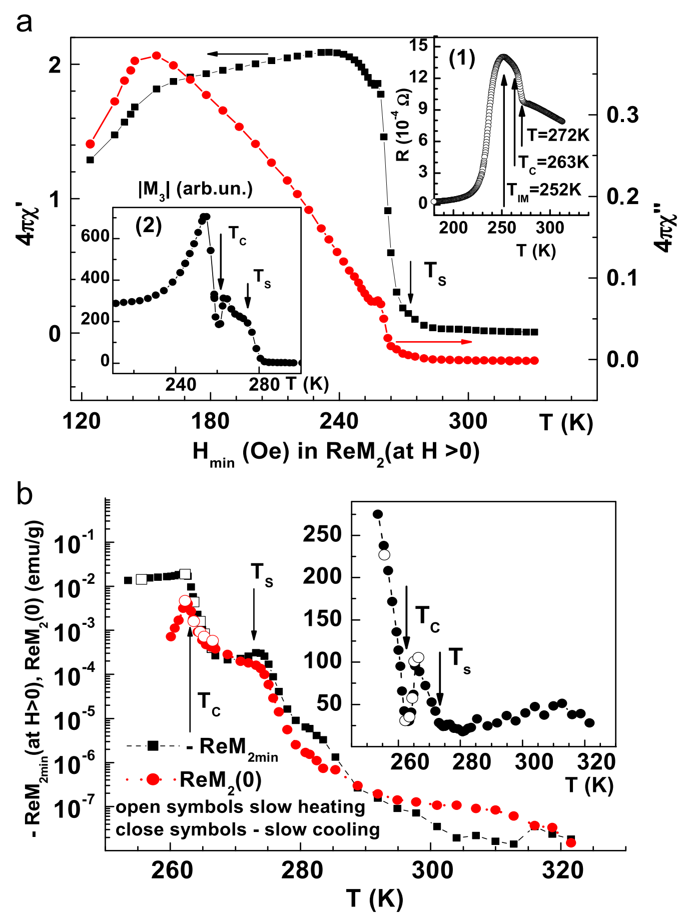

Fig. 1. $La_{0.83}Sr_{0.17}MnO_3$. Temperature dependences of ac susceptibility χ (panel (a)) and values of M_2 phase components in extremes (panel (b)). Insets of panels (a) show the T-dependences of the sample resistance—(1), of $|M_3|$—(2); and inset of panel (b) presents the H-position of the extremum in the Re $M_2(H)$.

isolated one-domain F clusters [5]. It is the first stage of clusters formation, resulting probably from lattice defects. Upon cooling, this "single-particle" signal increases sharply without the noticeable changes in its shape from $T^\#$ down to $T_S \approx 272$ K, where its value is about 10^3 times larger than the value at $T^\#$ (see Fig. 2 at close $T=273.9$ K and Fig. 1b). Stability of the H_{min} over this T-interval indicates that the signal shape does not change. It is the second stage of the evolution of a clustered state. Then Re $M_{2min}(T)$ exhibits a kink from T_S down to 267 K which is a result of the structural transition (Fig. 1b). Below 267 K, the signal sharply increases, reaching an extremum at T_C. As Fig. 1b shows, Re $M_2(0)$, that does not equal zero due to a remnant magnetization of the sample, reveals a very close T-dependence in this T range. The described behavior may be attributed solely to increasing cluster density. But, as we will see below, a comparison of this result with the data for a similar stage in LSCO shows that the growth (from $T^\#$ to T_S) here is several times larger than that in LSCO, where the matrix is non-F. This suggests that in LSMO an interaction of the F-clusters with the matrix that is in the critical regime can be very important, leading to an enhancement of the cluster signal.

Below T_C, a new hysteretic signal appears in very weak H (Fig. 2, 260.7 K) which is certainly related to development of an F ordering of the matrix. It exhibits a growth accompanied by a transformation on cooling down approximately to 244.5 K (Fig. 2, the panels below 260.7 K). As a comparison of Fig. 2a for Re $M_2(H)$ at 244.5 and 225.6 K shows, this signal almost disappears at 225.6 K. Fig. 2a for Re $M_2(H)$ also indicates that the response of