INFLUENCE OF PHASE SEPARATION ON THE LOW-FIELD MAGNETIC PROPERTIES OF $La_{1-x}Sr_xMn_{1-y}Fe_yO_3$

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Abstract

The magnetic properties of ceramic $La_{1-x}Sr_xMn_{1-y}Fe_yO_3$ samples with $x = 0.3$ and $y = 0.15–0.25$ are investigated in magnetic fields of $B = 10–10^3$ G at a temperature T between 5–310 K. A ferromagnetic transition at T_C, decreasing considerably with y, is accompanied with strong magnetic irreversibility at $B = 10$ G. The critical behavior of $\chi\ (T) \sim (T/T_C - 1)^{-\gamma}$ is governed at $y = 0.15–0.20$ by the percolative ($\gamma \approx 1.8$) and Heisenberg ($\gamma \approx 1.4$) spin systems.

1. Introduction

$La_{1-x}Sr_xMn_{1-y}Fe_yO_3$ (briefly LSMFO) belongs to a family of hole-doped mixed-valence ($Mn^{3+/4+}$) manganite perovskites, attracting considerable attention due to a rich magnetic phase diagram and interesting transport properties, including colossal magnetoresistance [1]. The electronic and magnetic properties of these compounds are strongly affected by the interplay between the spin ordering, charge and orbital degrees of freedom [1], as well as by the phase separation or generation of nanosize hole-rich ferromagnetic (FM) particles in the paramagnetic or antiferromagnetic host matrix [1, 2].

The magnetic properties of manganite perovskites are quite sensitive to doping with Fe due to two possible reasons. The first one is connected to a direct replacement of Mn^{3+} by Fe^{3+}, which do not support the FM double-exchange (DE) interactions in Fe^{3+}–Mn^{4+} pairs suppressing the FM properties of the material [3]. The second reason is determined by an additional microscopic disorder, induced by doping with Fe [4].

In this work, we study the low-field magnetic properties of LSMFO to obtain information about the microscopic magnetic state of this material.

2. Results and discussion

LSMFO samples with $x = 0.3$ and $y = 0.15$, 0.20, and 0.25 (marked below as #15, #20 and #25, respectively) were obtained with the conventional solid-state reaction method [1]. Magnetization $M\ (T)$ was measured using an RF-SQUID magnetometer after cooling the sample in a zero magnetic field (M_{ZFC} or zero-field cooled) or in fields of $B = 10$ G, 0.5 kG and 1 kG (M_{FC} or field-cooled).

In the left panel of Fig. 1, both χ_{ZFC} (T) and χ_{FC} (T) (where $\chi \equiv M / B$) at B = 10 G exhibit a FM transition at the Curie temperature T_C determined by inflection points of the corresponding curves ($T_C^{(in)}$). This transition is accompanied by magnetic irreversibility or deviation of χ_{ZFC} (T) from χ_{FC} (T) in #15 and #20, with evidence for frustrated magnetic ground state of LSMFO.

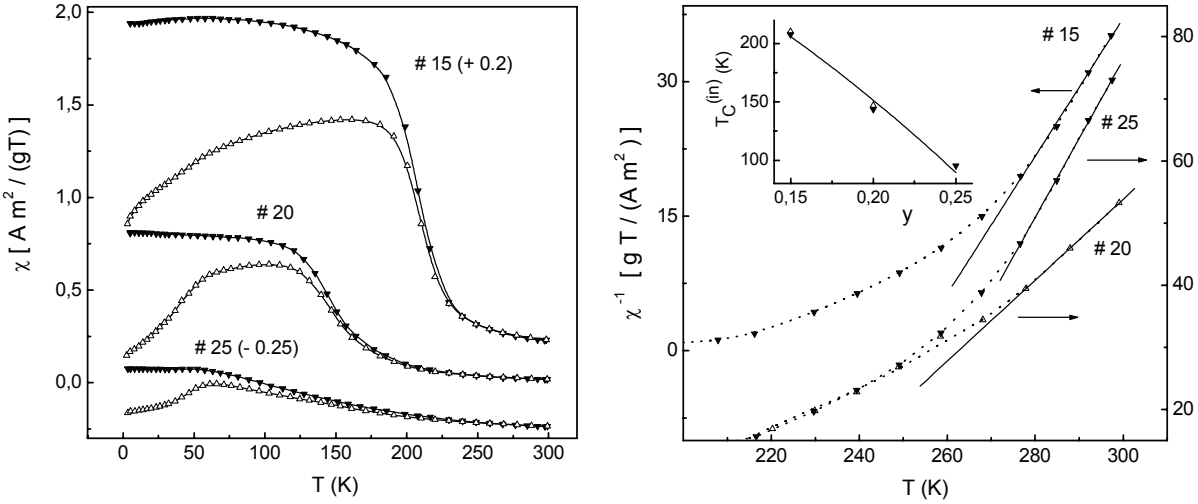

Fig. 1. Dependences of χ_{ZFC} (Δ) and χ_{FC} (▼) on T at B = 10 G. Some plots are shifted along the vertical axes by the values given in parenthesis (left panel). Plots of χ^{-1} vs. T in the ZFC (Δ) and FC (▼) regimes. Solid lines are linear fits (right panel). Inset: plot of $T_C^{(in)}$ vs. y, obtained in the ZFC (Δ) and FC (▼) regimes. The line is the fit with Eq. (1).

As the field increases, the irreversibility is damped strongly and disappears completely in #15 at B = 0.5 kG and in #20 at B = 1 kG, whereas in #25 it persists below ~30 K even up to B = 1 kG. In addition, a considerable decrease of χ (T) with increasing y takes place, whereas $T_C^{(in)}$ is damped appreciably with y (inset to the right panel of Fig. 1).
Below, we analyze T_C with the Varma model [5] given by the expression

$$kT_C \approx 0.05\, W\, c\, (1 - c), \qquad (1)$$

where c is the concentration of the holes or Mn^{4+} and W is the width of the electron band [5]. The direct substitution of Mn^{3+} for Fe^{3+}, which does not support the FM DE interactions (see above), leads to $c \approx c_0 - y$, where $c_0 \sim x$. Fit of the dependence of $T_C^{(in)}$ (y) with Eq. (1), given by the solid line in the inset to the right panel of Fig. 1, yields the values of $W = 2.6 \pm 0.1$ eV and $c_0 = 0.31 \pm 0.01$. Hence, c_0 is close to x and W to the value of $W \approx 2.5$ eV in LSMO [5]. Therefore, the decay of T_C with y in LSMFO is governed by the breaking of the DE interaction by Fe^{3+}, whereas the disorder induced by doping with Fe plays a negligible role.

At $T >> T_C$ the irreversibility disappears and an asymptotic Curie-Weiss behavior (the right panel of Fig. 1), given by the law χ (T) = $C/(T - \theta)$, is observed. Here $C = p_{eff}^2\, \mu_B^2 N/(3\, k)$ is the Curie constant, p_{eff} is the effective Bohr magneton (μ_B) number per magnetic ion, N is the concentration of the magnetic ions and θ is the Weiss temperature. Linear fit of the plots of χ^{-1} vs. T (the solid lines in the right panel of Fig. 1) yields the values of $p_{eff}^2 \sim 140 - 300$, exceeding considerably those of single ions, $p_{eff}^2 = 24$ (Mn^{3+}), 15 (Mn^{4+}) and 35 (Fe^{3+}). This implies the onset of the phase separation already above the room temperature.

As the temperature lowers, a violation of the Curie-Weiss law in the right panel of Fig. 1 is attributable to the critical behavior of χ (T) ~ $(T/T_C - 1)^{-\gamma}$ at $T \to T_C$, where γ is the

critical exponent depending on the nature and dimensionality of a spin system [6]. Below, the critical behavior of χ (T) is analyzed, using the law above in the two equivalent forms:

$$\chi^{-1} - \chi_C^{-1} \sim \tau^{\gamma} \text{ and } d\,\chi^{-1}/d\,T \sim \tau^{\gamma-1}, \quad (2)$$

where $\tau = T/T_C - 1$ and χ_C^{-1} (T) $\equiv \chi^{-1}$ (T_C), by interpolation of χ (T) and variation of T_C by a step of 0.5–1 K, to achieve the minimum standard deviation (SD) of the plots of ln ($\chi^{-1} - \chi_C^{-1}$) vs. τ and ln ($d\,\chi^{-1}$ / $d\,T$) vs. τ. This analysis yields the pairs of T_C and γ or T_C and $\gamma - 1$, applying the first or the second of Eqs. (2), respectively, and the optimum temperature interval, $\Delta\,T$, corresponding to the minimum of the SD vs. T_C plots.

The results of the above analysis are shown selectively in Fig. 2. In all samples, in both conditions of cooling at B = 10 G, two different intervals $\Delta\,T_1$ and $\Delta\,T_2$ were found, where the formal dependence of SD (T_C) exhibited a well-defined minimum.

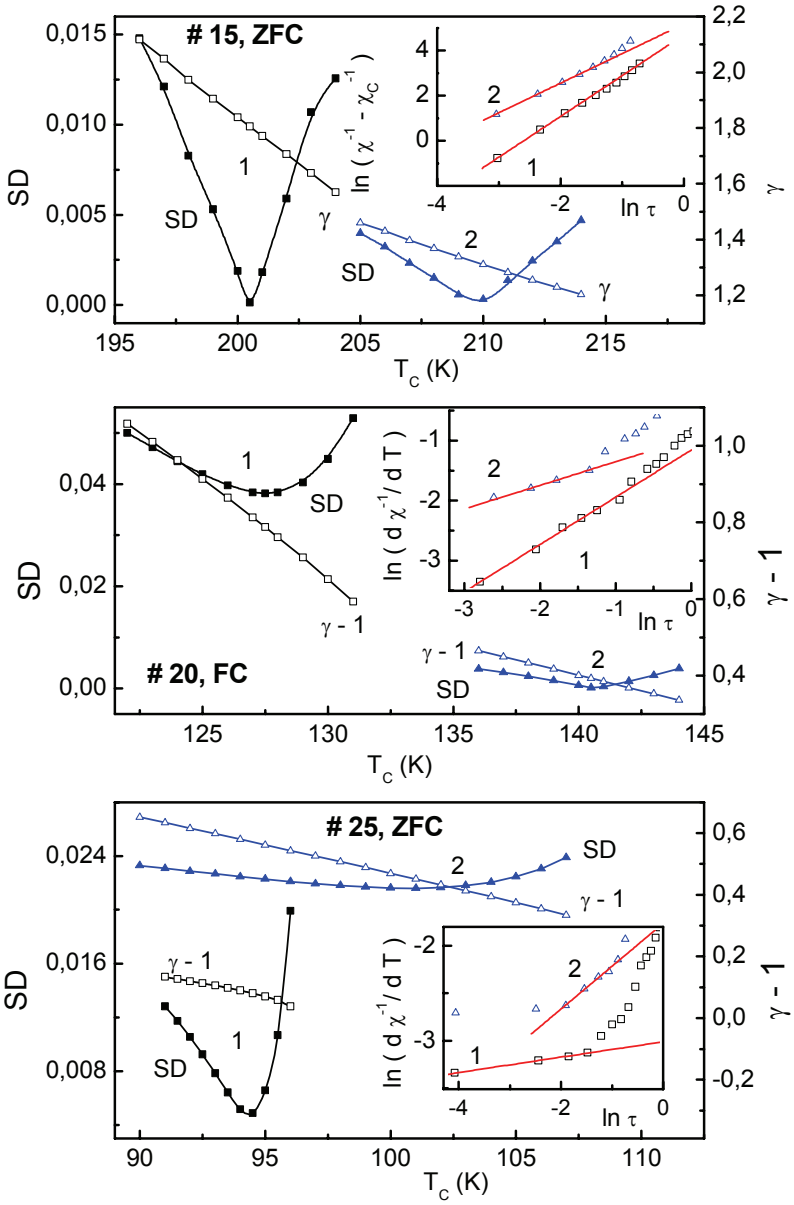

Fig. 2. Plots of SD vs. T_C and γ (top panel) or $\gamma - 1$ (middle and bottom panels) vs. T_C in the intervals $\Delta\,T_1$ (1) and $\Delta\,T_2$ (2). Insets: dependences of ln ($\chi^{-1} - \chi_C^{-1}$) (in arbitrary units, top panel) and ln ($d\,\chi^{-1}$ / $d\,T$) (in arbitrary units, middle and bottom panels) on τ in the intervals $\Delta\,T_1$ (1) and $\Delta\,T_2$ (2). The lines are linear fits.

These minima yield the values of $T_C^{(1)}$ and $T_C^{(2)}$ ($T_C^{(1)} < T_C^{(2)}$) corresponding to different values of γ_1 and γ_2. In #15 and #20, the values of γ_1 and γ_2 are concentrated around those of $\gamma_p = 1.80$ and $\gamma_H = 1.39$, respectively, predicted for a percolative spin system [7] and non-percolative (Heisenberg) spin system [6], respectively. In #25 γ_2 is close to γ_H, and γ_1 to the mean-field value, $\gamma_{mf} = 1$ [6], implying the absence of the percolation behavior.

Therefore, the complicated critical behavior of χ (T) in #15 and #20 reflects the coexistence of two different spin systems. The first of them is connected to generation of large and strongly correlated FM clusters attributable to the phase separation and percolation by joining of nanoscale FM particles into critical clusters with lowering temperature [2]. The second system consists presumably of smaller and weakly correlated magnetic units of the material, which do not enter the percolative critical clusters. In #25 the percolative spin system in is not observed. At the same time, the FM properties in #25 are pronounced much weaker than in #15 and #20, but the irreversibility is enhanced (Fig. 1). Hence, it is possible to attribute ferromagnetism of LSMFO to the first or percolative spin system, whereas frustration and magnetic irreversibility to the second and non-percolative system.

The coexistence of the percolative and non-percolative processes influencing the critical behavior of χ (T) was observed in $La_{1-x}Ca_xMn_{1-y}Fe_yO_3$ [4], $La_{1-x}Ca_xMnO_3$ [8], and $La_{1-x}Ba_xMnO_3$ [9]. Similar to these works, one can find the mean radius of FM particles $r \approx$ 3.4 and 3.8 nm and the mean magnetic moment $\mu/\mu_B \approx 1.4\times10^5$ and 1.5×10^5 in #15 and #20, respectively, which are typical of the nanosize FM clusters in manganite perovskites [4, 8, 9].

Conclusions

The low-field magnetic properties of LSMFO give evidence of the coexistence of the FM and irreversible magnetic properties. A substantial decay of T_C with Fe doping is governed by the damping of the FM DE interactions by Fe^{3+}. The asymptotic Curie-Weiss behavior well above T_C yields the values of p_{eff} considerably exceeding those of single magnetic ions. The complicated critical behavior of χ (T) indicates the existence of the percolative and non-percolative or Heisenberg spin systems. The low-field magnetic properties of LSMFO above are connected to the phase separation effect pertinent to the manganite perovskites.

References

[1] J.M.D. Coey, M. Viret, and S. von Molnar, Adv. Phys., 48, 167, (1999).
[2] E. Dagotto, Nanoscale Phase Separation and Colossal Magnetoresistance, Springer, Berlin, 452, 2002.
[3] K.H. Ahn, X.W. Wu, K. Liu, and C.L. Chien, Phys. Rev. B, 54, 15299, (1996).
[4] R. Laiho, K.G. Lisunov, E. Lähderanta, J. Salminen, and V.S. Zakhvalinskii, J. Magn. Magn. Mater., 250, 267, (2002).
[5] C.M. Varma, Phys. Rev. B, 54, 7328, (1996).
[6] H.E. Stanley, Introduction to Phase Transitions and Critical Phenomena, Clarendon, Oxford, 307, 1971.
[7] A.G. Dunn, J.W. Essam, and D.S. Ritchie, J. Phys. C, 8, 4219, (1975); S. Kirkpatrick, Phys. Rev. Lett., 36, 69, (1976).
[8] H. Huhtinen, R. Laiho, E. Lähderanta, J. Salminen, K.G. Lisunov, and V.S. Zakhvalinskii, J. Appl. Phys., 91, 7944, (2002).
[9] R. Laiho, K.G. Lisunov, E. Lähderanta, V.S. Zakhvalinskii, V.L. Kozhevnikov, I.A. Leonidov, E.B. Mitberg, and M.V. Patrakeev, J. Magn. Magn. Mater., 293, 892, (2005).