



Pressure induced antiferromagnetism in the manganite $\text{La}_{0.7}\text{Sr}_{0.3}\text{Mn}_{0.83}\text{Nb}_{0.17}\text{O}_3$



M.T. Vu ^{a, b, *}, D.P. Kozlenko ^a, S.E. Kichanov ^a, I.O. Troyanchuk ^c, E.V. Lukin ^a, L.H. Khiem ^b,
B.N. Savenko ^a

^a Frank Laboratory of Neutron Physics, JINR, 141980, Dubna, Moscow Reg., Russia

^b Institute of Physics, Vietnam Academy of Science and Technology, 100000, Hanoi, Viet Nam

^c Scientific-Practical Materials Research Center NAS of Belarus, 220072, Minsk, Belarus

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ABSTRACT

The crystal and magnetic structure of a manganite $\text{La}_{0.7}\text{Sr}_{0.3}\text{Mn}_{0.83}\text{Nb}_{0.17}\text{O}_3$ have been studied by means of a neutron diffraction method at pressures up to 4.5 GPa and in temperature range 4–290 K. At ambient pressure and temperature below $T_C = 150$ K a ferromagnetic FM state is stabilized in this compound. At pressures $P \geq 1.5$ GPa and temperature below $T_N \sim 110$ K appearance of a new A-type antiferromagnetic AFM phase in $\text{La}_{0.7}\text{Sr}_{0.3}\text{Mn}_{0.83}\text{Nb}_{0.17}\text{O}_3$ has been observed. A further pressure increasing leads to an increase in the volume fraction of the A-type antiferromagnetic AFM phase, which coexists with the suppressed ferromagnetic phase in the whole experimental pressure range. The Curie and Néel temperature increase upon lattice contraction with comparable pressure coefficients $dT_C/dP = 2.1$ K/GPa and $dT_N/dP = 1.6$ K/GPa. The structural mechanisms of the changes in magnetic structure of $\text{La}_{0.7}\text{Sr}_{0.3}\text{Mn}_{0.83}\text{Nb}_{0.17}\text{O}_3$ manganite are discussed.

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1. Introduction

Manganites of a perovskite type $\text{R}_{1-x}\text{A}_x\text{MnO}_3$ (where are R – rare earth and A – alkali earth elements) exhibit distinguished magnetic and electronic properties, especially the colossal magnetoresistance (CMR) effect [1]. In addition, they exhibit unique physical properties like the electron, orbital and spin ordering, metal-insulator phase transition, mesoscopic phase separation, rich magnetic and electronic phase diagrams depending on the R or A elements [2,3]. It should be noted, that rich spectra of a manganites magnetic properties depend substantially on delicate balance between the ferromagnetic (FM) interactions mediated by itinerant charge carriers via double – exchange mechanism [4–6] and the superexchange antiferromagnetic (AFM) interactions between localized spins of manganese ions [7]. The FM double exchange is usually stronger than AFM superexchange for the doping levels $0.18 < x < 0.5$, while for higher x values AFM superexchange interaction becomes dominant.

More recently it was found that ferromagnetic state can be also formed in manganites containing only Mn^{3+} ions via orbital-disorder mechanism, providing ferromagnetic character of superexchange interactions due to virtual electron transfer from half-filled e_g orbitals to empty ones [8]. An interesting model system allowing study of evolution of the magnetic state of optimally doped manganites by variation of the formal manganese valence is $\text{La}_{0.7}\text{Sr}_{0.3}\text{Mn}_{1-x}\text{Nb}_x\text{O}_3$ [9]. The parent compound $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ is a semimetal with the rhombohedral structure demonstrating one of the largest Curie temperatures among doped manganites, $T_C \approx 373$ K [10]. A substitution with Nb^{5+} ions up to $x = 0.15$ leads to a variation of the formal Mn valence from 3.3 to 3.0 and significant changes in physical properties. The $\text{La}_{0.7}\text{Sr}_{0.3}\text{Mn}_{0.85}\text{Nb}_{0.15}\text{O}_3$ compound is a semiconductor with the orthorhombic structure, demonstrating CMR effect, comparable with conventional mixed valence manganites and much reduced Curie temperature $T_C = 123$ K [8].

The balance between FM and AFM interactions in manganites can be directly tuned by a pressure application through the variation of the structural parameters like bond lengths and bonds angles [11–15]. In framework of the double-exchange model, the pressure application leads to enhancement of the double exchange $\text{Mn}^{3+}-\text{O}^{2-}-\text{Mn}^{4+}$ interactions and as result to an increasing of a

* Corresponding author. Joint Institute for Nuclear Research, FLNP, 141980, Moscow Reg., Dubna, Joliot-Curie st, 6, Russia.

E-mail address: vmthanh@iop.vast.ac.vn (M.T. Vu).

Curie temperature T_C , which is approximately equal to the insulator-metal transition temperature T_{IM} [1] for manganites with the doping levels $x < 0.5$ [16]. The pressure coefficient depends strongly on the electron phonon coupling strength, enlarging its value [17]. More interestingly, in orthorhombic manganites an appearance of the A-type AFM state coexisting with the FM ones was observed under pressure. This effect was attributed to anisotropic lattice compression, leading to orbital ordering effects [17].

Unlike conventional mixed-valence manganites, high pressure effects in compounds containing preferentially Mn^{3+} ions, which magnetic properties are mediated by $Mn^{3+}-O^{2-}-Mn^{3+}$ FM superexchange interactions, are much less studied. In the present study, the crystal and magnetic structure of $La_{0.7}Sr_{0.3}Mn_{1-x}Nb_xO_3$ manganite with $x \sim 0.15$ has been studied by means of neutron diffraction at high pressures up to 4.5 GPa and in the temperature range 4–290 K.

2. Experimental section

The polycrystalline sample $La_{0.7}Sr_{0.3}Mn_{1-x}Nb_xO_3$, $x \approx 0.15$, was prepared by means of solid-phase reaction according to a standard ceramic technology. The details of the synthesis procedure were described in previous work [18]. Further chemical analysis of the sample stoichiometry has shown that Nb content slightly exceeds the expected one and more precise chemical formula of the sample is $La_{0.7}Sr_{0.3}Mn_{0.83}Nb_{0.17}O_3$, providing formal manganese valence of 2.952.

Neutron powder diffraction measurements at ambient and high pressures up to 4.5 GPa were performed at selected temperatures in the range 4–300 K with the DN-6 diffractometer at the IBR-2 high-flux pulsed reactor (FLNP JINR, Dubna, Russia) using the sapphire anvil high pressure cells [19]. Several tiny ruby chips were placed at different points of the sample surface and the pressure was determined by a standard ruby fluorescence technique. Measurements of the pressure distribution on the sample yield typical pressure inhomogeneities of $\pm 15\%$. Diffraction patterns were collected at fixed scattering angle $2\theta = 90^\circ$ (with the resolution $\Delta d/d = 0.012$ at $d = 2 \text{ \AA}$). Experimental data were analysed by the Rietveld method using the Fullprof software [20].

3. Results

Neutron diffraction patterns of $La_{0.7}Sr_{0.3}Mn_{0.83}Nb_{0.17}O_3$ manganite measured at different pressure and temperatures are shown in Fig. 1. The relevant $La_{0.7}Sr_{0.3}Mn_{0.85}Nb_{0.15}O_3$ compound was previously found to crystallize in the rhombohedral structure of the $R\bar{3}c$ symmetry and on temperature lowering a phase transition to the orthorhombic $Pnma$ phase was detected [9]. For the sample $La_{0.7}Sr_{0.3}Mn_{0.83}Nb_{0.17}O_3$ studied in the present work, the orthorhombic structure with the $Pnma$ symmetry retains over the whole studied pressure and temperature range. The obtained from experimental data structural parameters at ambient pressure and room temperature are $a = 5.521(2)$, $b = 7.812(3)$ and $c = 5.557(1) \text{ \AA}$ and these values agree well with obtained previously [18].

The pressure dependence of the lattice cell parameters and unit cell volume of compound $La_{0.7}Sr_{0.3}Mn_{0.83}Nb_{0.17}O_3$ are shown in Fig. 2. The obtained linear compressibility coefficients of the lattice parameters $k_{ai} = -(1/a_{i0})(da_i/dP)_T$ ($a_i = a, b, c$) are $k_a = 0.0024(1) \text{ GPa}^{-1}$, $k_b = 0.0024 \text{ GPa}^{-1}$ and $k_c = 0.0031(4) \text{ GPa}^{-1}$.

The unit cell volume compressibility data (Fig. 2) are fitted by the third-order Birch–Murnaghan equation of state [21]:

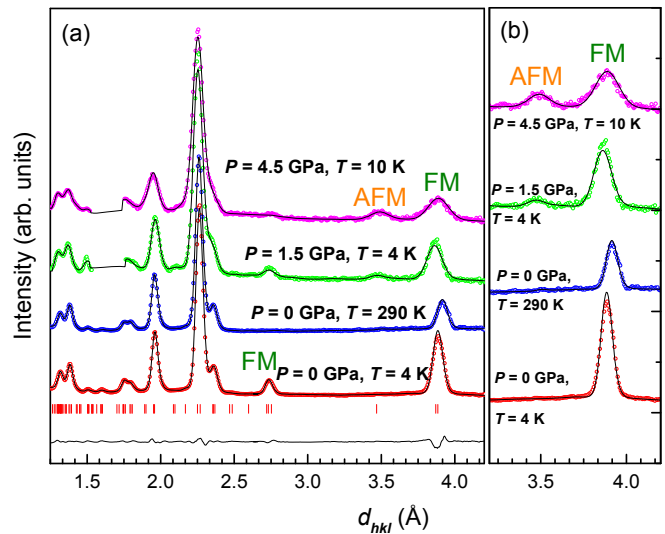


Fig. 1. a) Neutron diffraction patterns of $La_{0.7}Sr_{0.3}Mn_{0.83}Nb_{0.17}O_3$ at selected pressures and low temperature 4 K and room temperature 290 K. The experimental points and calculated profiles by the Rietveld method are shown. b) Enlarged spectra region corresponded to presence of the most intense AFM and FM peaks.

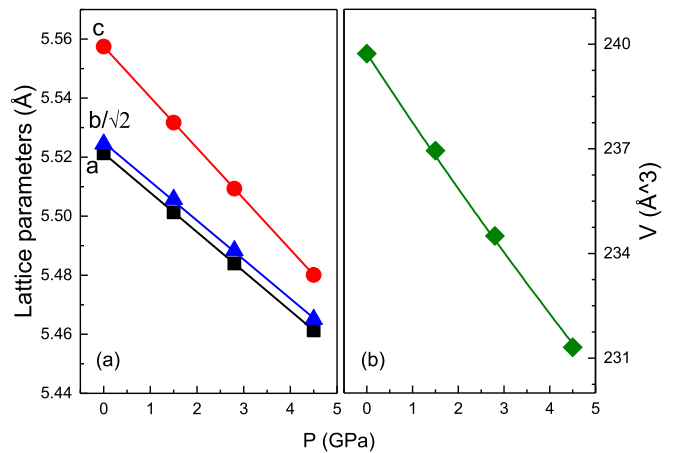


Fig. 2. a) Pressure dependence of lattice parameters and b) unit cell volume of $La_{0.7}Sr_{0.3}Mn_{0.83}Nb_{0.17}O_3$, the solid lines are linear fitting of lattice parameters and corresponded fitting of lattice volume by the third-order Birch–Murnaghan equation (1).

$$P = \frac{3}{2}B_0 \left(x^{-\frac{7}{3}} - x^{-\frac{5}{3}} \right) \left[1 + \frac{3}{4} (B' - 4) \left(x^{-\frac{2}{3}} - 1 \right) \right], \quad (1)$$

where $x = V/V_0$ is the relative change in volume, V_0 is the unit cell volume at $P = 0 \text{ GPa}$, $B_0 = -V(dP/dV)_T$ is the bulk modulus, and $B' = (dB_0/dP)_T$ is the derivative of the bulk modulus with respect to pressure. The best fit is achieved by the following values: $V_0 = 239.6(8) \text{ \AA}^3$, $B_0 = 118(3) \text{ GPa}$ and $B' = 4.0(1)$. The obtained bulk modulus is noticeably smaller in comparison with one $B_0 = 167 \text{ GPa}$ found for the rhombohedral $La_{0.7}Sr_{0.3}MnO_3$, with mixed valence Mn^{3+}/Mn^{4+} ions [22] and comparable with one $B_0 = 108 \text{ GPa}$ for the orthorhombic $LaMnO_3$, containing Mn^{3+} ions only [11].

In the orthorhombic structure of $La_{0.7}Sr_{0.3}Mn_{0.83}Nb_{0.17}O_3$ manganite, the oxygen octahedra MnO_6 contain three pairs of nonequivalent Mn–O bonds: Mn–O1 bonds directed along the b axis and Mn–O2a and Mn–O2b bonds lying in the (ac) plane (Fig. 3). At high pressure Mn–O bonds are compressed anisotropically and the

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