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Ru-induced spin rotation in $La_{1.2}Sr_{1.8}Mn_{1.9}Ru_{0.1}O_7$ bilayered manganite

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Abstract

We have studied the magnetic properties of Ru-doped $La_{1.2}Sr_{1.8}Mn_{1.9}Ru_{0.1}O_7$ bilayered manganite using neutron diffraction (ND) technique. The ND measurements show that the compound exhibits ferromagnetic ordering with the spins direction parallel to the *c*-axis. This is in sharp contrast to the parent compound where the spins are oriented along the *a–b* plane. The large spin–orbit coupling constant of Ru^{+4} ion ($\lambda = \sim 900\,cm^{-1}$) seems to play an important role on lifting of the orbital degeneracy of the e_g state by which the e_g electron prefers to occupy the $d_{3z^2-r^2}$ orbital and therefore, the spins align along the *c*-axis.

Keywords: Layered manganite; Neutron scattering; Spin rotation

The effect of dimensionality of the Mn–O–Mn networks has been studied on the magnetic and transport properties of layered perovskite manganites having the general formula $(La, Sr)_{n+1}Mn_nO_{3n+1}$; where the dimensionality, $n = 1, 2, \infty$ [1–4]. The crystal lattice of the layered perovskites consists of perovskite-like blocks of vertex sharing MnO_6 octahedra which extend to infinity in the *a–b* plane and have a thickness of *n* octahedra parallel to the *c*-axis; neighboring blocks are separated by a rock–salt layer such that the overall composition can be described as $[(La, Sr)MnO_3]_n(La, Sr)O$. Recently, the bilayered compound ($n = 2$) $La_{2-2x}Sr_{1+2x}Mn_2O_7$ ($0.3 \leqslant x \leqslant 0.5$) has become of special interest, because it shows large anisotropic magnetoresistance (AMR) than any other layered manganites. The Sr-doped bilayered manganite contains the mixed valent state of Mn; Mn^{+3} ($3d^4$) and Mn^{+4} ($3d^3$). For the octahedral symmetry of MnO_6 octahedra the configuration becomes $t_{2g}^3e_g^1(^5E_g)$ for Mn^{+3} and $t_{2g}^3(^4A_{2g})$ for Mn^{+4}. The e_g electrons are considered as mobile charge carriers interacting with the localized

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Mn^{+4} spins in the framework of Zener's double exchange model [5]. This induces ferromagnetic and metallic behavior in the compound. However, experimental results have revealed that the observed features in bilayered manganite cannot be attributed only to the double exchange interaction but also to spin, orbital and lattice degrees of freedom [3,4]. Okamoto et al. [6] have studied the correlation between orbital structure and magnetic ordering for a large number of bilayered compounds based on the mean field approximation, and found that the relative stability of $d_{x^2-y^2}$ and $d_{3z^2-r^2}$ orbitals of the e_g state dominates the magnetic structure as well as the magnetic transition temperature. This result also shows that there is an optimal mixing between the $d_{x^2-y^2}$ and $d_{3z^2-r^2}$ orbitals in the ferromagnetic ordering state. Recent band structure calculations and angle resolved photoemission spectroscopy (ARPES) measurements reported by Dessau et al. [7] in single crystal of $La_{1.2}Sr_{1.8}Mn_2O_7$ indicate that the relative energy of $d_{x^2-y^2}$ is very close to the $d_{3z^2-r^2}$ orbital and these bands overlap substantially along the in-plane k vector. As the energy difference between $d_{x^2-y^2}$ and $d_{3z^2-r^2}$ is very small, a small change either in the axial or equatorial bond lengths of the regular MnO_6 octahedron through hole doping [2], pressure [3], temperature [4] and magnetic field [8] alters the ground state of e_g electrons, which in turn alters the magnetic and electronic properties of bilayered manganites. The small energy difference between $d_{x^2-y^2}$ and $d_{3z^2-r^2}$ orbital also indicate that the ground state of e_g electrons can be altered by a doping ion, which is having strong spin–orbit coupling constant (orbital magnetic field), at the Mn-site in bilayered mangnaite systems. Consequently, one can manipulate in the magnetic structure of bilayered manganites by means of doping other 3d or 4d transition elements at the Mn-site and hence, the magnetic and electronic properties. Here, we demonstrate that 5% Ru (spin–orbit coupling constant $\lambda = 900\,cm^{-1}$) is able to rotate the spins of $La_{1.2}Sr_{1.8}Mn_2O_7$ from the a–b plane to the c-axis.

Bulk samples of $La_{1.2}Sr_{1.8}Mn_2O_7$ and $La_{1.2}Sr_{1.8}Mn_{1.9}Ru_{0.1}O_7$ were prepared by the conventional solid-state method. The phase purity of the samples was characterized by X-ray diffraction using Cu K_α source, which shows a single-phase pattern for both parent compound as well as Ru-doped compound. Magnetization measurements were carried out using an Oxford VSM model 3001. The ferromagnetic transition temperature (T_c) for the parent compound $La_{1.2}Sr_{1.8}Mn_2O_7$ and $La_{1.2}Sr_{1.8}Mn_{1.9}Ru_{0.1}O_7$ is found at ~115 K and ~140 K, respectively. These transition temperatures are well matched with the earlier reported value [9]. Powder neutron diffraction (ND) patterns at various temperatures were recorded on the powder diffractometer ($\lambda = 1.249$ Å) at Dhruva reactor, Trombay. The Rietveld refinement was carried out using the program FULLPROF [10].

Fig. 1 shows ND patterns for $La_{1.2}Sr_{1.8}Mn_{1.9}Ru_{0.1}O_7$ at temperature $T = 12$ K. To obtain evidence about the direction of the magnetic moment vector, we have analyzed all the magnetic reflections in detail. Because the crystal structure is body centered tetragonal cell only reflections $h+k+l$ even are expected for simple ferromagnetic order. In our case, peaks with $h+k+l =$ even were only found while absence of $h+k+l =$ odd magnetic peaks, indicating that all the spins are aligned ferromagnetically below T_c. Moreover, the absence of magnetic contribution at (0 0 4) (shown by arrow mark in Fig. 1) which corresponds to the spins direction along the a–b

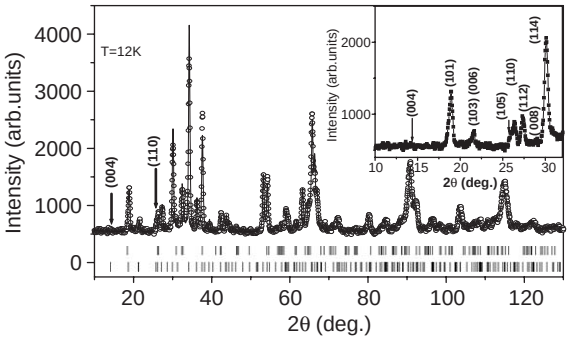

Fig. 1. Observed and calculated powder ND patterns at 12 K for $La_{1.2}Sr_{1.8}Mn_{1.9}Ru_{0.1}O_7$. Reflections positions are marked for both the majority (lower) and minority phases (upper). The low angle peaks are magnified (Inset of Fig. 1). This data shows that the (0 0 4) magnetic reflection, which corresponds to the spins direction along the a–b plane, is absent.