Vol. 97 (2000) *ACTA PHYSICA POLONICA A* No. 5

Proceedings of the European Conference "Physics of Magnetism '99", Poznań 1999

CORRELATION BETWEEN MAGNETIC PHASES AND INSULATOR–METAL TRANSITION IN $La_{1/3}Nd_{1/3}Ca_{1/3}MnO_3$ PEROVSKITE

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The $La_{1/3}Nd_{1/3}Ca_{1/3}MnO_3$ ceramic perovskites are investigated using the neutron diffraction technique and resistance measurements in the temperature range from 260 K down to 5 K. We have found that the integrated intensity of the antiferromagnetic peaks was growing with decreasing temperature and reached the maximum at a temperature about 20 K lower than that of the semiconductor–metal transition and did not vanish at 5 K. The semiconductor–metal transition is correlated with the temperature of the maximum of the lattice c-parameter and the percolation of ferromagnetic domains. Our results suggest that although the ferromagnetic long-range order is established through the semiconductor–metal transition induced percolation network, locally the antiferromagnetic correlation can remain.

PACS numbers: 71.30.+h, 75.25.+z

1. Introduction

Rare earth-based manganites, with the perovskite structure have recently attracted considerable attention because of their magnetoresistive (MR) properties. From the basic point of view, the scientific studies concentrate on the interplay between the spin, charge, and lattice structure. Nevertheless, the exact magnetic structure as well as the origin of the MR remain controversial. The electrical conductivity and ferromagnetism in such perovskites were explained in terms of the double exchange interaction, i.e. the transfer of electrons between Mn^{3+} and Mn^{4+} ions [1]. The low temperature and low-field MR has been attributed to the spin-polarized intergranular tunneling [2], spin-dependent scattering [3] or micromagnetic behavior associated with alignment of magnetic domains at the grain boundaries [4]. From the neutron experiment one can obtain the qualitative information on the change of the magnetic structure, i.e. the antiferromagnetic (AFM) to ferromagnetic (FM) phase transition, and verify its concomitance with the semiconductor-to-metal transition [5].

In this paper we present both the neutron diffraction results and the resistance measurements for the $La_{1/3}Nd_{1/3}Ca_{1/3}MnO_3$ system. In this compound, according to the magnetic inhomogeneity model of Rao et al. [6], simultaneous occurrence of metallic La-rich ferromagnetic domains and semiconducting Nd-rich nonferromagnetic domains is expected. The magnetic inhomogeneity can originate from different local distortions of the lattice around La^{3+} and Nd^{3+} ions. Consequently, in the crystal structure of $La_{1/3}Nd_{1/3}Ca_{1/3}MnO_3$ compound Nd^{3+} ions are in tension, while La^{3+} ions are in compression.

The aim of this study is to determine the influence of the change of the lattice parameters and the AFM–FM phase transition on the resistivity behavior, i.e. the semiconductor–metal (S–M) transition in the $La_{1/3}Nd_{1/3}Ca_{1/3}MnO_3$ perovskite prepared by the standard ceramic method.

2. Experimental

Polycrystalline $La_{1/3}Nd_{1/3}Ca_{1/3}MnO_3$ samples have been prepared by a conventional solid state reaction method, details were described in Ref. [7]. The neutron diffraction measurements were carried out in Hahn-Meitner-Institut in Berlin, Germany. To determine the type of the magnetic structure we have measured powder patterns for the neutron wavelength $\lambda = 0.2442$ nm. The lattice parameters of the orthorhombic perovskite structure are $a = 0.5480$ nm, $b = 0.5550$ nm, and $c = 0.7737$ nm. All magnetic features can be indexed in the chemical unit cell. The neutron powder diffractograms were collected with the angle resolution of $0.1°$. The DC-resistance R of the samples was measured by the standard two-point probe method over a temperature range of $4.2 \div 260$ K.

3. Results and discussion

The temperature dependences of the resistance $R(T)$ and the Bragg reflection $2\Theta(T)$ for crystallographic [004] direction are shown in Fig. 1. The peak in $R(T)$ indicates the occurrence of the S–M transition at a temperature of 104 K for cooling. The resistance exhibits a metallic behavior ($\mathrm{d}R/\mathrm{d}T > 0$) at low temperatures and a semiconducting behavior ($\mathrm{d}R/\mathrm{d}T < 0$) at high temperatures. From the plot of 2Θ versus T it results that the lattice constant c of the unit cell is approximately temperature independent apart from the range $100 \div 108$ K, in which a sharp increase in its value by about 0.01 nm is observed. Therefore, the change of the resistance type (S–M transition) is correlated with the modification of the unit cell c-parameter.

The $R(T)$ curve for the $La_{1/3}Nd_{1/3}Ca_{1/3}MnO_3$ perovskite is shown in Fig. 2 together with the temperature evolution of the integrated intensity (neutron results) corresponding to the (100) and (110) antiferromagnetic and (110) ferromagnetic reflections, respectively. The integrated intensity of the AFM peaks increases with decreasing temperature and has the maximum value at a temperature 20 K below the S–M transition in resistance measurements.

The onset of the ferromagnetic ordering at a temperature 20 K above the S–M transition supports the interpretation proposed by Rao et al. [6], which postulates the presence of multiple magnetic phases. We assume that lowering the

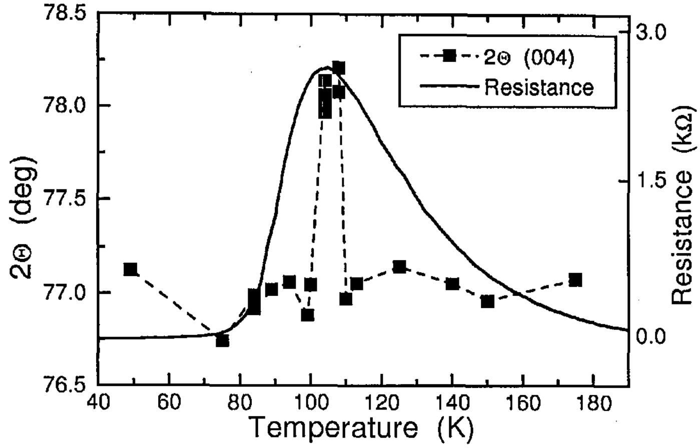

Fig. 1. The angle 2Θ of the structural (004) reflection and the DC-resistance R as a function of temperature in the cooling mode for $La_{1/3}Nd_{1/3}Ca_{1/3}MnO_3$ perovskite.

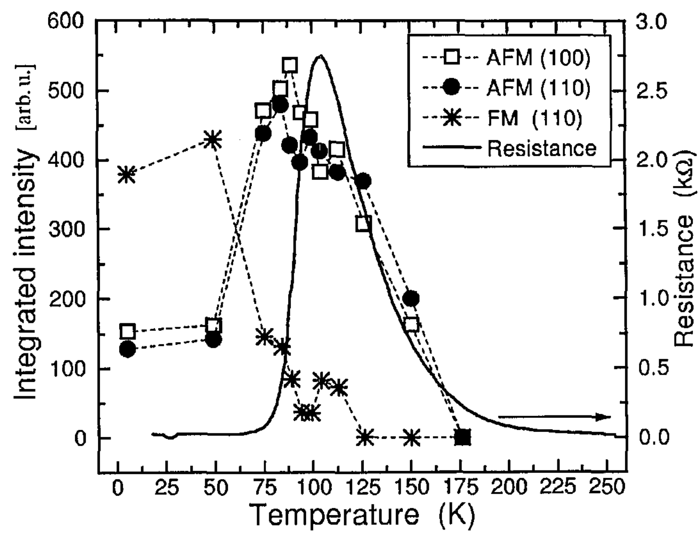

Fig. 2. The integrated peak intensity of anti- and ferromagnetic phases as a function of temperature for $La_{1/3}Nd_{1/3}Ca_{1/3}MnO_3$ perovskite. The temperature dependence of the DC-resistance is shown for comparison.

temperature increases the domains with ferromagnetic ordering, therefore they get into contact, which is accompanied by the peak in resistance. The AFM reflections do not vanish at 5 K implying that even when the FM long-range order is established, locally AFM correlations can remain. The work of Moudden et al. corroborated the same situation [8].

4. Conclusions

Finally, our data can be explained assuming that the local percolation of spin clusters leads both to the peak in resistance (S–M transition) and to the long-range ferromagnetic order. The transitions appear at the same temperature as the change of the lattice c-parameter.

The AFM phase does not show the direct coincidence with the transport properties.

Acknowledgments

This work was supported in part by the Committee for Scientific Research, Poland (grant No. 7 T08D 011 13). We also gratefully acknowledge Hahn-Meitner-Institut in Berlin for making the neutron measurements possible.

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