STRUCTURE AND MAGNETORESISTANCE PROPERTIES OF $(\mathrm{La}_{1-y}\mathrm{Nd}_y)_{1-x}\mathrm{Sr}_x\mathrm{MnO}_3$ POLYCRYSTALLINE SPECIMENS

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Structure, electric and magnetoresistance properties of $(\mathrm{La}_{1-y}\mathrm{Nd}_y)_{1-x}\mathrm{Sr}_x\mathrm{MnO}_3$ ($y = 0 \div 1$; $x = 0.15 \div 0.3$) polycrystalline specimens are studied. Electric measurements are performed in the temperature range 77—370 K, magnetoresistance is measured in fields up to 15 kOe. It is revealed that a substitution of neodymium for lanthanum leads to a substantial increase in magnetoresistance and a decrease in the temperature, at which magnetoresistance displays a maximum. The principal changes in positions of the peaks on the temperature dependences of electro- and magnetoresistance are shown to occur within the range $0 \leq y \leq 0.25$, which correlates with the character of a change of structural parameters. The evidence is presented that the peculiarities of the behavior of $(\mathrm{La}_{1-y}\mathrm{Nd}_y)_{1-x}\mathrm{Sr}_x\mathrm{MnO}_3$ specimens near $y = 0.25$ are associated with the appearance of a structurally and magnetically inhomogeneous state.

Introduction

An intense interest to $\mathrm{La}_{1-x}\mathrm{M}_x\mathrm{MnO}_3$ (M-alkaline-earth elements) doped manganites with the distorted perovskite structure is associated with a strong sensitivity of their electric properties to a magnetic field (the colossal magnetoresistance effect), which makes such materials prospective for the application in magnetoelectronic devices [1,2]. However, in most cases, the considerable changes of magnetoresistance are achieved only in strong fields or at low temperatures, and this seriously limits the abilities of their application. As results from the recent studies, substitutions in lanthanum or manganese sublattices can serve as an effective tool to govern both the magnetoresistance and the temperature range within which its value is maximal [3,4].

The magnetoresistance effect in $\mathrm{La}_{1-x}\mathrm{M}_x\mathrm{MnO}_3$ compounds results from a competition between the different types of interaction among manganese ions. The parent compound LaMnO_3 is an antiferromagnetic insulator with the triply ionized manganese ions which have the $t_{2g}^3 e_g^1$ electron configuration [5,6]. Three electrons located on the lower (t_{2g}) level are coupled by the strong internal (correlation) exchange interaction and behave as a single localized spin ($S = 3/2$). Unlike to the lower level, the e_g-level of manganese is strongly hybridized with the $2p$ level of oxygen. For this reason, the electrons located on it can move between the neighboring manganese sites. A deviation of the filling of e_g-level from 1 (as a result of the substitution of La by one- or divalent elements, or a change in the oxygen content) leads to a drastic transformation of both the magnetic state and conductivity of doped manganites — these materials become highly conductive ferromagnets [1, 2, 5, 6].

A theoretical consideration of the reasons underlying the strong correlation between magnetism and conductivity in doped manganites was first carried out by Zener [7]. He considered the transfer of an e_g-electron between manganese ions in a Mn^{3+}—O—Mn^{4+} chain with either parallel or antiparallel orientation of their spins, and showed that, in the first case (parallel orientation), the realization of such a process can lead to a decrease in the free energy of the system, but it is energetically unfavorable in the second one. Such a conclusion is based on the assumption of conservation of the electron spin orientation during the electron transfer from one cation to another, as well as on the requirement of the existence of a strong correlation interaction which stabilizes the spin orientation of a transferred electron relative to the cation spins, in accordance with the Hund's rules. As a result, the effective coupling appears between the magnetic moments of both cations, which is equivalent to the magnetic interaction. Since the electron transfer is a sequence of two processes, of which the first one corresponds to the transfer of the oxygen p-electron to Mn^{4+} ion, and the second one – the transfer of Mn^{3+} d-electron on its site, the resulting coupling is called double exchange. The domination of double exchange over other types of interactions peculiar to manganites (electron-phonon interaction, antiferromagnetic superexchange, charge ordering etc.) favors the delocalization of charge carriers and the establishment of ferromagnetic ordering [5, 6, 8].

The presence of structural deformations in $La_{1-x}M_xMnO_3$ not only influences the effective transfer integral of e_g-electrons, changing thus the double exchange energy, but also modifies a hierarchy of other interactions, and this can result in a considerable complication of the magnetic structure [3—6]. Hwang *et al.* [3], and Sun *et al.* [4] studied the influence of a substitution of lanthanum by other rare-earth elements and revealed that a decrease in the average ion radius in the lanthanum sublattice leads to a reduction of the magnetic transition temperature as well as to a considerable increase in the magnetoresistance value. More recent works have shown that local distortions of the crystalline lattice near the ions with different radii also play an important role in the above-mentioned effects [6, 9, 10]. To date, the study of an influence of the substitution of lanthanum by neodymium on the structure, magnetism, and electric conductivity has mainly been limited to that for compounds of the $La_{1-x}Ca_xMnO_3$ [4, 10—12], $La_{1-x}Ba_xMnO_3$ [13] systems, and only few papers have been devoted to the investigation of the $La_{1-x}Sr_xMnO_3$ system [13—16].

The aim of this work is a study of the correlation between the structural, electric and magnetoresistance properties of specimens of the $(La_{1-y}Nd_y)_{1-x}Sr_xMnO_3$ system in a wide range of the strontium ($x = 0.15 \div 0.3$) and neodymium ($y = 0 \div 1$) concentrations.

Experiment

The specimens for this investigation were prepared by solid-state reactions, using chemically pure $SrCO_3$ and high-purity La_2O_3, Nd_2O_3, and Mn_2O_3 as raw materials. The starting mixtures were homogenized by ball milling with bidistilled water. After evaporating the residual water, the mixtures were dried at 370—390 K, passed through a capron sieve, and then fired at 1050 °C for 2 h. The resultant powder was pressed into disks 12 mm in diameter and 3—4 mm in thickness, which were then sintered at $T_s = 1300$ °C for 2 h. In the present work, the properties of 3 series of specimens (with $x = 0.15$, 0.2, and 0.3) with the chemical composition presented in Table 1 were studied.

Table 1. Chemical composition of synthesized specimens of the $(La_{1-y}Nd_y)_{1-x}Sr_xMnO_3$ series

Specimen	Chemical composition
Nd_{01}	$(La_{0.75}Nd_{0.25})_{0.85}Sr_{0.15}MnO_3$
Nd_{02}	$(La_{0.5}Nd_{0.5})_{0.85}Sr_{0.15}MnO_3$
Nd_{03}	$(La_{0.25}Nd_{0.75})_{0.85}Sr_{0.15}MnO_3$
Nd_{04}	$(La_0Nd_1)_{0.85}Sr_{0.15}MnO_3$
Nd_{05}	$(La_{0.75}Nd_{0.25})_{0.8}Sr_{0.2}MnO_3$
Nd_{06}	$(La_{0.5}Nd_{0.5})_{0.8}Sr_{0.2}MnO_3$
Nd_{07}	$(La_{0.25}Nd_{0.75})_{0.8}Sr_{0.2}MnO_3$
Nd_{08}	$(La_0Nd_1)_{0.8}Sr_{0.2}MnO_3$
Nd_{09}	$(La_{0.75}Nd_{0.25})_{0.7}Sr_{0.3}MnO_3$
Nd_{10}	$(La_{0.5}Nd_{0.5})_{0.7}Sr_{0.3}MnO_3$
Nd_{11}	$(La_{0.25}Nd_{0.75})_{0.7}Sr_{0.3}MnO_3$
Nd_{12}	$(La_0Nd_1)_{0.7}Sr_{0.3}MnO_3$

X-ray diffraction (XRD) measurements were made on a DRON 3M powder diffractometer (CuK_α radiation, 40 kV, 18 mA). Structural parameters and the phase composition were determined by the Rietveld profile analysis method, using the FullProf program and JCPDS Powder Diffraction File data. XRD patterns were run in the angular range $2\theta = 10 \div 150°$ in a step-scan mode with a step size of 0.02° and a counting time of 10 s per data point. As the external standards, SiO_2 (2θ calibration) and Al_2O_3 (NIST SRM1976 intensity standard) were used. Measurements of electric resistance were carried out by the four-probe method in the temperature range 77—370 K. The specimens were rectangular in shape, 2×3×10 mm in dimensions. Electric contacts were made by applying and annealing a silver paste. Magnetoresistance (MR) was measured in fields up to 15 kOe and is defined as $(R - R_H)/R$, where R is the electric resistance of the specimen in zero field and R_H — in the external magnetic field H.

Results and Discussion

X-ray diffraction measurements have shown that $(La_{1-y}Nd_y)_{1-x}Sr_xMnO_3$ specimens studied in the present work have the rhombohedrally distorted perovskite structure ($R\bar{3}c$ space group). Structural parameters refined with the use of the full-profile Rietveld analysis are presented in Table 2. For each of the series of the specimens, the volume of the elementary cell, V, decreases with an increase in y, as could be expected at the substitution of larger lanthanum ions by smaller neodymium ones (for the nearest environment with the coordination number 6, the ion radii of lanthanum, $R^i_{\rm La}$, and neodymium, $R^i_{\rm Nd}$, are 1.061 and 0.995 Å, respectively). The $V(y)$ dependence is almost linear (Fig. 1), which agrees with the results obtained at the substitution of neodymium for lanthanum in the $La_{2/3}Ca_{1/3}MnO_3$ compound [10]. The shift of the oxygen

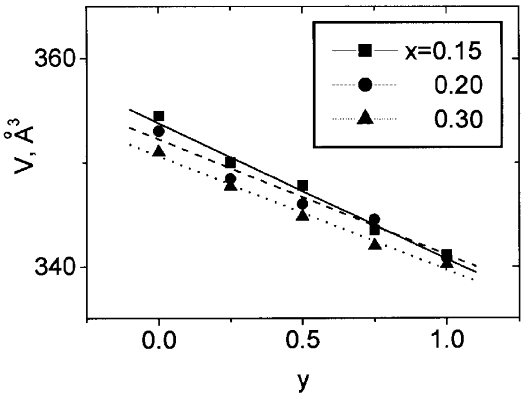

Fig. 1. Volume of the elementary cell vs neodymium content for $(\mathrm{La}_{1-y}\mathrm{Nd}_y)_{1-x}\mathrm{Sr}_x\mathrm{MnO}_3$

coordinate x_O monotonically changes with a decrease of y. However, this change is non-uniform as a function of Nd concentration: first, a sharp drop occurs upon the increase of y from 0 to 0.25, then the change becomes more slower (Fig. 2,*a*). Sharper changes at low levels of Nd doping ($0 \leq y \leq 0.25$) are also characteristic of the concentration dependence of c/a (Fig. 2,*b*) — the parameter, which characterizes a degree of the rhombohedral distortion of the perovskite structure [17]. Here, c and a are the room-temperature lattice constants of the specimens.

It is known that the perovskite cell, being quite compact, puts rather strict restrictions on the ionic radii of the elements it consists of [5, 6]. Though the range of the stability of the perovskite structure with chemical formula ABO_3 is determined by the ratio between the *average* values of R^i_A and R^i_B, the presence of the elements with different ionic radii in A or B sublattice leads to the appearance of considerable local distortions. Taking into consideration that $R^i_{\mathrm{Nd}^{3+}}$ differs from $R^i_{\mathrm{La}^{3+}}$ and even more from $R^i_{\mathrm{Sr}^{2+}}$, an increase in the neodymium content should lead to an enhancement of the inhomogeneity on a local level. Accounting for the strong sensitivity of the electric and magnetic properties of manganites to the average values of structural parameters as well as to the level of local distortions and strains [3—6], the essential changes are expected to occur in the electric and magnetoresistive behavior of the specimens with a change of the neodymium concentration, especially upon the increase of y from 0 to 0.25.

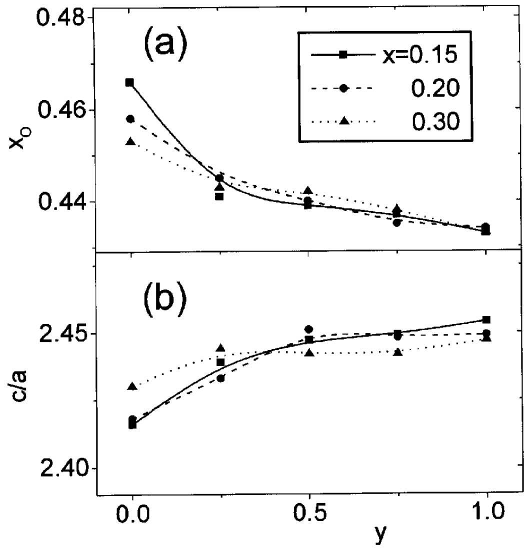

Fig. 2. Oxygen coordinate x_O (*a*) and the ratio between the parameters of the rhombohedral lattice c/a (*b*) vs neodymium content for $(\mathrm{La}_{1-y}\mathrm{Nd}_y)_{1-x}\mathrm{Sr}_x\mathrm{MnO}_3$ specimens

One of the key parameters that determines the physical properties and the type of magnetic ordering in the doped lanthanum manganites is the concentration of a divalent element, namely Sr in $\mathrm{La}_{1-x}\mathrm{Sr}_x\mathrm{MnO}_3$ compound. In accordance with the literature data [6, 19], for $0.15 < x \leq 0.30$, the ground (low temperature) state of $\mathrm{La}_{1-x}\mathrm{Sr}_x\mathrm{MnO}_3$ is ferromagnetic with a metallic character of conductivity ($dR/dT > 0$). For the high-temperature (paramagnetic) state, the conductivity of this compound is of activating character ($dR/dT < 0$). Therefore, $R(T)$ curve displays a maximum near the temperature of the magnetic transition. The temperature of the paramagnetic to ferromagnetic transition, T_C, depends on the strontium concentration and changes from 230 to 370 K as x grows from 0.15 to 0.30 [6]. The application of an external magnetic field H significantly decreases the electric resistance, leading to the appearance of the magnetoresistance effect. Magnetoresistance MR of the single crystalline specimens of doped manganites displays a maximum

Table 2. Structural parameters of $(\mathrm{La}_{1-y}\mathrm{Nd}_y)_{1-x}\mathrm{Sr}_x\mathrm{MnO}_3$ compounds

Specimen	Parameters of the elementary cell			Agreement factors	
	a, Å	c, Å	V, Å^3	R_b, %	R_f, %
Nd_{01}	5.4881(7)	13.390(2)	349.26(7)	7.5	8.3
Nd_{02}	5.4745(9)	13.398(2)	347.8(1)	8.5	9.9
Nd_{03}	5.451(1)	13.350(3)	343.5(1)	8.8	10.85
Nd_{04}	5.434(1)	13.338(3)	341.1(1)	10.95	10.45
Nd_{05}	5.4881(7)	13.358(1)	348.42(8)	7.05	7.4
Nd_{06}	5.4602(9)	13.402(3)	346.0(1)	8.0	8.55
Nd_{07}	5.4594(9)	13.368(2)	345.1(1)	8.0	10.1
Nd_{08}	5.436(1)	13.318(3)	340.8(1)	10.35	10.5
Nd_{09}	5.4341(8)	13.318(3)	347.71(9)	10.7	10.9
Nd_{10}	5.463(1)	13.342(4)	344.8(2)	8.0	7.25
Nd_{11}	5.4475(9)	13.308(3)	342.0(1)	9.9	10.45
Nd_{12}	5.438(1)	13.309(3)	340.9(1)	8.75	10.75

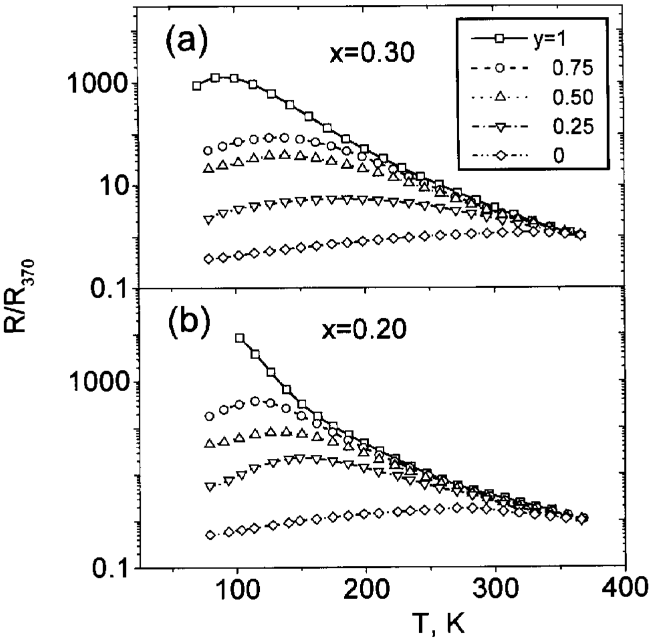

Fig. 3. Normalized electric resistance vs temperature for $(La_{1-y}Nd_y)_{1-x}Sr_xMnO_3$ specimens

near the Curie temperature and declines upon the temperature deviation from T_C [19]. In polycrystalline specimens, however, an additional contribution appears in the low temperature range, which monotonically increases with the temperature lowering. The appearance of this contribution is associated with a spin-dependent scattering of charge carriers in the intergrain area [20] or spin-polarized tunneling through the intergrain boundaries [21].

Electric and magnetoresistance properties of $La_{1-x}Sr_xMnO_3$ specimens with $x = 0.15$, 0.20 and 0.30, which were parent for the study of the influence of the substitution of neodymium for lanthanum in the present work, were in details analyzed in [17, 22]. For the whole range of strontium concentrations, studied in the above papers, the character of the temperature dependences of electric resistance $R(T)$ was shown to be similar to that observed in $La_{1-x}Sr_xMnO_3$ single crystals [19]. $R(T)$ curves displayed a maximum at a certain temperature, T_R, which increased with x. For each value of x, T_R was by 10—15 K lower than the correspondent value for the single crystalline specimens. This was supposed to be the result of a small deviation of the oxygen content from the stoichiometric value [22].

In Fig. 3, the temperature dependences of the normalized electric resistance $R/R_{370}(T)$, where R_{370} is the electric resistance at 370 K, are presented for $(La_{1-y}Nd_y)_{1-x}Sr_xMnO_3$ with $x = 0.20$ and 0.30. For both the series of the specimens, the increase in the neodymium content leads to a considerable decrease in T_R. So, the complete substitution of lanthanum by neodymium in the specimens with $x = 0.30$ decreases T_R from 330 to 92 K, and it falls from 276 to 80 K in the specimens with $x = 0.20$. For the third series ($x = 0.15$), the position of the peak is near 77 K at $y = 0.5$ and is lower than 77 K for the specimens with $y > 0.5$.

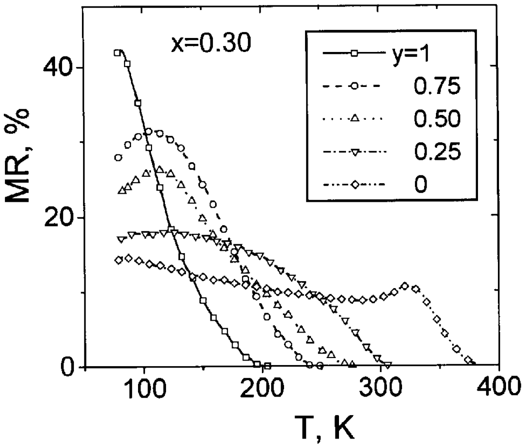

Fig. 4. Magnetoresistance of $(La_{1-y}Nd_y)_{0.70}Sr_{0.30}MnO_3$ specimens as a function of temperature

In Fig. 4, we present the data on the magnetoresistance vs temperature, MR(T), obtained in a field of 15 kOe for $(La_{1-y}Nd_y)_{0.70}Sr_{0.30}MnO_3$ specimens. At room temperature, the noticeable magnetoresistance effect (MR $\sim$ 10 %) is observed only in the specimen which does not contain neodymium. The substitution of 25 % lanthanum by neodymium (here and below, the atomic percentage is implied) leads to both a strong decrease in the magnetoresistance peak temperature, T_{MR}, and to a drastic change of the character of MR(T) curve. Whereas for the $La_{0.70}Sr_{0.30}MnO_3$ specimen, the interval of the temperatures, ΔT, within which MR changes from 0 to its peak value as the temperature is lowered, equals 55 K, for the specimen with $y = 0.25$, it is considerably greater and equals $\Delta T \approx 180$ K. On the further increase of the neodymium content, ΔT decreases again and, upon the complete substitution of La by Nd, equals 110 K. Such a character of the change of magnetoresistance behavior upon the increase in the neodymium concentration is also characteristic of the other series of specimens.

In Fig. 5, the dependences of T_{MR} and the maximal value of magnetoresistance, MR_{max}, as functions of the neodymium content are showed. For each the series of the specimens, the complete substitution of lanthanum by neodymium leads to a $(1.5 \div 2)$-fold decrease in T_{MR}, and a $(2 \div 4.5)$-fold increase in MR_{max}. The principal growth of $\mathrm{MR}_{\max}$ occurs at $y > 0.25$. By contrary, T_{MR} scarcely depends on the neodymium concentration in the area of large y, and strongly decreases as y changes from 0 to 0.25, which correlates with the character of the change of structural parameters (see Fig. 2).

Similar trends in the behavior of T_{MR} and $\mathrm{MR}_{\max}$ were also observed in doped manganites upon the substitution of La by other rare-earth elements [10—14]. As was noted above, it is manganese ions that are magnetically active in doped manganites, and the interaction between these ions occurs with the participation of oxygen anions. Substitutions in the lanthanum sublattice influences the strength of the Mn—Mn interaction through a change of local tensions and distortions. For the perfect perovskite ABO_3 structure, the crystalline symmetry laws put the strict restrictions on the ionic radii and the distance between the ions: $d_{\mathrm{A-O}}/d_{\mathrm{B-O}} = \sqrt{2}$ [5, 6]. Here, $d_{\mathrm{A-O}}$ ($d_{\mathrm{B-O}}$) is a distance between the ions A and O (B and O). In the imperfect perovskite lattice, to keep such a ratio near $\sqrt{2}$, the tensions appear and the equilibrium in the system can only be achieved through the appearance of local deformations. The presence of such deformations influences both the inter-ionic distance and the value of the angle in the B—O—B chain, leading to its deviation from 180° (for a perfect perovskite cell, the correspondent angle is 180°). Therefore, an increase in the degree of local distortions, caused by the substitution of a part of lanthanum ions by neodymium, leads to a decrease in the overlapping of the electron orbitals of neighboring ions, and thus, to a weakening of the exchange interaction between manganese ions. In our case, the general character of $T_{\mathrm{MR}}(y)$ and $\mathrm{MR}_{\max}(y)$ dependences agrees with such a point, but the details are to be discussed separately.

In [10], which is devoted to a study of properties of the $\mathrm{La}_{(2-z)/3}\mathrm{Nd}_{z/3}\mathrm{Ca}_{1/3}\mathrm{MnO}_3$ system, the anomalous behavior was revealed for the specimens with $z \sim 1$, i.e. for those, in which 50 % of La is substituted by Nd. The complex analysis of the structural, electric, magnetic and magnetoresistive properties allowed the authors to conclude that, contrary to the expectation of the homogeneous picture, stable near $z = 1$ is the structure with an inhomogeneous distribution of Nd atoms within the specimen volume. Such specimens consist of the areas enriched either by La or by Nd and are structurally and magnetically inhomogeneous. In this case, two peaks are characteristic of the temperature dependences of magnetic susceptibility, with positions of the peaks being close to the correspondent temperatures of the magnetic transitions in $\mathrm{Nd}_{2/3}\mathrm{Ca}_{1/3}\mathrm{MnO}_3$ (T_{Nd}) and $\mathrm{La}_{2/3}\mathrm{Ca}_{1/3}\mathrm{MnO}_3(T_{\mathrm{La}})$. $R(T)$ and $\mathrm{MR}(T)$ curves, however, display a single broad peak in the range between T_{Nd} and T_{La}. In our opinion, a correlation of the results obtained in the present work with those presented in [10] indicates that a similar inhomogeneous state is typical of $(\mathrm{La}_{1-y}\mathrm{Nd}_y)_{1-x}\mathrm{Sr}_x\mathrm{MnO}_3$, in particular, near $y = 0.25$. The reason for the appearance of such an inhomogeneity, as was concluded by the authors of [10], lies in a different character of lattice deformations near La^{3+} and Nd^{3+} ions and in an interaction of the correspondent fields of deformation between each other.

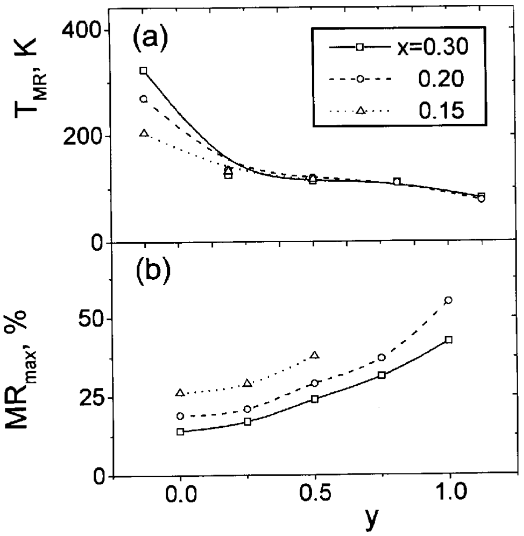

Fig. 5. Temperature of the MR maximum (*a*) and the maximal MR value (*b*) as a function of the Nd content

Thus, in the present work, the structural, electric, and magnetoresistive properties of specimens of the $(\mathrm{La}_{1-y}\mathrm{Nd}_y)_{1-x}\mathrm{Sr}_x\mathrm{MnO}_3$ system were studied in a wide range of the strontium ($x = 0.15 \div 0.3$) and neodymium ($y = 0 \div 1$) concentrations. It is shown that the dependence of the volume of the elementary cell on the neodymium concentration is close to a linear one in the whole range of y from 0 to 1. The main changes of the other structural characteristics, the oxygen coordinate x_{O} and the ratio between the parameters of the elementary cell c/a, occur in the range of $0 \leq y \leq 0.25$. A correlation has been established between the character of the change of structural parameters x_{O} and c/a, on the one hand, and that of the temperatures T_R та T_{MR} at which the maximum is observed on the $R(T)$ and $\mathrm{MR}(T)$ curves, on the other hand. The data are presented, which suggest that the structural and magnetic state is highly inhomogeneous in the specimens with y near 0.25.

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СТРУКТУРА ТА МАГНЕТОРЕЗИСТИВНІ ВЛАСТИВОСТІ ПОЛІКРИСТАЛІЧНИХ ЗРАЗКІВ $(\mathrm{La}_{1-y}\mathrm{Nd}_y)_{1-x}\mathrm{Sr}_x\mathrm{MnO}_3$

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Резюме

Досліджено структурні, електричні та магнеторезистивні властивості полікристалічних зразків $(\mathrm{La}_{1-y}\mathrm{Nd}_y)_{1-x}\mathrm{Sr}_x\mathrm{MnO}_3(y = 0 \div 1;\ x = 0,15 \div 0,3)$. Електричні вимірювання проводили у температурному діапазоні 77—370 К, магнетоопір вимірювали в полях до 15 кЕ. Показано, що заміщення лантану неодимом приводить до суттєвого підвищення магнетоопору і зниження температури, при якій магнетоопір має максимум. Виявлено, що основні зміни в положенні піків на температуратурних залежностях електро- та магнетоопору відбуваються в інтервалі $0 \leq y \leq 0{,}25$, що корелює з характером зміни структурних параметрів. Особливості поведінки зразків $(\mathrm{La}_{1-y}\mathrm{Nd}_y)_{1-x}\mathrm{Sr}_x\mathrm{MnO}_3$ поблизу $y = 0,25$ пов'язуються з утворенням структурно- і магнітно-неоднорідного стану.