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LETTER TO THE EDITOR

Peculiarities of magnetic, elastic and transport properties near the Curie temperature in $Nd_{1-x}Sr_xMnO_3$ manganites

A I Abramovich, A V Michurin, O Yu Gorbenko, A R Kaul
M V Lomonosov Moscow State University, Vorobyevy Gory, Moscow 119899, Russia

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Abstract. The close interplay between magnetic, transport and elastic properties in $Nd_{1-x}Sr_xMnO_3$ ($x = 0.33, 0.45$) compounds is found. Near the Curie point we have observed: (i) an abrupt minimum on the temperature dependence of negative volume magnetostriction ω (T), (ii) the large volume contraction ($\Delta V/V \approx 0.1\% for x = 0.33$ compound), (iii) a maximum on the temperature dependence of resistivity ρ (T), (iv) a maximum on the temperature dependence of absolute value of negative magnetoresistance. We have obtained large negative volume magnetostriction $\omega \approx -10^{-4}$ in a relatively low magnetic field $B = 0.9$ T and colossal negative magnetoresistance in the same magnetic field in the wide temperature region. These properties are explained by the existence of the conducting magnetic two-phase state in our compounds.

1. Introduction

At present the rare-earth manganites with perovskite structure are studied intensively. These materials have close interplay between electronic, magnetic subsystems and crystal lattice, resulting in anomalies of their magnetic, electric, optic and elastic properties. The most interesting effects from the theoretical, as well as the practical point of view, are colossal magnetoresistance (CMR) and giant magnetostriction (MS) observed in them near the Curie point T_C. Generally, CMR materials can be used as highly sensitive and electrically readable magnetic-field sensors for the read-head of the magnetic memory and compounds with large MS in devices, which convert electrical energy to mechanical energy. For this purpose it is necessary to stimulate a search for materials having CMR and very high MS at room temperature in the low magnetic field. There are many papers devoted to the study of CMR, while MS is explored little. Earlier, the giant negative volume MS $\omega \approx -5 \times 10^{-4}$ has been found by us for $Sm_{0.55}Sr_{0.45}MnO_3$ compound in a low magnetic field of 0.9 T. It is accompanied with negative CMR equal to 44% in the same magnetic field [1]. We present our results of the study of magnetization, ac initial magnetic and paramagnetic susceptibility, MS, thermal expansion (TE), resistance and magnetoresistance (MR) of $Nd_{1-x}Sr_xMnO_3$ ($x = 0.45; 0.33$) compounds.

2. Synthesis and experimental results

The ceramic samples were prepared as follows: ash-free paper filters were soaked with water solution of the metal nitrates with the concentration at approximately 1 mol/l, then the ashes formed by burning the dried filters were annealed at 973 K, the powder was pressed into pellets and sintered in air at 1473 K for 12 hours. The phase composition and lattice parameters were

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controlled by x-ray diffraction with Siemens D5000 diffractometer (Cu K_α radiation). The ceramics found are pure single-phase perovskite with the orthorhombic *Pnma* structure.

The magnetization measurements were performed by a vibrating magnetometer in a magnetic field up to 4 T; the initial magnetic susceptibility in an ac magnetic field with frequency from 0.8–8 kHz was measured with a F-5063 ferrometer; the paramagnetic susceptibility was measured using a balance (weighing) method with electromagnetic compensation. The resistance was measured by a four-probe method and contacts to the sample were made using a silver paste. The magnetostriction and thermal expansion were measured with strain gauges with resistance $92.30 \pm 0.01\Omega$ and tensosensitivity factor 2.26. One gauge was glued to the flat surface of the sample and the other gauge was glued to quartz. During the measurements the gauges were arranged identically on the sample and the quartz with respect to the direction of the magnetic field.

The measurements of the magnetization at 4.2 K showed that it is saturated under magnetic field 2 T. The spontaneous magnetic moment of the compound with $x = 0.45$ is equal to $3.50\mu_B$/mol which is close to value $3.55\mu_B$/mol corresponding ferromagnetic (FM) ordering Mn^{3+} and Mn^{4+} ions. The spontaneous magnetic moment of $x = 0.33$ compound is equal to $4.20\mu_B$/mol which is considerably more than the value of $3.67\mu_B$/mol corresponding FM ordering Mn ions only. Apparently, this difference is due to the magnetic moment of Nd^{3+} ions. As was shown by neutron diffraction method on $Nd_{0.7}Sr_{0.3}MnO_3$ compound, the Nd^{3+} magnetic moment reaches $0.8\mu_B$ and points in the same direction as the Mn magnetic moments [2]. The Curie temperatures T_C were estimated from measurements of the ac initial magnetic susceptibility in the magnetic field 10^{-4} T of 8 kHz frequency (figure 1). They were determined as the temperatures at which the $\partial\chi/\partial T(T)$ curves show a minimum and T_C is equal to 263 K and 242 K for $x = 0.45$ and 0.33 compounds respectively. The paramagnetic susceptibility obeys the Curie–Weiss law with paramagnetic Curie point $\theta = 331$ K and the effective magnetic moment $p_{eff.} = 5.63\mu_B$/mol for $x = 0.45$ and $\theta = 299$ K, $p_{eff.} = 5.16\mu_B$/mol for $x = 0.33$ compound. Figure 2 displays the temperature dependence of resistivity $\rho(T)$ and figure 3 displays the temperature dependence of MR $\Delta\rho/\rho = [\rho(H) - \rho(0)]/\rho(0)$ in the magnetic field of 0.9 T for both compounds. The compound with $x = 0.33$ has peak on $\rho(T)$ dependence near T_C, and resistivity of $x = 0.45$ compound shows the weak temperature dependence. The $\Delta\rho/\rho(T)$ dependences of both compounds exhibit a peak near T_C. We can see that MR shows the steady drop with increasing T, interrupted by an increase slightly near T_C. Two contributions in the MR are clearly seen from figure 3, one is the peak of MR near T_C and the other is the low-temperature MR, which is usually very small in single-crystals and epitaxial films of manganites, but reaches approximately 12% value at 80 K for our samples. The low-temperature MR is usually related with intergrain spin-polarized tunnelling [3] and spin-dependent scattering of polarized electrons at the grain boundaries [4], which usually coincide with domain walls. We assume that the first mechanism is predominant for our compounds because for manganites the strong p–d exchange takes place and they have the thick domain walls. So, the domain wall thickness is $10^3 a$ (where a is the lattice parameter) in accordance with the estimation made in [5]. Within such a wide domain wall the spins are turned gradually, and because of a strong p–d exchange, the charge carrier spin is arranged parallel with respect to spin of ion, on which it is located at the moment. In this case the charge carrier has not scattered. The MR peak near T_C seems to arise from MR contribution inside the grains. In such a manner for the compunds under investigation we obtained the CMR in the relatively low magnetic field and wide temperature region, which is important for practical device applications.

The longitudinal ($\lambda_\parallel$), transverse ($\lambda_\perp$) MS and TE were measured with strain gauges technique in the 80–300 K temperature region under magnetic field up to 1 T. For $x = 0.33$ compound the volume MS $\omega = \lambda_\parallel + 2\lambda_\perp$ changes sign at 160 K, it is positive below this

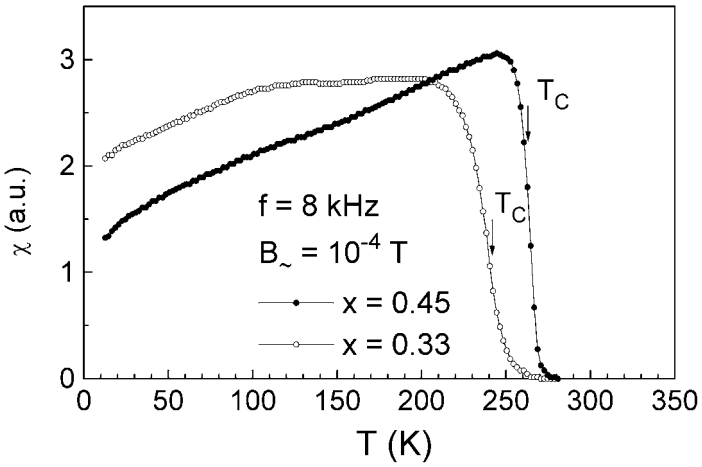

Figure 1. The temperature dependence of the ac initial magnetic susceptibility.

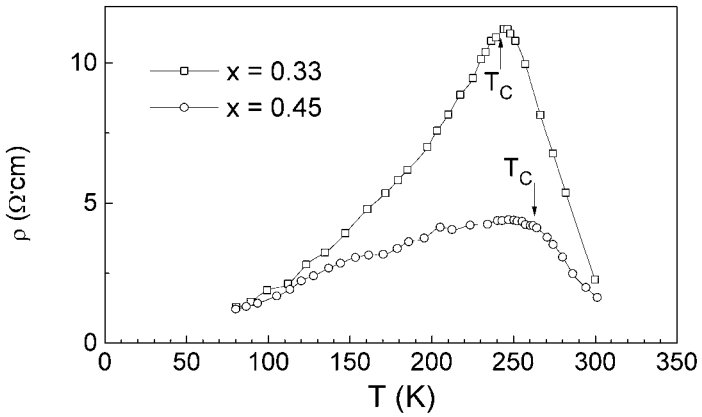

Figure 2. The temperature dependence of the resistivity.

temperature and negative above. The $\omega(T)$ dependence has an abrupt minimum at T_C, being $|\omega|$ is 7×10^{-5} at $B = 0.9$ T. The anisotropic MS $\lambda_t = \lambda_\parallel - \lambda_\perp$ is positive and equal to 3×10^{-5} at 80 K in the same magnetic field and it drops continuously to zero near T_C (figure 4). For $x = 0.45$ compound the volume and anisotropic MS behaviour is similar, but the MS value is less than for the $x = 0.33$ compound ($|\omega| = 1.5 \times 10^{-5}$ at $B = 0.9$ T near T_C). For both compounds the volume MS isotherms are not saturated in the magnetic field up to 1 T and no hysteresis are observed during increase and decrease of the magnetic field. The sharp change of TE is observed in the T_C region for $x = 0.33$ compound (the volume change $\Delta V/V = 3\Delta l/l \approx 0.1\%$), and the more smooth change for $x = 0.45$ compound (figure 5).

3. Discussion

There are different ideas concerning the nature of the CMR effect in manganites, such as a transition from jumping to polaron regime of conductivity near the Curie temperature T_C,

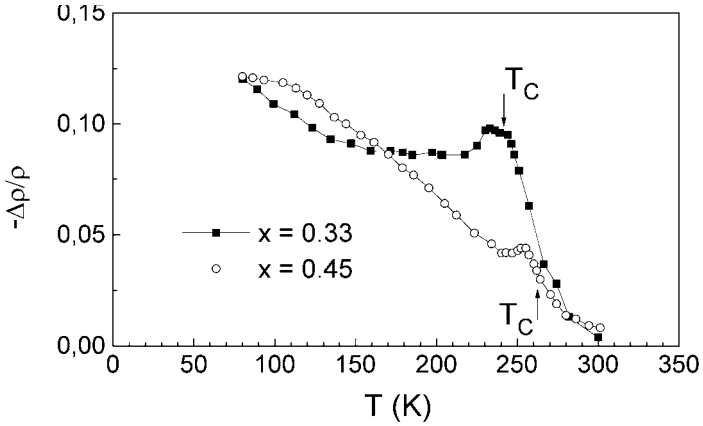

Figure 3. The temperature dependence of the MR.

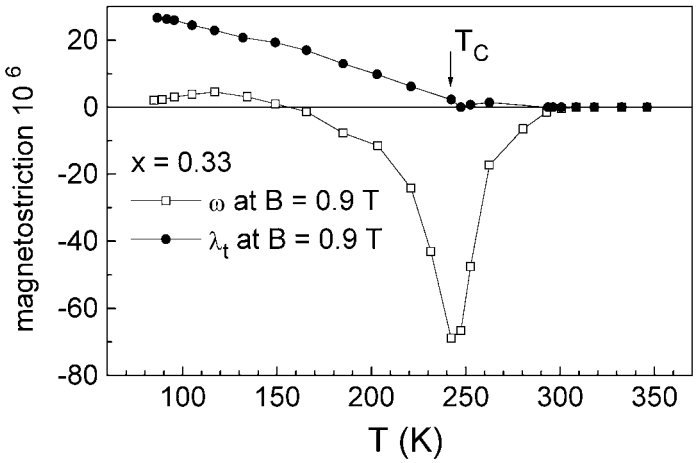

Figure 4. The temperature dependence of the volume and the anisotropic MS for $Nd_{0.67}Sr_{0.33}MnO_3$ compound.

melting of a charge ordered state under the magnetic field, decay of polarons under the magnetic field, etc. At present, more and more theoretical [6–8] and experimental [9,10] evidence appears for the existence of inhomogeneous states and electronic phase separation in manganites. One should note that the behaviour of ρ and CMR in manganites is similar to that in the usual magnetic semiconductors of EuSe and $CdCr_2Se_4$ type, where it was explained by the existence of a magnetic two-phase state (MTPS) [11]. It is well known that in magnetic semiconductors the charge carrier energy is minimal when the total ordering in the crystal is FM. For this reason, due to the gain in the s–d exchange energy, the electrons produce FM microregions (droplets) in an antiferromagnetic (AFM) semiconductor and stabilize these droplets by self-localization in them (insulating MTPS). As the impurity concentration increases, such FM droplets in an insulating AFM host increase in size and at a sufficiently high doping level they undergo percolation. In the process, another MTPS is formed: the insulating AFM droplets are located in the conducting FM host (conducting MTPS). So the $Nd_{1-x}Sr_xMnO_3$ compounds are a strongly doped AFM semiconductor $NdMnO_3$ in which, as

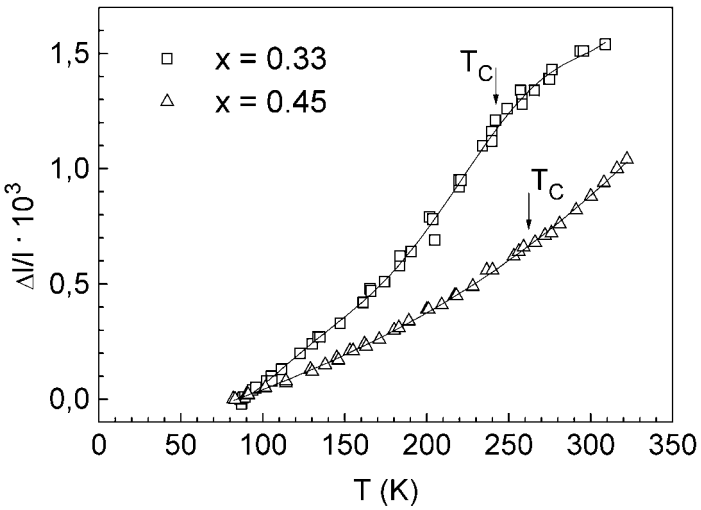

Figure 5. The temperature dependence of the TE.

we propose, at low temperatures MTPS is realized. It is confirmed by: (i) the spontaneous magnetization at 4.2 K is less then expected for full FM ordering (including magnetic moment of Nd^{3+} ions), (ii) the unusual low-temperature dependence of the χ for $x = 0.33$ compound (figure 1). We consider that conducting MTPS takes place in our compounds, because at $T < T_C$ are observed the metallic type of conductivity (figure 2). We have compared our results with those of [12]. The resistivity of the single crystal with $x = 0.45$ is $\backsim 10^{-4}\Omega$ cm at 4.2 K and $\backsim 10^{-3}\Omega$ cm in the T_C region [12]. This means that the exchange through charge carriers is predominant in this single-crystal. The T_C of our ceramic with $x = 0.45$ coincide with one of the same single-crystal [12]. This means that the exchange through charge carriers is predominant in the ceramic too, and the increase of their resistivity by three orders of magnitude in comparison with single crystal is due to intergrain boundaries. In the case of conducting MTPS there are two mechanisms of impurity-magnetic interaction influence on the resistance: (i) the scattering of charge carriers which reduces their mobility (ii) the formation of band tails, consisting of the localized states. The decrease of charge carrier mobility and their partial localization in band tails are most prominent in the T_C region. The MR is caused by the suppression of the impurity-magnetic scattering and band tails by the magnetic field [13].

At present there is no theory for the explanation of MS and TE of manganites. We propose that the MTPS is also the cause of the MS and TE anomalies. Yanase and Kasuya showed that the lattice parameters decrease in the FM part of crystal, since this results in the screening of the new charge distribution and lowers its energy by increasing the overlapping of the charge clouds of the central ion and its closest neighbours [14]. The lattice contraction in $La_{1-x}Ca_xMnO_3$ manganites near T_C was confirmed by the neutron diffraction study [15]. In the absence of the magnetic field the FM part of crystal breaks down thermally in the T_C region, and the excess TE of the sample that was observed in the present work takes place (figure 5). It is known that the imposition of an external magnetic field at $T \geqslant T_C$ increases the degree of FM order more strongly near the impurities than on the average over the crystal, since the effect of the field is intensified by the s–d exchange. That is, the magnetic field produces the FM part of the crystal, which were destroyed by heating, and a corresponding compression of

the lattice. However, the process of field-induced production of the FM clusters occurs in a limited temperature range not far above T_C. For this reason, the curves $\omega(T)$ pass through a minimum and $|\omega|$ drop rapidly as the temperature increases further (figure 4).

Acknowledgements

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