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Magnetic and resonance properties of the $Y_{0.5}Sr_{0.5}Cr_{0.5}Mn_{0.5}O_3$ polycrystal

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Abstract. The magnetostatic and magnetic resonance properties of the $Y_{0.5}Sr_{0.5}Cr_{0.5}Mn_{0.5}O_3$ polycrystalline system have been experimentally studied. The intracrystalline ferromagnetic interaction turned out to be prevalent while the intercrystalline interaction appears to have antiferromagnetic character. We found that two absorption lines are observed in the spectrum in the magnetic ordering region at $T < 80$ K. The high-field line corresponds to the interacting parts of polycrystal related to the disordered shells and the low-field peak is system of ferromagnetic particles.

1. Introduction

The compounds belonging to the complex oxide materials are intensively investigated and attractive for application because of their chemical inertness and stability. Inhomogeneous media with the expressed interfaces between mesoscopic structural elements are of great interest, e.g. systems with phase separation, film structures, and nanoscale composites [1]. In this direction, the manganite compounds with the general formula $A_xB_{1-x}Mn_yMe_{1-y}O_3$ (where A is the rare-earth element, B is, as a rule, the alkaline-earth element, and Me is the 3d-metal) have been studied most thoroughly [2]. The basic compound is $LaMnO_3$. In particular case, doping of this compound with Sr^{2+} ions lead to a rich variety of properties and multiplicity of transitions between different magnetic phases, depending on the impurity concentration. Generally, the magnetic and electrical properties of such systems are determined by the ratio between manganese ions Mn^{3+}/Mn^{4+}. On the other hand, in mixed chromite manganites, the effect of exchange bias arises due to the competition of exchange interactions between similarly charged and oppositely charged Cr and Mn ions with different valences [3-5]. Introducing yttrium impurities instead of lanthanum may result in the anomalous change in the relative volume, the change in the inverse susceptibility, and the relaxation magnetization processes typical for spin glasses are also observed. The foregoing demonstrates a noticeable effect of yttrium ions on the magnetic properties of manganite crystals. Upon transition to the nanoscale, the spatial inhomogeneity inside particles arises and the interparticle interactions are added. The properties of yttrium-based manganites have received little attention in literature, and introducing chromium ions instead of manganese ions expands the variety of possible exchange interactions.

2. Material and methods

A mixture of oxides Y_2O_3, $SrCO_3$, Cr_2O_3, and Mn_2O_3 was used to obtain the $Y_{0.5}Sr_{0.5}Cr_{0.5}Mn_{0.5}O_3$ manganate. The obtained samples of complex oxides were prepared by the sol-gel method [6]. The phase state of the final products was monitored by X-ray phase analysis with a Miniflex 600 X-ray diffractometer (Rigaku). Electron microscopic measurements were performed by a JEOL JEM-2100 microscope. Magnetic characteristics were studied on the MPMS-XL SQUID-magnetometer in fields up to 50 kOe. Electron magnetic resonance (EMR) spectra were measured by means of a EPR spectrometer ELEXSYS E580, Bruker, Germany operating at a frequency of ω_{MWF} = 9.48 GHz. Magnetic resonance measurements were performed in the temperature range from 5 K to 300 K.

3. Experimental

The X-ray spectroscopy method showed that the crystals correspond to the nominal composition and belong to the orthorhombic syngony with the cell parameters a = 0.7065 nm, b = 0.7375 nm, and c = 0.6741 nm. The density of the substance was $p \approx 3.95$ g / cm3. The average size of the crystallites which can be derived from the electron microscopic image is nearly $L \geq 10$ μm. Local values of the chemical elements content obtained by transmission electron microscopy from spots with a diameter of ~ 20 nm in different places of the sample, both in atomic content and in weight, correspond to the nominal values with an accuracy of 5-6 %.

3.1. Magnetic properties

Previously [7] the magnetostatic measurements showed there are symmetric hysteresis loops of magnetization at low temperatures. Below 30 K a loop of hysteresis is open, but the saturation of magnetization is not reached down to the fields of $H \geq 50$ kOe. At the temperatures $T > 35$ K a magnetization curve is anhysteretic, and at temperatures $T > 75$ K dependence of $M(H)$ becomes linear. The inverse susceptibility ($1/\chi$) (figure 1) was obtained at different magnetic fields. For a standard model of two-sublattice antiferromagnet [8], we have $\chi = C/(T - \Theta)$, where Θ corresponds to the point of intersection of the asymptote with the horizontal axis and depends on the ratio between the "intra"- and "interlattice" exchanges. In addition, the inset shows the qualitative $1/\chi$ behavior for an antiferromagnet at different magnetic field directions relative to the antiferromagnetic axis.

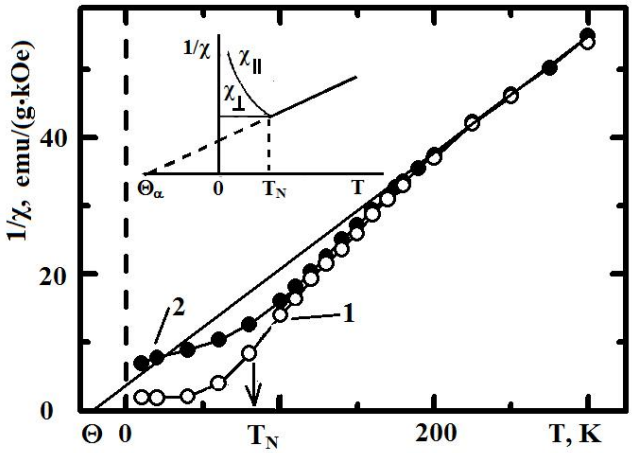

Figure 1. Temperature dependences of the inverse susceptibility at (*1*) 2.5 and (*2*) 40 kOe. Inset: model dependence for $1/\chi(T)$ from [8].

It can be seen that the experimental $1/\chi(T)$ dependence qualitatively corresponds to the antiferromagnetic ordering of a polycrystal. At a measuring field H = 2.5 kOe, the $1/\chi(T)$ curve bending becomes pronounced in the range of $T_N \approx 75$–90 K, which can be related to the Neel temperature. For all measuring fields, we have Θ = –27 K, while the temperature T_N depends on the measuring field.

The qualitative results can be explained within the model of a two-sublattice antiferromagnet. If $J_1 > 0$ and $J_2 < 0$ are the intra- and interlattice exchange interactions, then, according to [8], we have $\Theta = J_1 + J_2$ and $T_N = J_1 - J_2$. Using the experimental Θ and T_N values, we obtain $J_1 \approx +29$ K and

$J_2 \approx -56$ K. That is the magnetic measurements data show that individual ferromagnetic crystallites in policrystal are bound together by an antiferromagnetic interaction through shell (at $T < T_N$). Note that in the phase diagram for the $La_{1-x}Sr_xMnO_3$ system, the substitution concentration x = 0.5 is boundary for the existence of ferromagnetic or antiferromagnetic phases [2].

3.2. Magnetic resonance

When measuring the resonant absorption, we found that at low temperatures the spectrum consists of two lines: 1 (low-field) and 2 (high-field) (see figure 2, part a). The resonant field of a high-field line is practically independent of temperature; the intensity of this line (defined as the area under the curve and proportional to the magnetization) decreases in magnitude and the line disappears at $T \approx 80$ K. This temperature coincides with the temperature identified as the temperature (T_N) of the magnetic ordering in the sample during magnetostatic measurements. However, as seen in figure 2a, this line has a relatively narrow width. Line 1 is wide and is observed over the entire temperature range studied. These data implies that the high-field peak originates from the sample part that is coupled with the polycrystal shell and, when the long range order vanishes, this part cannot be seen against the low-field peak background. Thus, the low-frequency absorption is related to the resonance from a disordered system of ferromagnetic clusters (crystallites). As the temperature rises, the line 2 disappears, a set of clearly seen lines arises near its location but with an extremely low intensity.

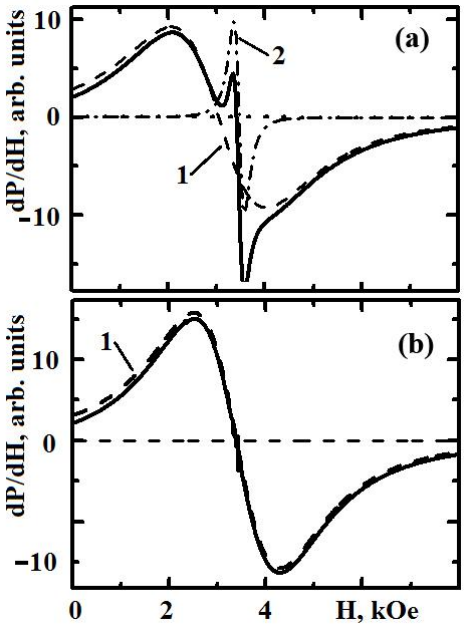

Figure 2. The resonance absorption spectrum of the $Y_{0.5}Sr_{0.5}Cr_{0.5}Mn_{0.5}O_3$ polycrystal at temperatures: (a) – T = 39 K, (b) – T = 270 K. 1, 2 – Lorentzian curve, solid line – experimental curve.

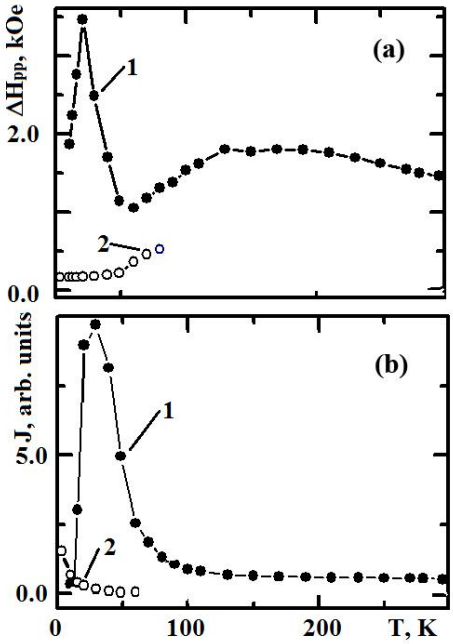

Figure 3. The temperature dependences: (a) – linewidths of resonant curves, (b) – line intensities. The numbers of the lines correspond to the lines in figure 2.

The resonant field of line 1 increases in magnitude from a value of $H_r \approx 1.5$ kOe (T = 4.5 K) to a value of $H_r \approx 3.4$ kOe (90 K). At temperatures $T > 90$ K, the resonant field of line 1 is practically independent on temperature, which indicates the paramagnetic behavior of the system. The behavior of the intensities (J) of absorption lines (figure 3, part b) at high temperatures ($T > 90$ K) is similar to the behavior of resonant fields. In the temperature range of the existence of line 2 ($T < 80$ K), its intensity decreases with increasing temperature, and the intensity of the main resonance (line 1), passing through the maximum at low temperatures, also becomes temperature-independent in the high-temperature range.

The resonant absorption linewidths (ΔH) show a more complex behavior. The width of line 2 increases slightly as it approaches the temperature T_N. For line 1, ΔH has two distinct maxima. One of them is narrower and more intense in the magnetic ordering region and another is wide in the paramagnetic region.

4. Discussion

The features of the width of line 1 in figure 3a do not correspond to the behavior inherent in the ensemble of superparamagnetic particles, where a significant widening of the absorption line at low temperatures and its narrowing at temperatures above the blocking temperature is typical. In our case, the shape of the temperature dependence ΔH is qualitatively similar to that observed, for example, in the $Mn_xFe_y0_4$ crystals [9], where the behavior of the linewidth is described by ion relaxation processes [10].

As known from early studies [9], the low-temperature ($T \sim$ 15-20 K) maximum of the linewidth in magnetically ordered manganese-containing ferrites is associated with the ionic relaxation of the Mn^{3+} ions. This ion has a $3d^4$ configuration and a ^5D electron term with L = 2 and S=2. In the crystal field of octahedral symmetry, the D-term splits into a doublet and a low-lying triplet [11], which further splits into singlets due to uniaxial distortions and the strong Jahn-Teller interaction. This ion can be considered as a rapidly relaxing ion making the main contribution to the linewidth at low temperatures.

If we pay attention to the high-temperature maximum of the linewidth, the possible reason of its existence may be the contribution from the Cr^{4+} ion subsystem. This ion has a $3d^2$ configuration and a ^3F electron term with L = 3 and S=1. In the crystal field of octahedral symmetry, this term splits into a singlet and two low-lying triplets. With a further decrease in symmetry, the main one is the orbital singlet, which has a three-fold spin degeneracy, and the remaining orbital levels are high-lying and do not significantly affect the state of the main singlet. The spin degeneracy is removed by the spin-orbit interaction and the low symmetric contribution of the crystal field, which results in the quasi-Ising behavior of the Cr^{4+} ion [11].

The results obtained from magnetic resonance studies are in the satisfactory agreement with the data of magnetostatic measurements. The sample is a set of crystallites interacting with each other at the interface. At the same time, comparing the temperature dependences of the intensities of the resonant absorption lines 1 and 2 in the region close to the transition ($T_N \approx 80$ K), we can estimate the volume of the core and the outer shell. Since the intensity of the absorption line is proportional to the volume of the magnetic phase, $J_2/J_1 \sim (r^3 - R^3)/R^3$, where r is the outer radius of the granule (crystallite) and R is the radius of the core. The experiment shows that this ratio is $\leq 10^{-2}$. To estimate the core and the outer shell radiuses, let's assume that the granules have a spherical shape, then $r \approx 5 \cdot 10^3$ nm and the shell thickness is $r - R \approx 20$ nm.

Analyzing the behavior of the magnetic resonance linewidth, we can conclude that the low-temperature maximum for line 1 corresponds to the magnetic ordering region in the crystallite, while the high-temperature maximum corresponds to the paramagnetic region. More detailed information on relaxation mechanisms will be provided elsewhere.

Acknowledgements

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