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Giant volume magnetostriction and colossal magnetoresistance at room temperature in $La_{0.7}Ba_{0.3}MnO_3$

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Abstract

Giant volume magnetostriction at room temperature is found for the first time in a $La_{0.7}Ba_{0.3}MnO_3$ single crystal, achieving 2.54×10^{-4} in a magnetic field of 8.2 kOe. An even greater value of the volume magnetostriction, equal to 4×10^{-4} in the same magnetic field, is observed at the Curie point $T_C = 310$ K. Volume magnetostriction and magnetoresistance exhibit similar dependences on temperature and magnetic field in the T_C-region, that is explained by the presence in this compound of a magnetic two-phase ferromagnetic–antiferromagnetic state due to strong s–d exchange.

The interest in the perovskite Mn oxides is associated with colossal magnetoresistance (CMR), that was observed in certain compounds near room temperature. This effect makes it possible to use them in various sensor devices. The magnetostriction of manganites is less well understood [1–6]. In works [1, 2] an anomalous thermal expansion contribution in the temperature interval from the Curie temperature T_C to 1.8 T_C in two compounds, $(La_{1-x}A_x)_{2/3}Ca_{1/3}MnO_3$ (A = Y or Tb), was explained by the polaronic lattice effect provided by a narrow polaronic band ('small polaron', that is strong electron–phonon interaction), along with the ferromagnetic character of clusters. Under an applied magnetic field the polaronic band is expanded, that is responsible for a volume magnetostriction and CMR. However, how the transition to the double exchange at T_C occurred was not clear. An additional point to emphasize is that strong electron–phonon interaction is favoured by antiferromagnetic (AF) pairing of the electron spins, that is observed in superconductors. In works [3–5] the giant volume magnetostriction, exceeding 10^{-4}, in a magnetic field of 8.5 kOe, was found in single-crystal $La_{1-x}Sr_xMnO_3$ ($0.1 \leqslant x \leqslant 0.3$) and $La_{0.8}Ba_{0.2}MnO_3$ near the Curie point T_C. Volume magnetostriction and magnetoresistance exhibit similar dependences on temperature and magnetic field in the T_C-region, that is explained by the presence in these compound of a magnetic two-phase ferro-antiferromagnetic state due to strong s–d exchange. The possibility of the presence of that magnetic two-phase state (MTPS) in the system $La_{1-x}Sr_xMnO_3$ was

confirmed by our investigations: in the compound $La_{0.9}Sr_{0.1}MnO_3$ we found a giant red shift of the absorption edge ∼0.4 eV, connected with ferromagnetic (F) order [7]. This means that the energy of the charge carriers (holes in the present case) decreases with increasing degree of F order, and for this reason it was energetically favourable for holes to become localized with not too high density near impurities, maintaining F order around them. Here their localization, besides a gain in s–d exchange energy, gave rise to Coulomb attraction of holes to acceptors. These F microregions or droplets were named ferrons [8–10], giant quasimolecules [11], magnetic polarons, etc. Nagaev showed that the radii of F droplets rise in an applied magnetic field [8]. On increase in the carrier density, the radii of F droplets begin to make contact with each other. Thus, percolation of the electron liquid occurs and another conductive MTPS is formed: the insulating AF microregions are set inside the conducting F host.

The $Re^{3+}_{1-x}A^{2+}_xMnO_3$ (Re = La and rare earth ions) composition represents the antiferromagnet $ReMnO_3$, doped by A^{2+} ions, which are acceptors. In works of Nagaev [8–10] and Dagotto with coauthors [12] CMR is connected with the presence in manganites of MTPS due to a strong s–d or d–d exchange (strong Hund coupling). CMR near the Curie point T_C for insulating MTPS is explained by the rise of F droplet radii and the alignment of their magnetic moments in the magnetic field that facilitates the hole tunnelling between FM droplets. In the end the field tends to destroy the FM droplets [9, 10]. The sharp increase of ρ in the T_C-region is characteristic of a conducting MTPS [8–10]. There are two mechanisms through which the impurity–magnetic interaction influences ρ: (i) the scattering of charge carriers which reduces their mobility; (ii) the formation of band tails, consisting of the localized states. As shown in [13], the decreases of the charge carrier mobility and their partial localization in the band tails are most prominent in the T_C-region. This is the cause of the ρ-maximum near the Curie point. The magnetic field increases the charge carrier mobility and excites the charge carriers from the band tails, that is the cause of CMR.

Yanase and Kasuya showed [11] that inside the F part of the crystal the lattice constants are reduced. The reason is that in the F part of the crystal the spacing between an impurity ion and its nearest magnetic ions is shortened to screen the new charge distribution and to lower the energy of the F part of the crystal by increasing the overlap between the valence electron shells of the impurity and the d-shells of the nearest magnetic ions. Hence it follows that near T_C a surplus thermal expansion may be observed, which is connected with the MTPS destruction. A switching on of an external magnetic field at $T \geqslant T_C$ would lead to a stronger increase of F ordering in the vicinity of impurities than on average over the crystal because its action is enhanced by s–d exchange. In other words, a magnetic field would reconstruct the MTPS, destroyed by heating, and the lattice compression, inherent in it. A sharp increase in the negative volume magnetostriction in the T_C-region could be observed. Here the case in point is the volume magnetostriction that is defined by the exchange interactions. However, the abovementioned process of MTPS reconstruction by a field can take place only in a limited temperature interval at $T \geqslant T_C$. Because of this the $|\omega|(T)$ curves could have a sharp maximum in the T_C-region and would quickly fall off with further increase in temperature.

At present an ever increasing number of scientists connect CMR in manganites with the existence in them of magnetic inhomogeneity (see the review [12] and monographs [14, 15]). As early as 1955 Wollan and Koehler found F and AF phases in $La_{1-x}Ca_xMnO_3$ ($0.1 \leqslant x \leqslant 0.3$) by neutron diffraction methods; the volumes of these phases depended upon the relative trivalent and tetravalent manganese ion content [16].

In our paper an attempt was made to reveal the giant volume magnetostriction and its connection with CMR near T_C in a $La_{0.7}Ba_{0.3}MnO_3$ single crystal leaning upon the arguments given above concerning MTPS due to strong s–d exchange. Recently, that connection was found in a $La_{0.8}Ba_{0.2}MnO_3$ single crystal [5] but giant volume magnetostriction and CMR were

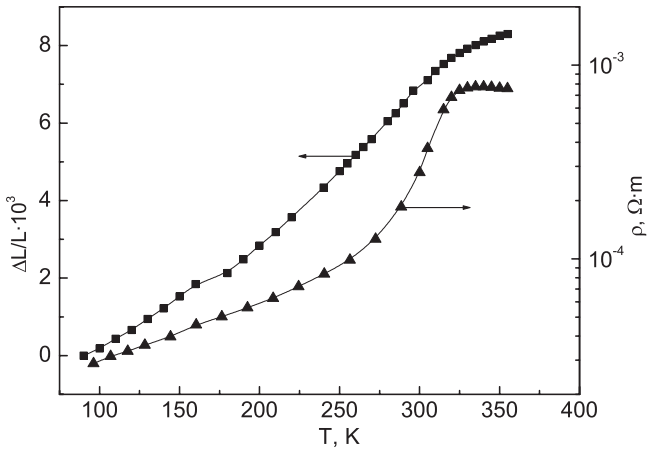

Figure 1. The temperature dependence of the thermal expansion $\Delta l/l$, and the electrical resistivity ρ, received on heating of the crystal.

observed below room temperature. In the $La_{0.7}Ba_{0.3}MnO_3$ single crystal $T_C = 310$ K. This promises well that giant volume magnetostriction and CMR will take place in $La_{0.7}Ba_{0.3}MnO_3$ at room temperature. That discovery would make it possible to fabricate magnetomechanical devices possessing CMR.

In this work the thermal expansion $\Delta l/l$, magnetostriction λ, electrical resistivity ρ and magnetoresistance $\Delta\rho/\rho = (\rho_H - \rho_{H=0})/\rho_{H=0}$ of single-crystal $La_{0.7}Ba_{0.3}MnO_3$ were studied. The single crystal of this composition was made by the floating zone technique. The details of preparation have been described in [17]. Parallel ($\lambda_\parallel$) and perpendicular ($\lambda_\perp$) magnetostriction to an applied magnetic field H and thermal expansion $\Delta l/l$ were studied by the use of the strain gauge technique. A strain gauge was stuck to the plane parallel to the direction of crystal growth. This differed from the [100] axis by 4°. The accuracy of the $\Delta l/l$ measurements was better than 4×10^{-6}. The standard four-point method was used for measurement of ρ and $\Delta\rho/\rho$. Contacts on the samples were prepared with a silver paste. The Curie temperature was determined from a curve of the T-dependence of a magnetization in $H = 470$ Oe by extrapolation of the steepest part of this curve until intersection with the T-axis. The T_C-value determined in this way was equal to 310 K.

Figure 1 shows the temperature dependence of linear thermal expansion and in figure 2 are shown the $\lambda_\parallel(T)$ and $\lambda_\perp(T)$ curves in the magnetic field 8.2 kOe. From the experimental $\lambda_\parallel(H)$ and $\lambda_\perp(H)$ curves the isotherms of anisotropic magnetostriction $\lambda_t = \lambda_\parallel - \lambda_\perp$ and volume magnetostriction $\omega = \lambda_\parallel + 2\lambda_\perp$ were constructed. The isotherms of volume magnetostriction near the Curie point are shown in figure 3(a) and figure 4(a) demonstrates the temperature dependence of ω in such a temperature interval in certain values of magnetic field. On increase in temperature, the anisotropic magnetostriction reduces continuously down to zero in the T_C-region. As illustrated in figures 3(a) and 4(a), the volume magnetostriction is negative and its magnitude reaches a maximum near $T_C = 310$ K. On further heating $|\omega|$ vanishes rapidly. The maximum value of $|\omega| \approx 4 \times 10^{-4}$ at $H = 8.2$ kOe. In the same field at room temperature (300 K) $|\omega| = 2.54 \times 10^{-4}$. This is the giant value of volume magnetostriction that is important for a practical application. It should be mentioned that in the maximum field of measurement, equal to 8.2 kOe, the $\omega(H)$ curve is far from saturation as shown in figure 3(a).

Figures 1 and 4(b) show the temperature dependence of ρ and $\Delta\rho/\rho$ correspondingly and figure 3(b) the $\Delta\rho/\rho(H)$ curves in the T_C-region. From comparison of figures 3(a)

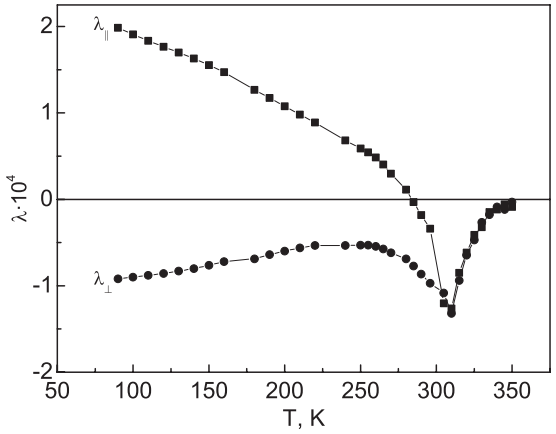

Figure 2. The temperature dependence of the parallel ($\lambda_{\parallel}$) and perpendicular ($\lambda_{\perp}$) magnetostriction to an applied magnetic field $H = 8.2$ kOe.

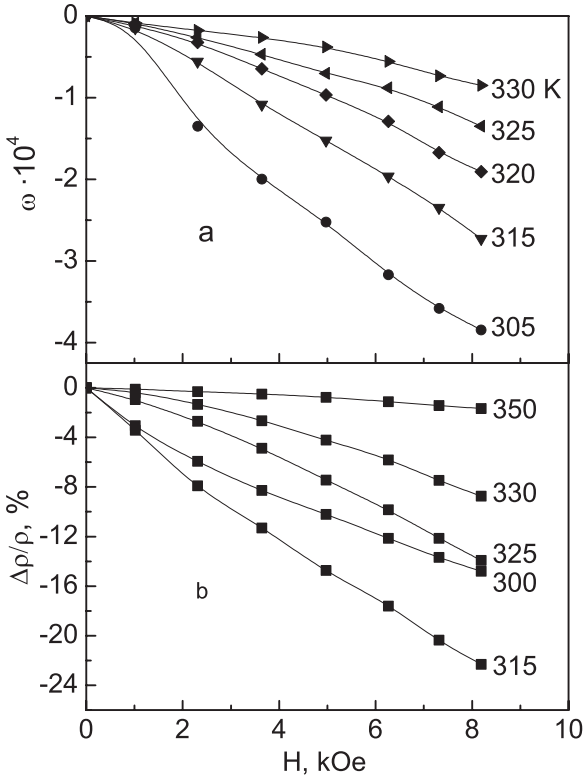

Figure 3. The isotherms of the volume magnetostriction ω (a) and the magnetoresistance $\Delta\rho/\rho$ (b) at the selected temperatures near the Curie point.

and (b), 4(a) and (b), it will be noted that the behaviour of ω and $\Delta\rho/\rho$ is similar near T_C. So, ω and $\Delta\rho/\rho$ are negative, their isotherms are not saturated up to the maximum field of measurement 8.2 kOe, the curves of $|\omega|(T)$ and $|\Delta\rho/\rho|(T)$ have maxima and at the maximum

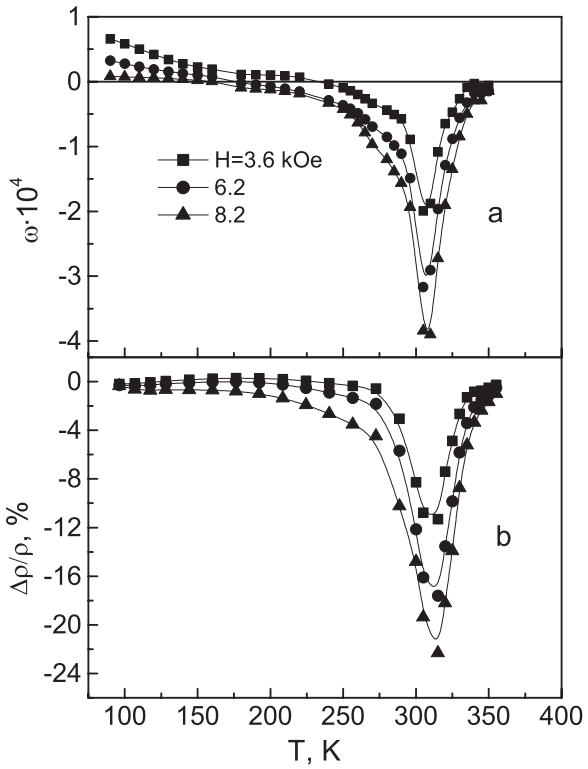

Figure 4. The temperature dependence of the volume magnetostriction ω (a) and magnetoresistance $\Delta\rho/\rho$ (b) in the selected magnetic field.

$|\omega|$ and $|\Delta\rho/\rho|$ achieve giant values of 4×10^{-4} and 22.7% correspondingly. Because of this it may be supposed that the peculiarities of ω and $\Delta\rho/\rho$ near T_C in $La_{0.7}Ba_{0.3}MnO_3$ are explained as well as in $La_{1-x}Sr_xMnO_3$, namely, by the presence in the sample of a magnetic two-phase ferromagnetic–antiferromagnetic state due to strong s–d exchange [8–10, 12]. However, in support of this conjecture it was necessary to receive proof of the presence of strong s–d exchange in the $La_{1-x}Ba_xMnO_3$ system. For this purpose we plan to study the temperature shift of edge of optical intrinsic absorption of this system. Revealing a strong red shift would show the presence of strong s–d exchange.

One further comment should be made. In MTPS theory the relation between T_C and the Neel temperature T_N is still elusive. Experiments exhibited in [12, 14, 15] frequently show that in MTPS manganites near the Curie point the AF state is substituted by the paramagnetic state. Such an MTPS was discussed in [18–20]. It is currently unknown which one of the two MTPSs listed above occurs near T_C in $La_{0.7}Ba_{0.3}MnO_3$.

Figure 1 shows that the $\rho(T)$ dependence has a metallic type of conductance at $T < T_C$ and a maximum near T_C. This behaviour is characteristic for a conductive MTPS. In the Curie point region the MTPS would be thermally destroyed, causing sample expansion. As may be seen from figure 1, at $T \ll T_C \Delta l/l$ increases linearly with T, that is caused by the phonon anharmonic contribution. The slope of the $\{\Delta l/l\}(T)$ line is considerably extended in the interval 260 K $\leqslant T \leqslant$ 315 K. It is felt that the increasing slope in this interval is connected with an extra contribution to thermal expansion due to the MTPS destruction. At $T > 315$ K this increase slows down. Evidently, the destruction of the MTPS is completed in this interval.

The peculiarity of the $\{\Delta l/l\}(T)$ curve in the interval 165 K $\leqslant T \leqslant$ 181 K may be attributed to a structural transition from Pbmn to R$\bar{3}$c phase.

By this means the data of this paper count in favour of the hypothesis that the giant volume magnetostriction is universally presented in CMR manganites since the volume magnetostriction and CMR are offered by the presence of the MTPS in the sample. We have undertaken the study of the magnetostriction and the magnetoresistance in $Nd_{0.6}Ba_{0.4}MnO_3$ and $La_{0.8}Ca_{0.2}CoO_3$ ceramics too. In these compounds magnetostriction and magnetoresistance are not found within the limits of experimental error, that is evidence of the hypothesis mentioned above.

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