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Magnetoelastic Properties of $La_{0.744}Ba_{0.186}MnO_3$ Single Crystals

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Magnetoelastic properties of $La_{0.744}Ba_{0.186}MnO_3$ single crystals have been studied. The basic magnetoelastic constants of the crystal have been estimated using the data of magnetostriction and magnetization measurements, which have been carried out in [010] and [110] crystallographic directions in a wide temperature range from 10 K to 150 K. The anomaly of the thermal expansion has been observed at about 185 K. Near this anomaly two almost degenerated phase transitions have been observed. One of these transitions may be related to the structural phase transition, while the second one to the occurrence of spontaneous strains. The magnetostriction of $La_{0.744}Ba_{0.186}MnO_3$ single crystal seems to arise due to the effect of applied field on the spontaneous strains.

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1. Introduction

The intensive investigations during the last years of the perovskite Mn oxides, $Re_{1-x}A_xMnO_3$ (Re = La, Y, Nd, Pr, Er, A = Ca, Sr, Ba, Pb), lead to the discovery of various interesting effects connected with magnetoelastic properties of these materials [1]. The large magnetostrictive effect in manganites has been discovered first by Ibarra et al. [2]. The anomalies of the longitudinal magnetostriction behavior allowed us to observe a field-induced structural phase transition in $La_{0.9}Sr_{0.1}MnO_3$ [3]. The existence of magnetic polarons above the Curie temperature in $La_{2/3}Ca_{1/3}MnO_3$ perovskites has been shown by means of magnetostriction and thermal expansion measurements [4]. Another interesting aspect of the role of magnetoelastic interactions in manganites was a discovery of the anomalous

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acoustoelectric effect in $La_{0.67}Ca_{0.33}MnO_3$ films [5], which seems to be connected directly with the magnetoelastic effect. The magnetoelastic effects are also responsible for technical parameters of magnetic and mechanical sensors. All these facts stimulate a great deal of interest in studying the magnetoelastic properties of doped manganites.

In this work, we investigate for the first time the magnetization M, magnetostriction λ, and thermal expansion $\Delta l/l$ of the $La_{0.744}Ba_{0.186}MnO_3$ single crystals at $10 \leq T \leq 150$ K. This compound was chosen because of the lack of data on magnetostriction in Ba-doped $LaMnO_3$ single crystals. The compound studied is a ferromagnet with the Curie temperature of about 251 K [6] and with a structural phase transition from a rhombohedral to orthorhombic structure at $T_s = 185$ K. The structural phase transition was shown to be of the first order [6].

2. Experimental details

The $La_{0.744}Ba_{0.186}MnO_3$ single crystals were grown using the floating zone technique. The details of preparation have been described elsewhere [7]. It possesses a rhombohedral structure (the space group $R\overline{3}c$). The X-ray diffraction analysis at 296 K has allowed to determine the lattice parameters: $a = b = 5.558$ Å, $c = 13.462$ Å. These values are in good agreement with the corresponding values known from literature (for similar composition) [8]. For magnetostriction and thermal expansion measurements the sample of $2.0 \times 2.15 \times 2.5$ mm^3 was chosen with edges along the [001], [010], and [100] directions (in the pseudo-cubic cell approximation) and practically free of twins.

The magnetization measurements were carried out using a SQUID magnetometer (MPMS 5, Quantum Design). The three-terminal capacitance technique was applied in order to measure magnetostriction and thermal expansion. Magnetostriction measurements were performed as a function of temperature in external magnetic field with intensity up to 120 kOe. The accuracy of the λ measurements was better than 0.5×10^{-6}.

3. Experimental results and analysis

Figure 1 presents the results of the longitudinal magnetostriction measurements along the [010] and [110] crystallographic directions at 50 K. It can be concluded that magnetostriction in the ferromagnetic (FM) phase of $La_{0.744}Ba_{0.186}MnO_3$ is anisotropic and it has the magnitude and magnetic field dependence being characteristic of ferromagnetic $3d$ metals. It has to be mentioned that both magnetostriction and magnetization have similar dependences on the applied field (Fig. 2). In both cases the saturation field has practically the same value (about 3.5 kOe).

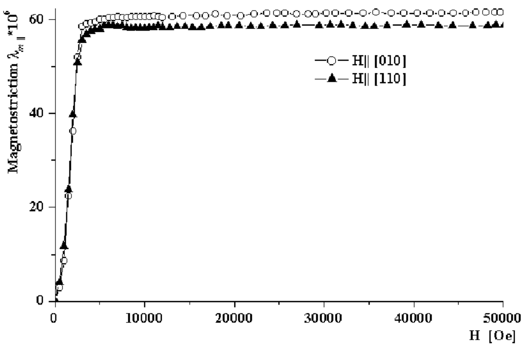

Fig. 1. The longitudinal magnetostriction of the $(La_{0.8}Ba_{0.2})_{0.93}MnO_3$ single crystal measured along [010], [110] crystallographic directions at 50 K.

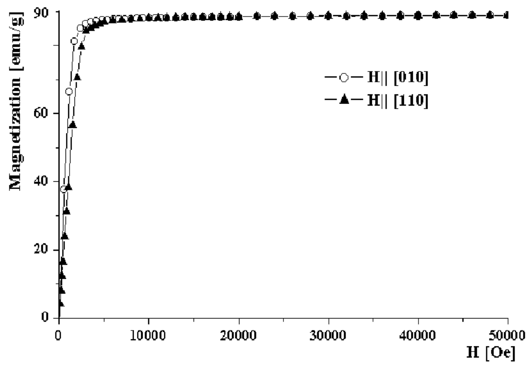

Fig. 2. The magnetization of single crystalline sample as a function of magnetic field applied along [010] and [110] crystallographic axes at a temperature of 50 K.

In order to determine the magnetostriction constants we have used the following expression for magnetostriction in a single crystal of the orthorhombic structure [9]:

$$\begin{aligned}
\lambda = {} & \tfrac{1}{3}\lambda_1^{\alpha,0} + \lambda_2^{\alpha,0}(\beta_3^2 - \tfrac{1}{3}) + \lambda_3^{\alpha,0}(\beta_1^2 - \beta_2^2) \\
& + \left[\tfrac{1}{3}\lambda_1^{\alpha,2} + \lambda_2^{\alpha,2}(\beta_3^2 - \tfrac{1}{3}) + \lambda_3^{\alpha,2}(\beta_1^2 - \beta_2^2)\right]\left(\alpha_3^2 - \frac{\alpha_1^2 + \alpha_2^2}{2}\right) \\
& + \left[\tfrac{1}{3}\lambda_1^{\alpha,2'} + \lambda_2^{\alpha,2'}(\beta_3^2 - \tfrac{1}{3}) + \lambda_3^{\alpha,2'}(\beta_1^2 - \beta_2^2)\right]\left(\frac{\alpha_1^2 - \alpha_2^2}{2}\right) \\
& + 2\lambda^{\beta,2}\beta_1\beta_2\alpha_1\alpha_2 + 2\lambda^{\gamma,2}\beta_2\beta_3\alpha_2\alpha_3 + 2\lambda^{\delta,2}\beta_1\beta_3\alpha_1\alpha_3, \qquad (1)
\end{aligned}$$

where α_1, α_2 α_3 — the direction cosines of magnetization and β_1, β_2, β_3 — the direction cosines of the elongation observed. In our experiment the measured magnetostriction λ_r determines the changes of sample dimension during the magnetization process starting from the fully demagnetized state

$$\lambda_{\mathrm{r}} = \lambda_{\mathrm{m}} - \lambda_{\mathrm{sp}}, \tag{2}$$

where λ_{m} — the magnetostriction induced by the changes of the magnetization in external magnetic field, λ_{sp} — spontaneous magnetostriction as the consequence of the magnetic ordering below T_{C}.

Finally we obtained

— for the magnetostriction parallel to the [010] direction:

$$\lambda_{\mathrm{r}010} = \left[\left(\frac{\lambda_1^{\alpha,2}}{6} - \frac{\lambda_2^{\alpha,2}}{6} - \frac{\lambda_3^{\alpha,2}}{2}\right) + \left(\frac{\lambda_1^{\alpha,2'}}{3} - \frac{\lambda_2^{\alpha,2'}}{3} - \lambda_3^{\alpha,2'}\right)\right] \times \left[1 - \frac{3}{2}\left(\frac{M}{M_{\mathrm{s}}}\right)^2\right], \tag{3a}$$

— for the magnetostriction parallel to the [110] direction:

$$\lambda_{\mathrm{r}110} = \left(\lambda_1^{\alpha,2} - \lambda_2^{\alpha,2} - \lambda^{\beta,2}\right)\left[1 - \frac{1}{2}\left(\frac{M}{M_{\mathrm{s}}}\right)^2\right]. \tag{3b}$$

Using a fitting procedure the following results have been obtained:

$$\left(\lambda_1^{\alpha,2} - \lambda_2^{\alpha,2} - \lambda^{\beta,2}\right) = -120 \times 10^{-6}$$

and

$$\left[\left(\frac{\lambda_1^{\alpha,2}}{6} - \frac{\lambda_2^{\alpha,2}}{6} - \frac{\lambda_3^{\alpha,2}}{2}\right) + \left(\frac{\lambda_1^{\alpha,2'}}{3} - \frac{\lambda_2^{\alpha,2'}}{3} - \lambda_3^{\alpha,2'}\right)\right] = -40 \times 10^{-6}.$$

From general considerations one should expect that the leading terms in above expressions should be $\lambda_1^{\alpha,2}$ and $\lambda_1^{\alpha,2'}$. It means that in this approximation

$$\lambda_1^{\alpha,2} = -120 \text{ ppms} \quad \text{and} \quad \lambda_1^{\alpha,2'} = -60 \text{ ppm}.$$

These, relatively high, magnetostriction constants seem to arise because of specific mechanism of spin-lattice coupling proposed by Chaudhari and Ghatak [10, 11].

The results of the thermal expansion measurements along the [010] and [110] directions are presented in Fig. 3. Thermal expansion measurements suggest the existence of at least two different regions with different temperature dependences. Up to 180 K, there is a region, in which the Gruneisen law is fulfilled. Above 180 K, there is a region ($180 < T < 192$ K) with the anomalous behavior of the thermal expansion. In this region we have observed a temperature hysteresis of thermal expansion being characteristic of the first-order phase transitions (not shown in Fig. 3). The similar behavior has been observed in measurement of magnetization as a function of temperature [12]. In our opinion the anomaly in thermal expansion seen at T_{S} in Fig. 3 seem to be rather not connected with the carrier localization process (the polaron effect) [13, 14]. The results of structural investigations [8] together with temperature dependences of the lattice parameters would suggest

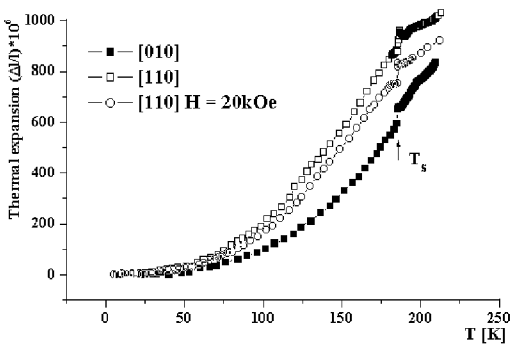

Fig. 3. The thermal expansion of the $La_{0.744}Ba_{0.186}MnO_3$ single crystal along [010] and [110].

that the anomalous thermal expansion could be attributed to the structural phase transition similar to that observed in $La_{1-x}Sr_xMnO_3$ [15].

We have determined also the linear thermal expansion coefficient α_T using the data of the thermal expansion measurements. The thermal expansion coefficient of $La_{0.744}Ba_{0.186}MnO_3$ single crystal has been calculated according to the following formula:

$$\alpha_T = \frac{1}{l_0}\frac{\mathrm{d}l}{\mathrm{d}T}. \tag{4}$$

The results of calculation are presented in Fig. 4. It can be clearly seen that the sharp changes of α_{T} occur in a vicinity of $T_{\mathrm{S}} = 185$ K, in a relatively narrow temperature range of about ± 5 K. These sharp anomalies of the thermal expansion at T_{S}, related to the changes of the lattice parameters, demonstrate that the precursor effects observed usually at structural phase transitions are not very important at the crystals studied.

The results of the thermal expansion measurement as a function of temperature in the external magnetic field (2 T) show that the field only slightly shifts the position of the lower anomaly toward the low temperature but distinctly enlarges the amplitude of the higher one up to 80 ppm. From the results presented in Fig. 4 one can obtain important information that the thermal expansion coefficient reveals two large λ-type phase transition anomalies of comparable amplitude but of opposite signs. We propose to assign this behavior to the combined effects of two different phase transitions, which are almost degenerated in temperature. One could suppose that the higher temperature phase transition (exactly at T_{s}) observed in Fig. 4 (see the inset) should be assigned to the first-order structural phase transition from the rhombohedral to orthorhombic phase. Below T_{s} spontaneous strains appear, therefore the phase transitions observed at lower temperature could be assigned to the second-order phase transition to the phase with spontaneous strain ε (being the order parameter).

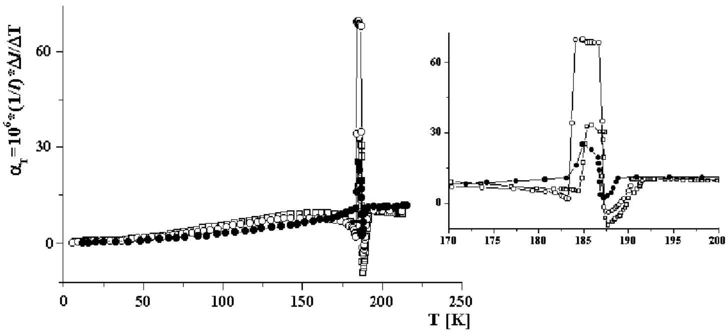

Fig. 4. The linear thermal expansion coefficient of the $La_{0.744}Ba_{0.186}MnO_3$ single crystal as a function of temperature. The inset shows the region of two large λ-type phase transition anomalies. The inset shows details of the $\alpha_T(T)$ structure near 185 K. The symbols have the meaning indicated in Fig. 3.

The spontaneous strains ε_{ij} are coupled to the order parameter M (magnetization), giving terms in energy as follows:

$$\mu_{ij} M^2 \varepsilon_{ij}, \tag{5}$$

where μ_{ij} denotes the coupling constants. It means that the Landau free energy F may be given as

$$F = F_0 + \tfrac{1}{2}a(T - T_{\mathrm{s}})M^2 + \tfrac{1}{4}U_4 M^4 + \tfrac{1}{6}U_6 M^6$$
$$+\tfrac{1}{2}c_{ijkl}\varepsilon_{ij}\varepsilon_{kl} + \mu_{ij}M^2\varepsilon_{ij}, \tag{6}$$

where the last two terms describe elastic and magnetoelastic contributions, respectively. Assuming the condition that a sample is free of stress one can obtain

$$\tfrac{1}{2}c_{ijkl}\varepsilon_{kl} + \mu_{ij}M^2 = 0. \tag{7}$$

This relation suggests that the temperature dependence of magnetostriction should be similar to that observed for magnetization in agreement with data presented in

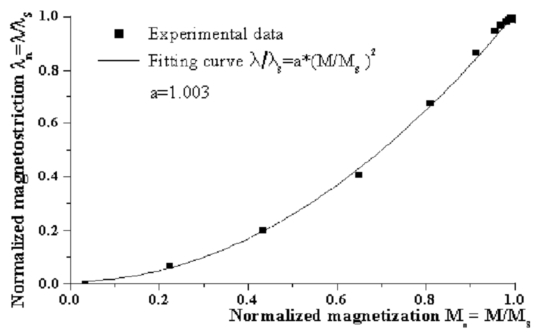

Fig. 5. The normalized magnetostriction as a function of normalized magnetization. The fitting curve $\lambda_n = aM_n^2$ (solid line).

Figs. 1 and 2. The observation that the longitudinal magnetostriction measured for the magnetization parallel to the [010] direction differs from that measured for the magnetization parallel to the [110] direction, results from the anisotropic character of the tensor μ. Simultaneously, Fig. 4 displays a strong effect of magnetic field on spontaneous strain. It suggests, in accordance to Eq. (7), that one of the important contributions to the magnetostriction of $La_{0.744}Ba_{0.186}MnO_3$ single crystals is related to the reduction of spontaneous strains by an external magnetic field. According to this assumption, λ should be proportional to M^2. The fitting curve presented in Fig. 5 confirms this assumption. It should be remarked that the dipolar mechanism of magnetostriction [9] leads to the same dependence. Nevertheless, a strong effect of the magnetic field on a spontaneous strain bears the evidence of the important contribution of proposed mechanism to the effective magnetostriction of the studied crystals.

4. Conclusion

Recently, the new mechanism of magnetostriction in the manganite system has been proposed [10, 11] basing on a theoretical model. In the model the e_g electrons of Mn^{3+} ions are coupled to the lattice due to the Jahn–Teller effect. Simultaneously, it is assumed that the exchange interactions are realized by the double-exchange mechanism. All conclusions have been based on numerical calculations therefore its application to the real system is rather limited.

In the present paper we have proposed a simplified model of magnetostriction in manganites based, to some extent, on ideas proposed by Chaudhari and Ghatak [10, 11]. First of all the following facts were taken into account:

- The $La_{0.8}Ba_{0.2}MnO_3$ manganites exhibit the structural phase transition from the rhombohedral to orthorhombic phase at $T_s = 175$ K, below the Curie temperature $T_C = 255$ K. The structural phase transition is of the first order.
- Both the hydrostatic pressure and magnetic field stabilize the rhombohedral phase [16]. It means that the transfer interaction of e_g carriers is enhanced in this case and consequently the double-exchange interaction becomes more effective.
- In the temperature region where the orthorhombic phase is stabilized the spontaneous strains appear.

On the base of these assumptions supported by the results of thermal expansion measurements, we have derived Eq. (7) relating spontaneous strains to the magnetization. In this way any changes of magnetization by external magnetic field, temperature, pressure, etc. lead to the change of spontaneous strains. It seems that the presented experimental data confirm the proposed mechanism of magnetostriction in the $La_{0.744}Ba_{0.186}MnO_3$ single crystal. The applied magnetic field suppresses spontaneous strain existing below the Curie temperature and it is in fact an origin of magnetostrictive effects. The proposed mechanism does not exclude other mechanisms giving contribution to the magnetostriction in manganites [1].

Acknowledgment

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