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MAGNETOELASTIC ANOMALIES AND SUPPRESSION OF JAHN–TELLER DISTORTIONS IN $La_{1-x}Sr_xMnO_3$ BY PULSED MAGNETIC FIELDS

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Thermal expansion $\Delta l/l(T)$ and magnetostriction $\lambda_{\parallel,\perp}(H,T)$ in the pulsed magnetic fields H up to 250 kOe were studied in the single $La_{1-x}Sr_xMnO_3$ crystals ($x = 0.1$, 0.125, and 0.15) at temperatures $T =$ 10–320 K. Anomalies in the $\Delta l/l(T)$ curves were observed near a charge (polaron) ordering (T_p) and structural transitions between orthorhombic Jahn–Teller distorted O' phase and high temperature orthorhombic (pseudocubic) O^* one (T_s'). A noticeable increase in the field-induced magnetostriction $\lambda_{\parallel,\perp}(H)$ was revealed in the O' phase at $T_p < T < T_s'$. Jumps in the $\lambda_{\parallel,\perp}(H)$ curves were observed at temperatures near T_p. The observed features may be related to a suppression of the Jahn–Teller O' phase by the magnetic field and field induced transition to a new orbital-ordered ferromagnetic state.

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The doped perovskites $R_{1-x}A_xMnO_3$, where R is a trivalent rare-earth ion and A is a divalent ion such as Ca, Sr, has attracted a considerable attention due to an observation of various interesting phenomena such as a colossal negative magnetoresistance, magnetic, structural and metal–insulator phase transitions, a charge ordering [1, 2]. Recent investigations of their magnetoelastic properties and finding of the field induced structural transitions [3, 4] indicate a strong coupling between the magnetic and crystal subsystems in these compounds. In this work we studied thermal expansion at a zero magnetic field and magnetostriction in pulsed fields of $La_{1-x}Sr_xMnO_3$. We focused our attention on the $x = 0.1$, 0.125, and 0.15 compositions which exhibit below a ferromagnetic ordering at T_C a structural phase transition from an orthorhombic Jahn–Teller distorted O' phase to a polaron (charge) ordered one at T_p [5–7]. Above T_C another structural transition between the orthorhombic Jahn–Teller O' phase and high temperature orthorhombic (pseudocubic) O^* phase also takes place at T_s'. Recently, unusual field induced transitions were observed in these compositions in the range $T_p < T < T_C$ accompanied by magnetization and positive magnetoresistance jumps [8–10]. These transitions were associated with a field induced orbital ordering. We observed that they are accompanied by magnetoelastic anomalies.

Single crystals of $La_{1-x}Sr_xMnO_3$ ($x = 0.1$, 0.125, and 0.15) were grown by the floating zone method with radiation heating. X-ray powder diffraction measurements showed that grown crystals were of single phase, however, they had a twin structure. The thermal expansion $\Delta l/l(T)$ was measured by a strain gauge technique at $T = 78$–300 K in a zero magnetic field. The longitudinal and transverse magnetostriction $\lambda_{\parallel,\perp}(H,T)$ was measured by a quartz sensor, stuck on the samples, in pulsed magnetic fields up to 250 kOe at $T = 10-330$ K.

In the temperature dependencies of the thermal expansion (Fig. 1) pronounced anomalies were observed in all compositions corresponding to the known spontaneous structural phase transitions. According to Refs. [5–7] the temperatures of the transitions are: $T_p \approx 130$ K, $T_C \approx 160-170$ K and $T_s' \approx 300$ K for $x = 0.1$, $T_p \approx 150$ K, $T_C \approx 200$ K and $T_s' \approx 250$ K for $x = 0.125$ and $T_p \approx 190-200$ K, $T_C \approx T_s' \approx 220$–$230$ K for $x = 0.15$. We note a sharp jump of the $\Delta l/l(T)$ at the charge (polaron) ordering at T_p for $x = 0.1$ and 0.125 and its transformation to a smooth feature for $x = 0.15$. The structural transition between the Jahn–Teller O' and pseudocubic O^* phases at T_s' is also accompanied by the clear jump for $x = 0.125$. A sign of this transition also appeared near 300 K for $x = 0.1$. In the $x = 0.15$ composition the feature is retained, however its amplitude is reduced. No distinctive anomalies in the $\Delta l/l(T)$ were observed at the Curie temperature T_C.

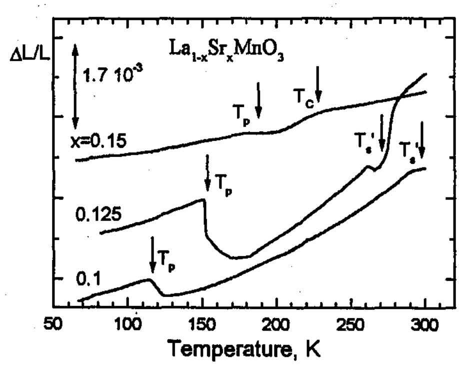

Fig. 1. Temperature dependence of the thermal expansion in $La_{1-x}Sr_xMnO_3$. Arrows indicate temperatures of the spontaneous phase transitions (see the text).

For magnetic field dependence of the magnetostriction $\lambda_{\parallel,\perp}(H,T)$ various anomalies were observed near the spontaneous phase transitions. The example of the $\lambda_{\parallel}(H)$ behavior is shown in Fig. 2a for the composition $x = 0.1$. The $\lambda_{\parallel}(H)$ practically had no anomalies below T_p and reached only the saturation value $\approx 10^{-4}$. However, a significant increase in the $\lambda_{\parallel}(H)$ was observed above T_p, which appeared distinctly in the 150 K curve. We note also a bend for field-up and field-down branches of this curve. The bend is transformed to a negative jump with a further temperature increase (curves 160 and 171 K), whose threshold field is increased and goes beyond our magnetic field range. At $T \to T_s'$ the magnetostriction is reduced and changes a sign (curve 301 K). Such complicated behavior of the magnetostriction indicates on the presence of field induced magnetic (structural) transitions. The similar peculiarities were also observed for the $\lambda_\perp(H)$. A feature of the $\lambda_\perp(H)$ is its large value, which exceeds nearly twice the $\lambda_\parallel(H)$ at some temperature and has the same sign. Noticeable hysteretic phenomena accompanied both the $\lambda_\parallel(H)$ and $\lambda_\perp(H)$ behavior.

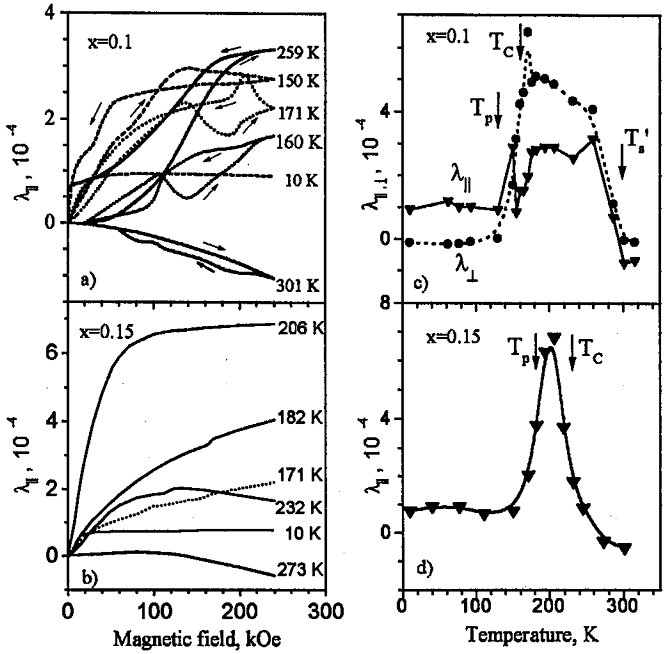

Fig. 2. (a, b) Magnetic field dependencies of the longitudinal magnetostriction in $La_{1-x}Sr_xMnO_3$ for $x = 0.1$ (a) and 0.15 (b); (c, d) temperature dependencies of the longitudinal and transverse magnetostriction for the fixed magnetic field 200 kOe for $x = 0.1$ (c) and 0.15 (d).

In the case of the $La_{0.875}Sr_{0.125}MnO_3$ the increase in the $\lambda_{\parallel,\perp}(H)$ is retained near T_p, as for the $x = 0.1$ composition. In $La_{0.85}Sr_{0.15}MnO_3$ the similar increase in the $\lambda_\parallel(H)$ up to 7×10^{-4} was observed at the range $T_p < T < T_C$, however, the jumps in the $\lambda_\parallel(H)$ behavior were not observed (Fig. 2b).

In order to elucidate magnetoelastic properties in the different phases we have plotted a temperature dependence of the $\lambda_{\parallel,\perp}(T)$ for the fixed magnetic field 200 kOe (Fig. 2c,d). We revealed an apparent correlation in the temperatures of the observed $\lambda_{\parallel,\perp}(T)$ features with the spontaneous transitions at T_p, T_C, and T_s'. At the temperatures slightly above T_p the field induced magnetostriction is positive and relatively large for all the compositions while in the vicinity of the T_C the $\lambda_\parallel(T)$ has a tendency to be reduced and change a sign for $x = 0.1$ and really becomes negative for the $x = 0.125$ composition. A remarkable feature of the $\lambda_{\parallel,\perp}(T)$ curves is a large field-induced value of the magnetostriction in the temperature range $T_p < T < T_s'$, where the distorted Jahn–Teller O' phase exists (we remind that $T_C \approx T_s'$ for $x = 0.15$). This feature may be related to a suppression of the distorted Jahn–Teller O' phase by the magnetic field and field-induced transition to a new orbital ordered state. According to the theoretical studies (for example, [11]) a magnetization process in manganites is strongly affected by the orbital states of Mn ions and is accompanied with the change of the orbital structure. Recently, unusual magnetization and positive magnetoresistance jumps were observed in $La_{1-x}Sr_xMnO_3$ ($x = 0.1$–0.125) at temperatures between T_p and T_C, which were associated with the field induced orbital ordering [8–10]. According to [8, 10] a spontaneous magnetization in the low temperature charge ordered phase is bigger than that in Jahn–Teller O' phase that results in an instability of the latter in the magnetic field. The observed magnetostriction behavior gives evidence that the instability of the distorted Jahn–Teller O' phase takes place not only in a ferromagnetic phase at $T_p < T < T_C$ but also in a paramagnetic state at higher temperature up to T_s'.

In conclusion, we studied the magnetoelastic properties of the $La_{1-x}Sr_xMnO_3$ in the strong magnetic fields, observed the unusual field induced transitions and revealed a remarkable sensitivity of the magnetostriction to the change of the ground state with temperature.

Acknowledgments

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