Magnetic-field-driven structural transition in a $La_{0.8}Ba_{0.2}MnO_3$ single crystal

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The crystal structure, magnetization, and resistivity of the $La_{0.8}Ba_{0.2}MnO_3$ single crystal are studied in the vicinity of the structural phase transition point. The x-ray experiments show the *Pbnm* and $R\bar{3}c$ phases to coexist at least within the interval $185<T<196$ K. The thermal hysteresis of magnetization and resistivity is observed. The interrelations between structural, magnetic, and resistivity properties are analyzed in terms of simple phenomenological theory.

The interest to the lanthanum manganites is caused by the colossal magnetoresistance (CMR) effect and metal-insulator transition, see Refs. 1–3. The strong interaction between charge carriers, localized spins, and lattice degrees of freedom is a characteristic feature of these materials, which results in complexity of the effects observed and difficulty in the interpretation of experimental data. The most attention is given to electronic transport near the magnetic phase transition temperature T_C because it is in the vicinity of T_C that the metal-insulator transition and the CMR take place. The phenomena accompanying the structural phase transition are less studied although they are also very interesting. Thus the experiments performed on $La_{1-x}Sr_xMnO_3$ single crystals have revealed the strong dependence of structural transition temperature T_S on a magnetic field, pronounced hysteretic behavior of resistivity, striction, and sound velocity, subtle interplay between lattice properties, magnetism, and charge ordering.[4–7] The microscopic origin of the effects was indicated in Ref. 4: this is the difference in transfer of e_g holes in two crystal phases as it follows from the double exchange model. A detailed theory, however, is absent.

In this article, we study a member of another manganites family, namely, $La_{0.8}Ba_{0.2}MnO_3$. A single crystal of this composition was made by floating zone technique. The details of preparation have been described elsewhere.[8] From the resulting rod, the samples for x-ray, magnetization, and resistivity measurements were cut. The crystal structure and composition were studied with DRON-3 diffractometer in Cr-K_α radiation. The temperature dependence of x-ray patterns was investigated over the range 80–300 K under warming. The patterns taken at room temperature were treated by means of the ''FULLPROF'' program.[9] The magnetization experiments were performed with vibrating sample and SQUID magnetometers with magnetic field being along the orthorhombic *c* axis. The resistivity ρ was measured with standard four-probe technique in magnetic field up to 120 kOe directed as in the magnetization experiments.

The Curie temperature T_C evaluated through Arrott-Belov plots is about 251 K. The saturation field H_S is lower 10 kOe. Near T_C, the ρ-T curve displays a standard peak, which is suppressed and shifted to higher temperatures by a magnetic field. The detailed study of magnetic and transport properties will be published elsewhere; in this article we focus on the effects near structural transition temperature.

Figure 1 shows the experimental x-ray pattern taken at $T=300$ K (open circles) and calculated one (solid line) of the sample studied. The temperature dependence of lattice parameters is presented in Fig. 2. In the range 80 to 185 K only the lines of orthorhombic *Pbnm* phase are observed. At the temperature of 185 K, there appear the weak lines of rhombohedral $R\bar{3}c$ phase. The intensity of the latter lines increases with increasing temperature while the intensity of the *Pbnm* lines decreases and above $T=196$ K these lines practically disappear. Thus the structural phase transition is of the first order, the *Pbnm* and $R\bar{3}c$ phase coexisting at least within the interval $185<T<196$ K.

Figure 3 shows the temperature dependence of magnetization *M* taken in the magnetic field of 10 kOe. The thermal hysteresis is seen approximately from 185 to 210 K; it is to be noted, however, that the boundaries of the hysteresis loop are fuzzy. Near the center of the loop, the difference between *M* measured under heating and cooling is about 4 G, which is less than 1% of magnetization value. The upper part of the loop corresponds to the cooling of the sample and the lower part refers to the heating.

The most pronounced hysteresis is observed on resistivity

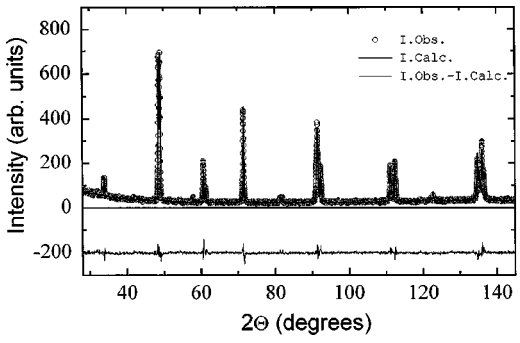

FIG. 1. X-ray patterns of $La_{0.8}Ba_{0.2}MnO_3$. Open circles, observed in experiment; solid line, calculated with FULLPROF program (Ref. 8).

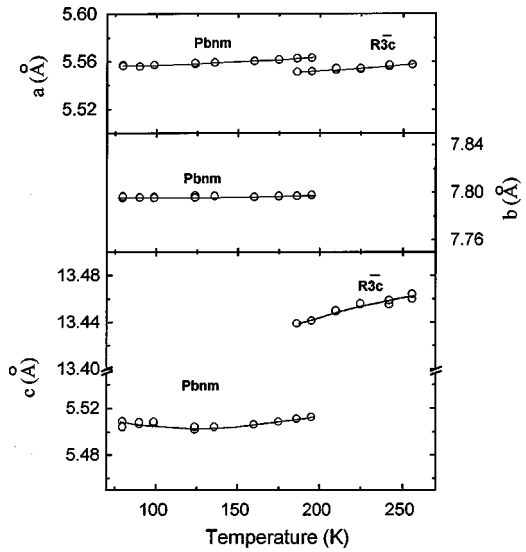

FIG. 2. Temperature dependence of lattice parameters.

versus temperature curves presented in Fig. 4. At $H=0$, the hysteresis loop ranges approximately from 180 to 210 K. Application of a magnetic field shifts the loop to lower temperatures, which suggests the decrease of T_S. In the inset of Fig. 5 we present the magnetic field dependence of T_1 which is defined as a position of the kink at the upper part of the ρ-T loop, see Fig. 4. The dependence of T_1 is approximately linear in H except weak fields ($H<10$ kOe); in the latter region the position of the kink is likely to be determined not only by the structural transition but also by magnetic domains. Magnetic field reduces also the difference between ρ at upper and lower parts of the loop and decreases thereby the area S of the loop, see Fig. 5.

Unlike the M-T loop, the upper part of the ρ-T loop corresponds to the heating while the lower part corresponds to the cooling. This fact helps to understand the origin of the ρ-T hysteresis. Indeed, below T_C (but not far from the magnetic transition point) the resistivity of manganites is decreasing function of magnetization. In the structural phase transition region, the high-temperature rhombohedral phase has higher magnetization than the low-temperature orthorhombic one and hence we may expect the resistivity of the rhombohedral phase to be lower than that of the orthorhombic one. This agrees with our data; therefore the temperature and magnetic field dependence of resistivity can be considered as a consequence of the change in magnetic state.

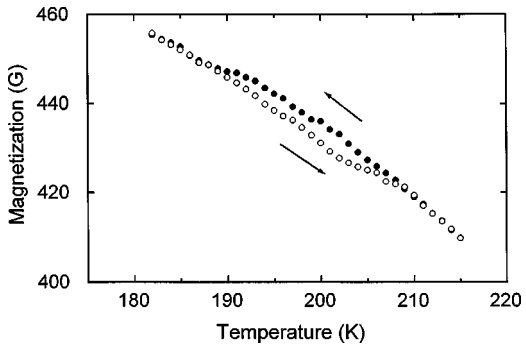

FIG. 3. Temperature dependence of magnetization under cooling (solid circles) and heating (open circles).

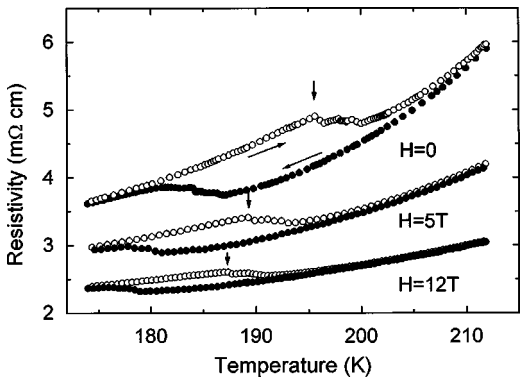

FIG. 4. Thermal hysteresis of resistivity. The arrows indicate T_1.

To understand the interrelation between magnetic and lattice properties it is useful to exploit the ideas developed more than thirty years ago in connection with the properties of MnAs (see, for example, the paper by Bean and Rodbell).[10] For simplicity we shall ignore the hysteresis. The starting point is the expression for the energy of the crystal which is assumed to have the form $F=F_M+F_{\rm lat}+\frac{1}{2}KM^2Q$ where F_M is the magnetic part, $F_{\rm lat}$ is lattice one, and the last term describes the interaction between magnetic and lattice degrees of freedom; K is the coupling constant, Q is the function of lattice variables which, as well as $F_{\rm lat}$ we do not need to specify; a possible form of $F_{\rm lat}$ and Q can be found in Ref. 5. The structural transition occurs in ferromagnetic state not too far from the Curie point, so that we may take magnetic part of energy in Landau form:

$$F_M=\frac{A}{2}M^2+\frac{B}{4}M^4-MH$$

with $A=\alpha(T-T_C^{(0)})$ where α and B are phenomenological constants of Landau theory, $T_C^{(0)}$ is the Curie temperature when $K=0$. The magnetization can be taken to be linear in H, i.e., $M(H)=M(0)+\chi H$ with χ being the paraprocess susceptibility. Since the change in T_S, caused by a magnetic field, is small, we shall restrict ourselves by linear approximation in coupling constant K. We assume that if $K=0$, the sample is in phase 1 at $T<T_S^{(0)}$ and it is in phase 2 when $T>T_S^{(0)}$; in other words, $F_{1\rm lat}-F_{2\rm lat}=\gamma(T-T_S^{(0)})$ where γ is positive constant. The magnetization $M_{1,2}$ obeys the equation

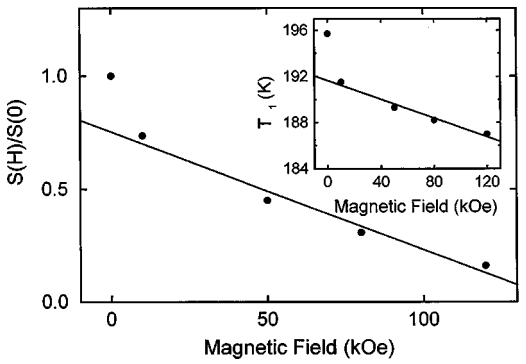

FIG. 5. Temperature dependence of the area of the resistivity hysteresis loop. The line is linear fit for $10\leqslant H\leqslant 120$ kOe. The inset shows T_1 versus H with linear fit for $10\leqslant H\leqslant 120$ kOe.

$$\alpha\left(T-T_C^{(0)}+\frac{KQ_{1,2}}{\alpha}\right)M_{1,2}+BM_{1,2}^3-H=0, \qquad (1)$$

where $Q_{1,2}$ should be taken at $K=0$. Equation (1) shows that the interaction results in renormalization of the Curie temperature and the jump in magnetization $\Delta M=M_1-M_2$ at the temperature of the structural transition T_S, which is also renormalized. The simple calculations give the following results. The jump ΔM is proportional to K and is reduced by magnetic field

$$\Delta M(H)=\Delta M(0)\left(1-\frac{2\chi H}{M(0)}\right), \qquad (2)$$

where $M(0)$ and χ should be taken at $T=T_S$. Strictly speaking, $M(0)$ and χ in the right hand side of the last equation refer to the $K=0$ case; in linear theory, however, we may neglect the difference between these quantities in the $K=0$ and the $K\neq 0$ cases. The H dependence of the structural transition temperature is given by

$$T_S(H)=T_S(0)+\frac{\Delta M(0)H}{2\gamma}. \qquad (3)$$

This equation expresses the well-known thermodynamic rule: a magnetic field favors the phase with higher magnetization. The difference in the Curie temperatures $\Delta T_C=T_{C1}-T_{C2}$ can be written as

$$\Delta T_C=\frac{\Delta M(0)}{\alpha\chi M(0)}. \qquad (4)$$

Let us compare the formulas derived with our experimental data. We shall take that $T_1\approx T_S$. From Fig. 3 we obtain $\Delta M(0)\approx -4$ G. Then from Eq. (3) it follows that $T_1(H)$ should be linear in H with negative slope; the inset in Fig. 5 shows that this is true if $H>H_S$.

Now we turn to the H dependence of the area S of the ρ-T hysteresis loop, see Fig. 5. Let us assume that the reduction of the area is due to the decrease of $|\Delta M|$ in a magnetic field. By making use of Eq. (2) we obtain $d\ln S/dH$ should be $-2\chi/M(0)$. From magnetization curve for $T=200$ K (not shown) we have found $M(0)=440$ G, $\chi=1.2\times10^{-3}$, so that $2\chi/M(0)=5.5\times10^{-6}$ Oe^{-1}. The experimental value of $d\ln S/dH$ for $H\geqslant 10$ kOe, see Fig. 5, is -6.9×10^{-6} Oe^{-1}, which agrees with the calculated one. Thus the change in resistivity is indeed the consequence of the change in magnetization, at least in its main part.

We can estimate also the difference in the Curie temperatures between orthorhombic and rhombohedral phases. When determining the Curie point from Arrott-Belov curves, we have found $\alpha=4.7$ K^{-1}. Substituting $\Delta M(0)$, χ and α into Eq. (4), we obtain $\Delta T_C=1.6$ K. One can see that this value is close to the distance (which is about 2.3 K) between the ascending and descending branches of hysteresis loop in Fig. 3 measured near the center of the loop at a fixed M. The fact that the Curie temperature of the orthorhombic phase is lower than that of the rhombohedral one indicates that exchange interaction in the former case is weaker, which agrees with double exchange picture outlined in Ref. 4.

The comparison of our results with those published previously shows that the main features of the effects near the structural phase transition point in $La_{1-x}Sr_xMnO_3$ and $La_{1-x}Ba_xMnO_3$ manganites are similar. The change in resistivity is caused by the change in magnetic state. The interconnection between lattice, magnetic, and resistivity properties can be described quantitatively in the frame of the simple phenomenological theory.

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