

Steplike Lattice Deformation of Single Crystalline $(\text{La}_{0.4}\text{Pr}_{0.6})_{1.2}\text{Sr}_{1.8}\text{Mn}_2\text{O}_7$ Bilayered Manganite

M. Matsukawa,^{1,*} Y. Yamato,¹ T. Kumagai,¹ A. Tamura,¹ R. Suryanarayanan,² S. Nimori,³ M. Apostu,² A. Revcolevschi,² K. Koyama,⁴ and N. Kobayashi⁴

¹*Department of Materials Science and Technology, Iwate University, Morioka 020-8551, Japan*

²*Laboratoire de Physico-Chimie de L'Etat Solide, CNRS, UMR8182 Universite Paris-Sud, 91405 Orsay, France*

³*National Institute for Materials Science, Tsukuba 305-0047, Japan*

⁴*Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan*

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We report a steplike lattice transformation of single crystalline $(\text{La}_{0.4}\text{Pr}_{0.6})_{1.2}\text{Sr}_{1.8}\text{Mn}_2\text{O}_7$ bilayered manganite accompanied by both magnetization and magnetoresistive jumps, and examine the ultrasharp nature of the field-induced first-order transition from a paramagnetic insulator to a ferromagnetic metal phase accompanied by a huge decrease in resistance. Our findings support that the abrupt magnetostriction is closely related to an orbital frustration existing in the inhomogeneous paramagnetic insulating phase rather than a martensitic scenario between competing two phases.

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The lattice transformation associated with a field-induced metamagnetic transition, observed in perovskite manganites and a class of rare-earth intermetallic compounds, gives rise to significant information for our understanding of the dynamics of first-order phase transitions in solid-state physics [1]. For example, the structural transformation of intermetallic compounds from an austenite (parent) to a martensite phase upon increasing an external field is accompanied by a metamagnetic transition from an antiferromagnetic (AFM) to a ferromagnetic (FM) state [2]. This type of transition contrasts with that activated thermally where thermal fluctuations drive the system from a metastable towards a stable state at constant external parameters (external magnetic field, temperature, and external stress) [3]. A martensite transformation is associated with a significant change of the unit cell from the parent to the new phase and takes place as a sequence of avalanches between metastable states due to local strain fields stored in the lattice [4].

Recent experimental and theoretical studies on doped manganites exhibiting a colossal magnetoresistance (CMR) effect have revealed the inhomogeneous coexistence of charge-ordered AFM insulating and FM metallic phases in a phase-separated ground state [5]. The low-temperature phase-separated state is composed of a charge-ordered (CO) matrix phase with a large lattice distortion due to a cooperative Jahn-Teller effect and metallic FM clusters with a different unit cell from the former phase, indicating the presence of substantial strains at CO/FM interfaces. Several recent studies on metamagnetic transitions of manganites have also shown that ultrasharp magnetization steps appear at low temperatures. To account for this, a model based on a martensitic scenario between competing CO/FM phases has been proposed though questions have been raised against this model [6–10].

The purpose of this Letter is to present a detailed investigation of anisotropic magnetostrictions of single crystalline $(\text{La}_{0.4}\text{Pr}_{0.6})_{1.2}\text{Sr}_{1.8}\text{Mn}_2\text{O}_7$, and to examine the dynamics of a steplike first-order transition from a paramagnetic insulating (PMI) to a ferromagnetic metal (FMM) phase in CMR manganites. Our data strongly favor an orbital frustration to account for our findings.

For the Pr-substituted $(\text{La}_{0.4}\text{Pr}_{0.6})_{1.2}\text{Sr}_{1.8}\text{Mn}_2\text{O}_7$ crystal, a spontaneous ferromagnetic metal phase (originally present with no Pr substitution) disappears at ground state, but a field-induced PMI to FMM transformation is observed over a wide range of temperatures. A magnetic (H, T) phase diagram, established from magnetic measurements, is separated into three regions labeled as PMI, FMM, and phase-separated (PS) states (see Refs. [11,12]). The proximity of free energies between the PMI and FMM states is of importance to stabilize the PS state. Application of an external field destabilizes the PMI ground state and drives the transition to the FMM state through the intermediate PS state. As seen from the magnetic phase diagram, a stronger field is needed to induce the PMI to FMM transition at low temperatures since the thermal energy is reduced upon lowering temperature. This tendency points to a thermally activated character of the metamagnetic transition.

Single crystals of $(\text{La}_{0.4}\text{Pr}_{0.6})_{1.2}\text{Sr}_{1.8}\text{Mn}_2\text{O}_7$ were grown by the floating zone method using a mirror furnace. The lattice parameters as well as the chemical homogeneity of the tetragonal crystal were reported earlier [13]. The dimensions of the $z = 0.6$ sample are $3.4 \times 3 \text{ mm}^2$ in the ab plane and 1 mm along the c axis. Measurements of magnetostriction, both in the ab plane and along the c axis, were done by means of a conventional strain gauge method at the Tsukuba Magnet Laboratory, the National Institute for Materials Science (NIMS), and at the High Field Laboratory for Superconducting Materials, Institute for

Materials Research, Tohoku University. The magnetization measurements were made by using a VSM magnetometer at NIMS. The sample was zero-field cooled from 300 K down to low temperatures and we then started measuring the isothermal magnetostriction upon increasing (or decreasing) the applied fields, parallel to the c axis. The normal sweep rate was set to be 0.26 T/min.

Figure 1 shows the isothermal magnetostriction data of a $(\text{La}_{0.4}\text{Pr}_{0.6})_{1.2}\text{Sr}_{1.8}\text{Mn}_2\text{O}_7$ single crystal at $T = 4.3$ and 10 K. First of all, we emphasize qualitative differences in the magnetostriction between ultrasharp transformations at low T observed here and broad variations at higher T

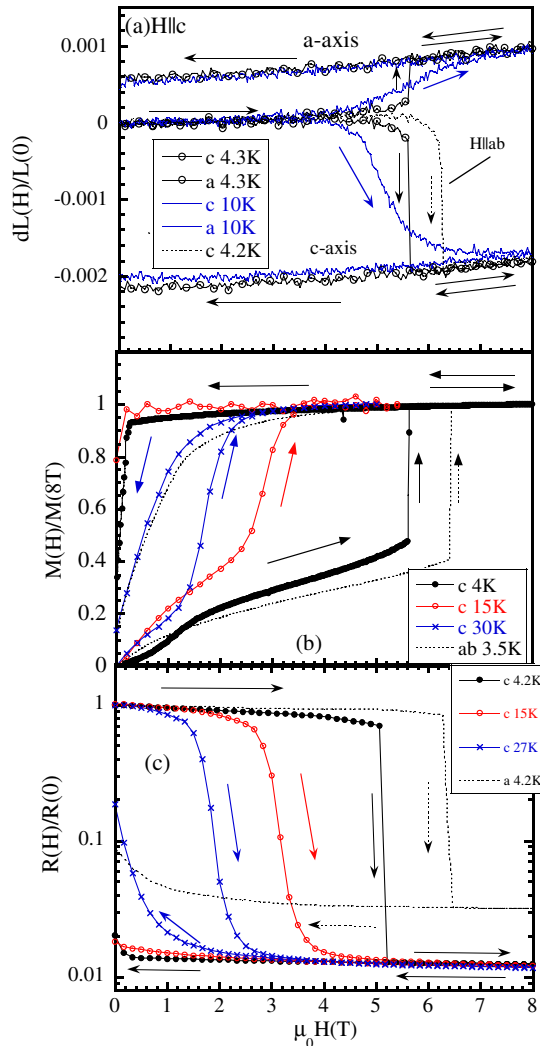


FIG. 1 (color online). (a) The c -axis and ab -plane isothermal magnetostriction of a $(\text{La}_{0.4}\text{Pr}_{0.6})_{1.2}\text{Sr}_{1.8}\text{Mn}_2\text{O}_7$ single crystal at $T = 4.3$ and 10 K ($H \parallel c$). A dotted curve represents the c -axis data at 4.2 K in the case of $H \parallel ab$. The small magnetoresistance effect of the strain gauge itself gives a slight increase in both dL_c and dL_{ab} . (b) Field dependence of the isothermal magnetization of the same crystal at selected temperatures ($H \parallel c$). For comparison, the ab -plane magnetization data are given at 3.5 K ($H \parallel ab$). (c) c -axis magnetoresistances of the same crystal at selected temperatures in the cases of $H \parallel c$. For comparison, the ρ_{ab} data at 4.2 K ($H \parallel ab$) are cited.

previously reported in [11] (width is approximately the order of 1 T). Both dL_c and dL_{ab} are independent of the applied field up to ~ 5 T. However, upon further increasing the field, the c axis abruptly shrinks by $\sim 0.2\%$ at low T , whereas the $a(b)$ axis expands simultaneously. The transition widths are estimated to be within 0.1% of the magnitude of the critical field H_c from the present data. Following a steplike change of the magnetostriction at H_c , the values of dL_c and dL_{ab} then remain almost constant upon increasing H up to 8 T and decreasing it down to zero field. Finally, the $dL_c(H)$ and $dL_{ab}(H)$ curves exhibit a large hysteresis and the remanent striction remains very stable for about one day after switching off the field. For comparison, the isothermal magnetization and magnetoresistance of the same composition crystal are presented in Figs. 1(b) and 1(c). Here, the value of M_c (8 T) reaches $\sim 3.5\mu_B/\text{Mn}$ site, close to the full moment of $3.6\mu_B/\text{Mn}$ site, while the value of M_{ab} is $\sim 3.0\mu_B/\text{Mn}$ site, which is by about 20% smaller than the ideal value [13]. The corresponding magnetization and magnetoresistance data also exhibit a steplike transition at the same critical field. We note that a steplike behavior of the magnetostriction is not observed for polycrystalline $(\text{Sm}, \text{Sr})\text{MnO}_3$ manganite in spite of the appearance of both a steplike magnetization and magnetoresistance [8]. The steps observed in $dL(H)$, $M(H)$, and $\rho(H)$ are almost independent of the field direction.

Next, we present in Fig. 2(a) the c -axis magnetostriction data as a function of cooling field, because the cooling field affects a fraction of the FMM region [6]. All $dL_c(H)$ data show a smooth decrease and then follow a steplike drop. The value of $dL_c(H)$ decreases from $\sim 0.18\%$ at the FC-1T run down to $\sim 0.1\%$ at the FC-1.6T run [see the caption of Fig. 2(a)]. The substantial difference in $dL_c(H)$ between the FC-1T and FC-1.6T runs indicates that the FMM phase is partially formed within the PMI matrix through each FC run, before magnetostriction measurements are done. The remarkable jump observed in $dL_c(H)$ is monotonically decreased upon increasing the cooling field from 1 T up to 1.6 T, accompanied by a nonlinear dependence of H_c from 5.6 T up to 7.6 T. For all measurements exhibiting the steplike transition, a divergent behavior of bath temperature was observed at the corresponding H_c , as shown in Fig. 2(b). The larger steps in $dL_c(H)$ accompany a more sudden temperature rise. In addition to it, the apparent striction in both $dL_c(H)$ and $dL_a(H)$ at the steplike transition suggests a rapid rise of the sample's temperature due to the released heat [14] [see the caption of Fig. 2(a)]. It has been reported from the magnetocaloric measurement on polycrystalline $\text{La}_{0.225}\text{Pr}_{0.4}\text{Ca}_{0.375}\text{MnO}_3$ that a released heat at the ultrasharp field-induced transition gives rise to the abrupt increase of the sample temperature $\delta T \sim 30$ K [9]. When the cooling field exceeds 1.6 T, the steplike behavior of $dL_c(H)$ disappears.

Then, after a first scan in zero-field cooling, we carried out successive measurements of the isothermal magnetostriction. At the beginning of further scans, we reset the

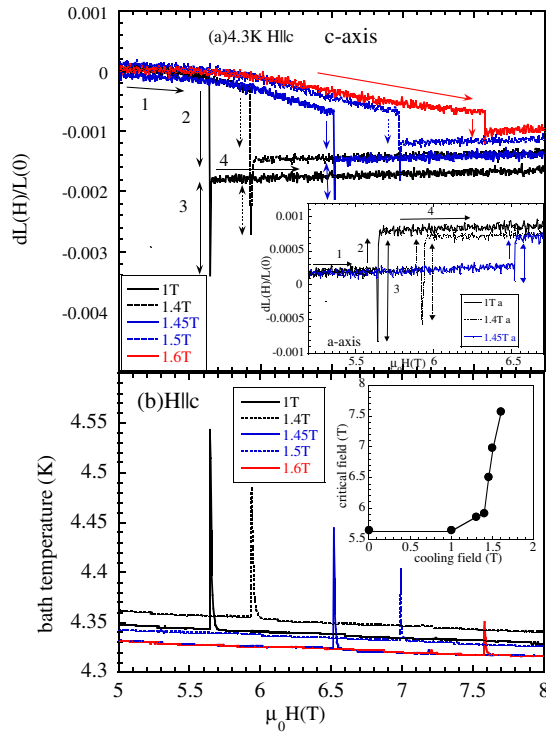


FIG. 2 (color online). (a) c -axis magnetostriction as a function of cooling field (1, 1.4, 1.45, 1.5, and 1.6 T). For comparison, the a -axis data are displayed in the inset of (a). First, warming the sample from 4.3 K up to 200 K in the absence of field and then cooling it from 200 K down to 4.3 K in the respective field during 30 min (FC), the applied field was removed. Finally, we start measuring the magnetostriction upon increasing the field up to 8 T. For each measurement, we repeated the same procedure. The arrows labeled 1 and 4 mark $dL_c(H)$, before and after the transition, respectively. Arrow 2 represents the net lattice deformation, while arrow 3 points to the apparent change caused by the rise of sample temperature. (b) The corresponding bath temperature versus applied field. The slight decrease in the bath temperature as a function of H arises from the magnetoresistance effect of the cernox thermometer. The inset of (b) represents the critical field H_c versus the cooling field, at 4.3 K.

sample from the remanent FMM to the PMI virgin state by a local heating (see Ref. [15]). In the second scan [Fig. 3(b)], once $dL(H)$ shows a continuous variation with the insulator-metal transition, near 6.3 T, a sudden jump along both the c axis and the a axis is also observed. However, the third scan data show no discontinuous step, similar to the normal $dL(H)$ curves at higher temperatures. The critical field of the second scan is increased up to 6.3 T from 5.3 T in the first scan. The qualitative differences in the $dL(H)$ curves among the three scans probably arise from a training effect due to the local heating from the strain gauges attached on the sample. Such a local heating erases the memories experienced by the sample, removes frustrations existing within the PMI matrix, and stabilizes the PMI state, resulting in a widely broad transition, as seen in the third scan. For comparison, we examine how the cooling rate from high temperatures down to 4.3 K affects

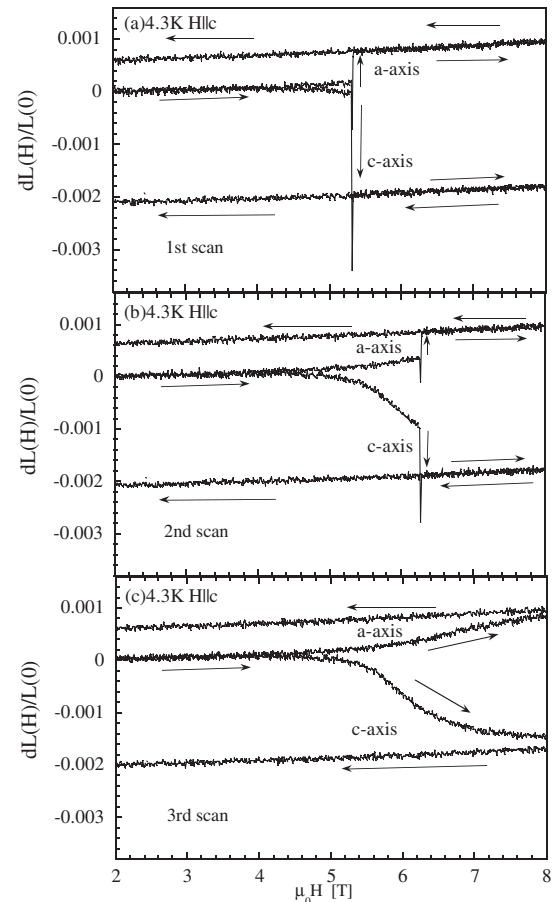


FIG. 3. Field dependence of the c -axis and a -axis isothermal magnetostriction, dL_c and dL_{ab} , at 4.3 K ($H \parallel c$). (a) First scan, (b) second scan, and (c) third scan. Here, first scan of the applied field denotes $0 \rightarrow 8 \rightarrow 0$ T. Second (third) scan: local heating $\rightarrow 0 \rightarrow 8$ (12) $\rightarrow 0$ T. (For details, see the text).

the critical field. In both rapidly and slowly cooling procedures, the step phenomena appear (Fig. 4), and we get the comparable values of $H_c = 5.36$ T (rapid) and 5.2 T (slow) in the case of 300 K. In a similar way, $H_c = 5.63$ and 5.58 T, rapid and slow cooling rates from 200 K, respectively. We found out that H_c is almost independent of the cooling rate and is suppressed in the cooling process from a higher temperature of 300 K. In addition, checking the sweep rate dependence of H_c , we get 5.6 T in both 0.52 and 0.26 T/min and 5.8 T at the slow rate of 0.052 T/min (not shown here).

Finally, we discuss now the origin of the magnetostrictive jump observed here. The parent bilayered manganite $\text{La}_{1.2}\text{Sr}_{1.8}\text{Mn}_2\text{O}_7$ exhibits a PMI to FMM transition around $T_c \sim 120$ K and an associated CMR effect [16]. In the half-doped bilayered manganite of $\text{La}_{2-2x}\text{Sr}_{1+2x}\text{Mn}_2\text{O}_7$ ($x = 0.5$), near the $x = 0.4$ crystal, a long-range orbital- and charge-ordered (OO/CO) state is established, accompanied by a CE-type (zigzag FM chains with AFM interchain coupling) antiferromagnetic structure [17]. Neutron scattering measurements on a bilayered manganite near optimal doping show that CE-type OO/CO clusters (short-

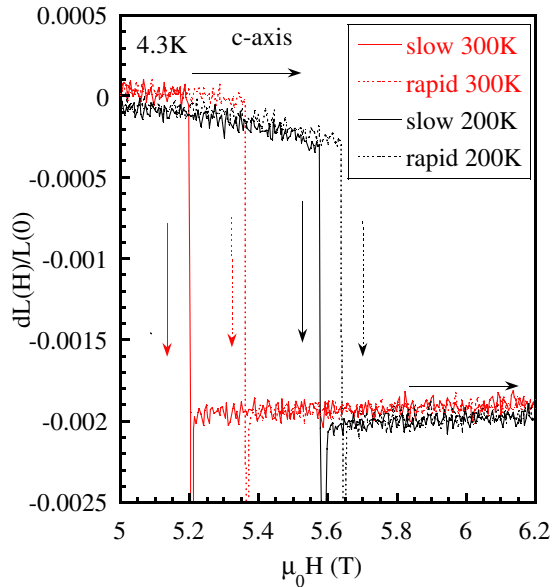


FIG. 4 (color online). Cooling rate dependence of $dL(H)$ from 200 and 300 K down to 4.3 K. In the case of 200 K, rapid and slow cooling rates are set at 30 min and 12 h. At 300 K, rapid and slow cooling rates in 20 min and 15 h, respectively.

range polaron correlations) freeze upon cooling to $T^* \approx 310$ K (T^* : glass transition temperature), as a magnetic frustration between FM and AFM interactions leads to a spin-glass state [18]. This finding strongly suggests the existence of an orbital frustration in the PMI phase, which prevents the formation of the CO phase. Thus, we believe that an orbital frustration exists within the PMI state of the present Pr-substituted sample. Once this type of frustration is initially formed at high temperatures within the sample, one expects that it survives down to low temperatures. In the successive isothermal scans (Fig. 3), the local heating process relaxes the frustration existing within the matrix of the sample, giving a standard broad transition. In other words, in the first scan, the frustration existing probably enhances the instability of the metastable state of the free energy under the magnetic field, resulting in a steplike behavior. These findings are consistent with the fact that the critical field is sensitive to the thermal profile undergone by the sample from high temperatures. This scenario also accounts for the cooling field dependence of H_c observed here and reported in magnetization steps [6,10]. The field-cooled run suppresses the CO clusters but increases the FM clusters. Accordingly, the reduced frustration affects a steplike transition, increasing the value of H_c , which is consistent with the positive dependence of H_c on the cooling field. The nucleation and growth of FMM domains due to an external field reduce the magnetic energy by the Zeeman energy, while they increase the elastic energy arising from strains stored at the interface of two-phase domains. It is true that this competition in

free energy has some relation with the transition, but it never reproduces quite different isothermal behaviors under the same external field scans.

In summary, we observed a steplike magnetostriction in a single $(\text{La}_{0.4}\text{Pr}_{0.6})_{1.2}\text{Sr}_{1.8}\text{Mn}_2\text{O}_7$ sample, accompanied by steps in both magnetization and magnetoresistance. Our data suggest that the abrupt lattice transformation reported here cannot be described by a martensitic transformation but is closely related to an orbital frustration existing in the inhomogeneous PMI phase.

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*matsukawa@iwate-u.ac.jp

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- [15] For the local heating, a current of 15 mA was applied to a series of two gauges during 10 sec so that the gauge resistance returned rapidly to an initial value before the application of field, corresponding to the PMI virgin state. After checking the stability of the bath temperature, we then restarted measuring $dL(H)$. For each measurement, the excited current of the gauge was set to be 1 mA.
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