

Stretched exponential behavior in remanent lattice striction of a $(\text{La,Pr})_{1.2}\text{Sr}_{1.8}\text{Mn}_2\text{O}_7$ bilayer manganite single crystal

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We have investigated the time dependence of remanent magnetostriction in a $(\text{La,Pr})_{1.2}\text{Sr}_{1.8}\text{Mn}_2\text{O}_7$ single crystal in order to examine the slow dynamics of lattice distortion in bilayered manganites. A competition between double-exchange and Jahn-Teller-type orbital-lattice interactions results in the observed lattice profile following a stretched exponential function. This finding suggests that spatial growth of the local lattice distortions coupled with e_g -electron orbital strongly correlates with the appearance of the field-induced colossal magnetoresistance effect.

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The discovery of the colossal magnetoresistance (CMR) effect in doped manganites with perovskite structure has stimulated considerable interest for the understanding of their physical properties.¹ Though the insulator to metal (IM) transition and its associated CMR are well explained on the basis of the double-exchange (DE) model, it is pointed out that the dynamic Jahn-Teller (JT) effect due to the strong electron-phonon interaction plays a significant role in the appearance of CMR as well as the DE interaction.^{2,3} Furthermore, Dagotto *et al.* propose a phase separation model where the ferromagnetic (FM) metallic and antiferromagnetic (AFM) insulating clusters coexist and their model strongly supports recent experimental studies on the physics of manganites.⁴

The bilayer manganite $\text{La}_{1.2}\text{Sr}_{1.8}\text{Mn}_2\text{O}_7$ exhibits a paramagnetic insulator (PMI) to ferromagnetic metal (FMM) transition around $T_c \sim 120$ K and its associated CMR effect.⁵ In comparison with cubic manganites, the MR effect of the compound under consideration, due to its layered structure, is enhanced by two orders of magnitude, at 8 T, around T_c . It is well known that Pr substitution on the La site leading to $(\text{La}_{1-z}, \text{Pr}_z)_{1.2}\text{Sr}_{1.8}\text{Mn}_2\text{O}_7$ causes an elongation of the c -axis length in contrast with a shrinkage of the $a(b)$ axis, resulting in a change of the e_g -electron occupation from the $d_{x^2-y^2}$ to $d_{3z^2-r^2}$ orbital.^{6,7} For $z=0.6$, a strong suppression of the e_g -electron bandwidth leads to a PM insulating ground state.⁸ However, the FM metastable phase with a metallic conduction is easily obtained by the application of a magnetic field. The field-induced first-order IM transition is further accompanied by a memory effect of magnetoresistance, magnetization, magnetostriction, and magnetothermal conductivity.⁸⁻¹⁰ This memory effect may be explained by a schematic picture of the free energy exhibiting two local minima corresponding to the FMM and PMI states, as shown in the inset of Fig. 1(a). In the absence of a magnetic field, the system stays in the stable PI state but, upon field application, the FMM state becomes metastable against the PMI state. After removing the field, the system should go back to a pure PMI ground state through the various kinds of mixed

states after a very long time. Recently, Gordon *et al.*⁹ have reported that the time dependence of the remanent magnetization of $(\text{La,Pr})_{1.2}\text{Sr}_{1.8}\text{Mn}_2\text{O}_7$ follows a stretched exponential function, which is closely related to a magnetic frustration between FM double-exchange and AFM superexchange interactions. In several reports concerning cubic manganites, the magnetization and resistivity relaxation effects have been discussed within the framework of the phase segregation between FM metal and AFM charge-ordered insulating states.¹¹⁻¹⁴ In addition, there are a few reports on magnetic and transport properties in doped bilayer manganites which extend our understanding of a spin-glass-like phase related to the phase separation model.^{9,15} However, to our knowledge, the slow dynamics of a lattice relaxation in doped manganites has not been reported so far.

In this paper, we report the time dependence of remanent magnetostriction in the Pr-substituted bilayer manganites. A frustration of lattice deformation between double-exchange and Jahn-Teller-type orbital-lattice interactions plays a crucial role in the observed slow relaxation going from the FM metal to PM insulator.

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 calculated lattice parameters were reported in Ref. 8. The dimensions of the $z=0.6$ sample are 3.4×3 mm² in the ab plane and 1 mm along the c axis. Magnetostriction, both in the ab plane and along the c axis, was measured by means of a conventional strain gauge method at the Tsukuba Magnet Laboratory, the National Institute for Materials Science and at the High Field Laboratory for Superconducting Materials, Institute for Materials Research, Tohoku University.¹⁶ The magnetization measurements were made by using a superconducting quantum interference device magnetometer at Iwate University. A magnetic phase diagram in the (H, T) plane was established from the magnetic measurements on the $z=0.6$ crystal as shown in the inset of Fig. 1(b).^{8,17} The phase diagram is separated into three regions labeled as the PMI, FMM, and mixed phases [the hatched area in the (H, T) plane]. In the mixed

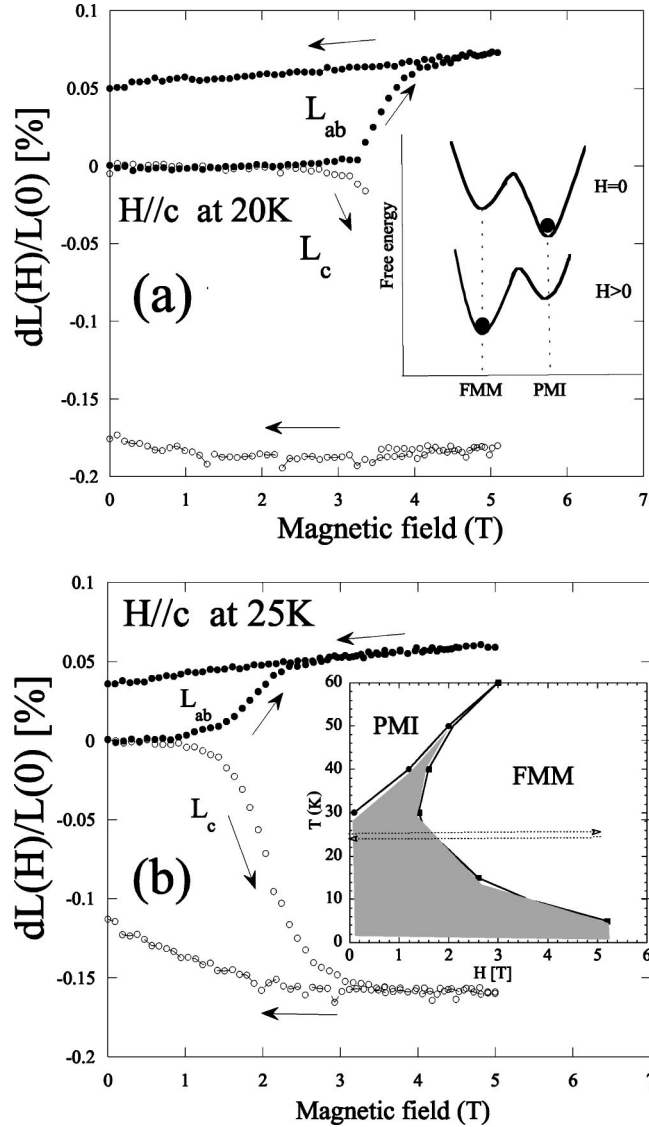


FIG. 1. Magnetostriction both in the ab plane and along the c axis, for single-crystalline $(\text{La}_{0.4}, \text{Pr}_{0.6})_{1.2}\text{Sr}_{1.8}\text{Mn}_2\text{O}_7$. A schematic picture of the free energy with two local minima corresponding to the FMM and PMI states is shown in the inset of (a). The inset of (b) represents a magnetic phase diagram in the (H, T) plane established from the magnetic measurements carried out on the $z=0.6$ crystal.

phase, the PMI and FMM clusters coexist, which is characterized by hysteresis in the magnetization curves. Thus, we tried to look for a relaxation effect for the $z=0.6$ crystal at temperatures selected between 20 and 30 K, covering a large to small hysteresis region. After cooling the sample down to the respective temperatures in a zero field, the field was applied along the c axis up to 5 T at a sweep rate of 0.2 T/min and was then decreased down to zero at the same rate. A dotted horizontal line in the phase diagram represents such an experimental procedure crossing the phase boundary separating the hysteresis and FM areas. Finally, magnetostriction data were recorded as a function of time just after the field was switched off.

Magnetostriction data both in the ab plane and along the c

axis, for the $(\text{La}_{0.4}, \text{Pr}_{0.6})_{1.2}\text{Sr}_{1.8}\text{Mn}_2\text{O}_7$ single crystal, are shown in Fig. 1 as a function of magnetic field applied parallel to the c axis at selected temperatures. At 20 K, the value of $dL_c(H)/L_c(0)$ exhibits an abrupt decrease around 3 T, while the value of $dL_{ab}(H)/L_{ab}(0)$ rises at the same field value. Here, the value of $dL_i(H)$ is defined as $L_i(H) - L_i(0)$. In other words, the c axis length suddenly shrinks around 3 T in contrast with the expansion of the $a(b)$ axis accompanied by the field-induced IM transition. Combining $dL_{ab}(H)/L_{ab}(0)$ and $dL_c(H)/L_c(0)$, a volume shrinkage associated with the IM transition is deduced, indicating a drastic suppression of the local lattice distortion associated with the transition from the localized to itinerant state.¹⁸ The resultant volume striction remains invariant down to zero field although its magnitude depends on the respective temperatures. In the insulating state of hole-doped bilayer manganites, there exists a lattice disorder between the Mn^{3+}O_6 and Mn^{4+}O_6 octahedra, with and without the JT local lattice distortion, respectively.¹⁹ As we pass T_c from the insulating state, the Mn-O bond length disorder is increasingly suppressed and, in the metallic state, it becomes less inhomogeneous than that found in the insulating state. Giant magnetostriction of the $z=0.6$ crystal is understood on the basis of a strong suppression of the local lattice disorder due to the field-induced FM phase, where the double-exchange interaction is favorable against the orbital-lattice interaction. Moreover, a polarized neutron diffraction study on the $z=0.6$ crystal suggests that the field-induced metallic state, where the field is parallel to the c axis, is characterized by a high population of the $d_{3z^2-r^2}$ orbital of Mn^{3+} .²⁰ This finding is consistent with the c axis distortion based on the local JT effect in the insulating state, preferring occupation of the $d_{3z^2-r^2}$ orbital through the orbital-lattice coupling.

Next, let us show in Fig. 2 the remanent c axis magnetostriction data of the $(\text{La}_{0.4}, \text{Pr}_{0.6})_{1.2}\text{Sr}_{1.8}\text{Mn}_2\text{O}_7$ single crystal as a function of time, just after the field was switched off. Here, the value of $dL_c(t)/dL_c(t_0)$ denotes the normalized $dL_c(t) = [L_c(t) - L_c(0)]$ by $dL_c(t_0) = [L_c(t_0 \ll 0) - L_c(0)]$ where $L_c(t_0 \ll 0)$ and $L_c(t=0)$ correspond to the virgin and initial values, before the application of field and just after the removal of field, respectively. If the system fully relaxes to the PM virgin state, $dL_c(t)$ should approach an equilibrium value of $dL_c(t_0)$. It should be noted that, at higher temperatures, above 30 K, the remanent striction instantaneously decays just after removing the field, while at low temperatures (5 K), the value of $dL_c(t)$ remains constant for 1 day. At 30 K, the lattice striction rapidly rises after removal of the field; then it relaxes by a half of the total value up to the relaxation time ($=1.7 \times 10^2$ s) and finally restores the ground-state value. Furthermore, at lower temperatures, it takes a much longer time for dL_c to decay to the ground-state value, as shown in Fig. 2. The temporal profile of the remanent magnetostriction follows a stretched exponential function $dL_c(t)/dL_c(t_0) = 1 - \exp[-(t/\tau)^\beta]$, where τ and β represent the characteristic relaxation time and exponent, respectively. The fitted parameters τ and β are plotted as a function of temperature in Fig. 3(a). Here, the temperature dependence of τ is well described by a thermally

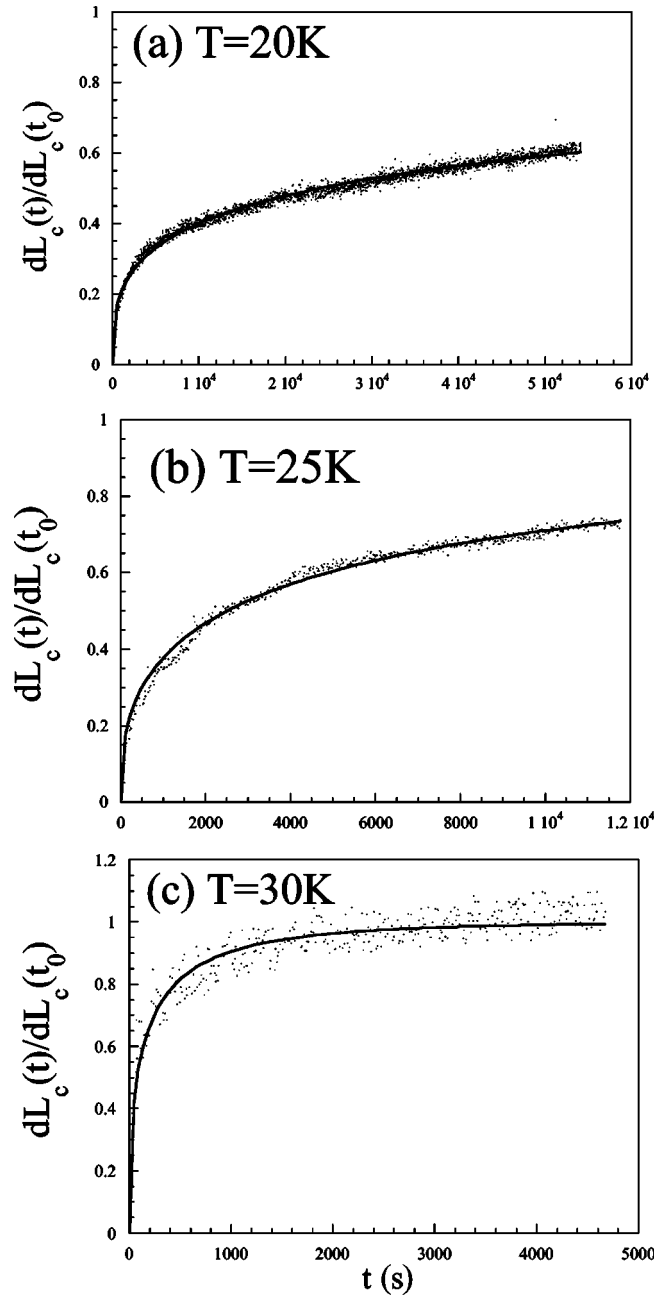


FIG. 2. The remanent c -axis normalized magnetostriction of single-crystalline $(\text{La}_{0.4}, \text{Pr}_{0.6})_{1.2}\text{Sr}_{1.8}\text{Mn}_2\text{O}_7$ as a function of time, just after the field was switched off. The solid lines correspond to a fit by a stretched exponential function, $dL_c(t)/dL_c(t_0) = 1 - \exp[-(t/\tau)^\beta]$, where τ and β represent the characteristic relaxation time and exponent, respectively. (a) $\tau = 6.9 \times 10^4$ s and $\beta = 0.35$ at 20 K, (b) $\tau = 6.0 \times 10^3$ s and $\beta = 0.42$ at 25 K, and (c) $\tau = 1.7 \times 10^2$ s and $\beta = 0.48$ at 30 K.

activated function $\tau = A \exp(\Delta/kT)$ where $A = 2.0 \times 10^{-2}$ and $\Delta = 30$ meV. Moreover, the value of the exponent is roughly fitted by a linear function of the form $\beta = 3 \times 10^{-2} + T/66.7$. Such a deviation from simple exponential decay needs further treatment on the basis of a multiple-relaxation-time approximation to describe the observed relaxation effect.²¹ The transition probability (τ^{-1}) from the FMM metastable to the PMI stable states is proportional to $\exp(-\delta U/kT)$, where δU

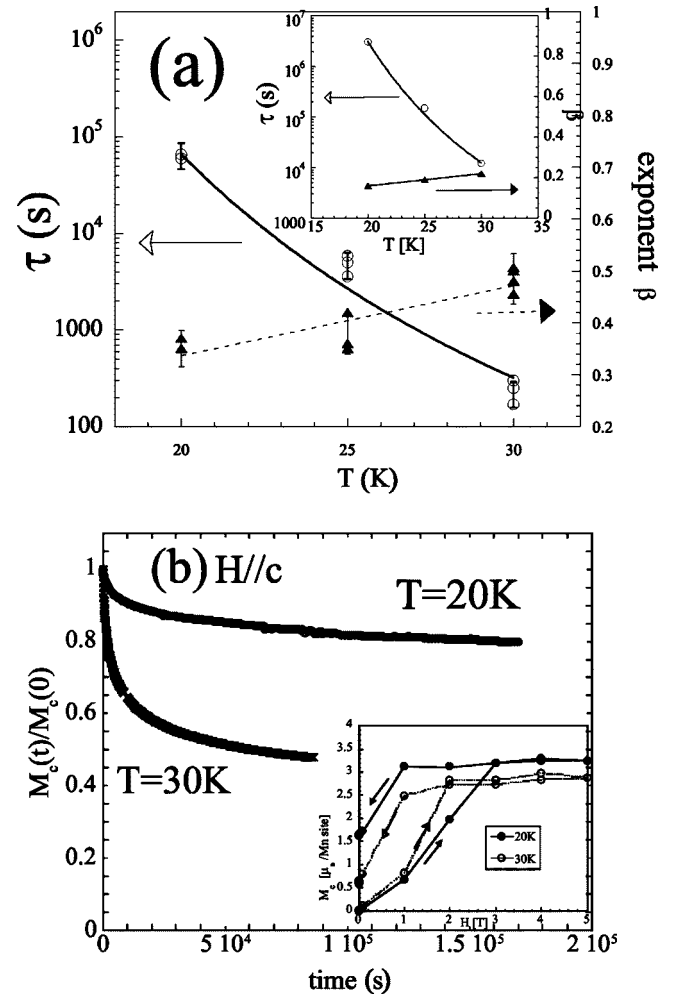


FIG. 3. (a) Remanent magnetostriction. Characteristic relaxation time and exponent τ and β , respectively, plotted as a function of temperature. The former is well fitted by a thermally activated function, while the latter follows a T -linear function. For comparison, in the inset of (a), the fitted parameters to remanent magnetization data are also shown. (b) c -axis normalized magnetization of the same crystal as a function of time after removing the field. $\tau = 3.1 \times 10^6$ s and $\beta = 0.16$, at 20 K, $\tau = 1.2 \times 10^4$ s and $\beta = 0.22$ at 30 K. In the inset of (b), the data collected for $M_c(H)$ before measuring the remanent magnetization are also shown.

represents potential barriers between the metastable state and the local maximum. A wide distribution of the potential barriers is related to a stretched exponential decay, leading to our understanding of the phase separation between FM and PM clusters. For comparison, the c axis magnetization of the same crystal is presented in Fig. 3(b) as a function of time after removing the field. In the inset of Fig. 3(b) are also shown the data collected for $M_c(H)$ before measuring the remanent magnetization. In the inset of Fig. 3(a), the parameters τ and β , obtained by fitting the magnetization data, are plotted as a function of temperature.²² In a similar way, the activation energy of the relaxation time is estimated to be 33 meV and the exponent is fitted to the T -linear function $\beta = 4 \times 10^{-2} + T/167$. We notice that the characteristic τ of L_c is about by two orders of magnitude smaller than

that of M_c on the same crystal, accompanied by a higher exponent.

Here, we comment on some differences in the relaxation processes between the magnetostriction and magnetization although both decay curves follow a similar stretched exponential function. First, from the viewpoint of a frustration among the competing interactions, both situations are different. The remanent magnetostriction slowly relaxes because of the competition between the double-exchange and JT-type lattice-orbital interactions. The former interaction causes a suppression in the local lattice distortion along the c axis through the itinerant state, but the latter favors a lattice deformation through the local John-Teller effect. On the other hand, in a previous study carried out on the $z=0.6$ crystal, the temperature variation of the magnetization exhibited a steep decrease in the zero-field-cooled (ZFC) scan below $T_g=28$ K, which is probably associated with a spin-glass state.⁸ It is thus expected from the spin-glass-like behavior of $M(T)$ that the decay process of remanent magnetization is closely related to a magnetic frustration between FM double-exchange and AFM superexchange interactions. Furthermore, we point out the discrepancy in the fitted relaxation time between $L(t)$ and $M(t)$ curves as shown in Fig. 3. A gradual drop in the $M(H)$ curve at 20 K with decreasing field is quite in contrast to the field-independent behaviors of $L(H)$ and $\rho(H)$ at low temperatures during the field sweep to zero.⁹ The similarity in remanent values between $L(H)$ and $\rho(H)$ data strongly suggests that the system still remains a metallic state with a volume shrinkage just after removing the field. As one of reasons for the difference between $M(H)$ and $L(H)$ [or $\rho(H)$], we think that the nucleation of magnetic domains in the metallic region probably gives a decrease in $M(H)$ at low fields.

Recently, Tokunaga has revealed the existence of magnetic domain structures in $(\text{La}_{0.4}, \text{Pr}_{0.6})_{1.2}\text{Sr}_{1.8}\text{Mn}_2\text{O}_7$ single crystals using a magneto-optical imaging technique.²³ It is

true that the formation of domain walls never produces a much longer time of the magnetic relaxation than the lattice relaxation. However, some of domain walls and/or each spin are pinned at lattice defect sites, giving a long lifetime of magnetic domains. Another scenario for the disagreement is concerning the existence of magnetic polarons in the insulating state, causing a magnetic contribution. In particular, in the hole-doped bilayer manganites, Zhou *et al.* proposed that Zener polarons occupying two Mn sites form a ferromagnetic cluster in the paramagnetic regime above T_c , to explain the pressure effect on thermoelectric power.²⁴ In addition, it should be noted that the Debye-Waller (DW) factor along the Mn-apical-oxygen bond in the insulating state is greater¹⁹ than the DW in the metallic one, at most by a factor of 2. In other words, the difference in the local lattice inhomogeneity between the metallic and insulating states is not as large as that in M between the ferromagnetic and paramagnetic states. Finally, we make some comments on the relaxation phenomena of remanent resistivity in manganites. Resistive relaxation studies support the view that the insulator-metal transition and its associated CMR effect are well described from the viewpoint of both the phase separation and its related percolation behavior of the metallic phase.^{11,25} The spatial growth of the local lattice distortions coupled with an e_g -electron orbital strongly correlates with the appearance of the field-induced CMR effect through the spatial evolution of percolating paths.

In summary, we have reported the time dependence of remanent magnetostriction in the Pr-substituted bilayer manganites. A frustration between double-exchange and Jahn-Teller-type orbital-lattice interactions plays a crucial role on the observed slow relaxation of lattice going from the field-induced FMM to PMI states.

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²² In the report of Gordon *et al.*, the estimated relaxation time was very short (10^{-2} s at 30 K) although the experimental value of $M_c(t)$ did not tend to decay during the estimated τ . Instead, τ is roughly expected to be of the order of 10^4 s from the M_c data in Fig. 3(b). If we try to fit our data with those of Gordon *et al.* using the same stretched exponential function $M(t)/M(0)=1-\exp[-(\tau/t)^\beta]$ then the characteristic relaxation time is estimated to be 1.2×10^4 s at 30 K, which is reasonable for a rough estimation.

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