Anomalous pressure effect on the remanent lattice striction of a $(La,Pr)_{1.2}Sr_{1.8}Mn_2O_7$ bilayered manganite single crystal

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We have studied the pressure effect on magnetostriction, both in the *ab* plane and along the *c* axis of a $(La,Pr)_{1.2}Sr_{1.8}Mn_2O_7$ bilayered manganite single crystal over the temperature region where the field-induced ferromagnetic metal (FMM) transition takes place. For comparison, we have also examined the pressure dependence of magnetization curves at the corresponding temperatures. The applied pressure reduces the critical field of the FMM transition and it enhances the remanent magnetostriction. An anomalous pressure effect on the remanent lattice relaxation is observed and is similar to the pressure effect on the remanent magnetization along the *c* axis. These findings are understood from the viewpoint that the double-exchange interaction-driven FMM state is strengthened by the application of pressure.

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I. INTRODUCTION

The observation of a phenomenon of a colossal magnetoresistance (CMR) effect has renewed the interest for doped manganites with perovskite structure.[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 was 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 of the DE interaction.[2,3] Furthermore, Dagotto *et al.* proposed a phase-separation model where ferromagnetic (FM) metallic and antiferromagnetic (AFM) insulating clusters coexist, which strongly supports recent experimental studies of the physics of manganites.[4]

Moritomo *et al.* have reported that the $La_{1.2}Sr_{1.8}Mn_2O_7$ bilayered manganite exhibits a paramagnetic insulator-(PMI) to-ferromagnetic metal (FMM) transition around $T_c \sim 120$ K and an associated CMR effect.[5] The Pr(Nd) substitution on the La site leading to $\{La_{1-z},Pr(Nd)_z\}_{1.2}Sr_{1.8}Mn_2O_7$ causes an expansion along the *c* axis but a shrinkage along the *a*(*b*) axis, resulting in a change of the e_g electron occupation from the $d_{x^2-y^2}$ to the $d_{3z^2-r^2}$ orbital.[6,7] These findings accompany not only a suppression of the PMI to FMM transition temperature T_c, but also a variation of the easy axis of magnetization from the *ab* plane to the *c* axis. For the $z=0.6$ crystal, the spontaneous ferromagnetic metal phase disappears at the ground state but the field-induced PMI to FMM phase is obtained over a wide range of temperatures. The magnetic phase diagram in the (H,T) plane, established from magnetic measurements carried out on the $z=0.6$ crystal, is presented in Fig. 1, with three regions labeled as PMI, FMM, and mixed states (white area). The white area is characterized by a hysteresis in the magnetization curves. The application of physical pressure is a powerful tool to investigate the lattice effect on magnetic and electronic properties of doped manganites as well as the *chemical-pressure* effect due to the other rare-earth ion substitution on the La site.[8] Many studies have been carried out so far on the effect of pressures on structural, magnetic and transport properties in bilayered manganites.[9–11] In half-doped bilayered manganite $La_{2-2x}Sr_{1+2x}Mn_2O_7(x=0.5)$, near the $x=0.4$ crystal studied here, a long-range orbital- and charge-ordered state appears over a limited temperature range between 100 and 210 K.[12] This finding is taken to be related to the polaronic state of the optimally doped crystal exhibiting the CMR effect through

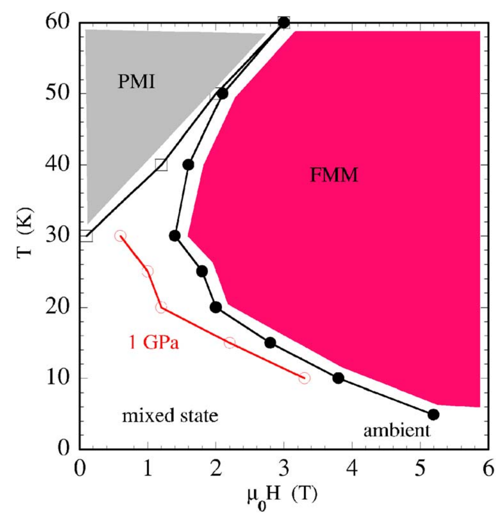

FIG. 1. (Color online) Magnetic phase diagram in the (H,T) plane established from the magnetic measurements carried out on the $z=0.6$ crystal $(H\|c)$. Three regions are distinguished and labeled as PMI, FMM, and mixed states (white area). The white area is characterized by a hysteresis in the magnetization curves. The phase-transition lines between PMI (or mixed phase) and FMM are defined as a maximum of dM/dH. For comparison, we show the PMI to the FMM phase-transition line under a pressure of 1 GPa determined from the pressure data of *MH* curves, as shown in the text.

an orbital frustration in the PI phase.[13] We report here the effect of pressure on magnetostriction, both in the *ab* plane and along the *c* axis, associated with the field-induced IM transition of a $(La_{1-z},Pr_z)_{1.2}Sr_{1.8}Mn_2O_7$ bilayered manganite single crystal. Just after removing the field, the system still remains in a metastable FMM state and it then comes back to the original PMI state through a mixed state made of FMM and PMI regions. Here, the FMM state lies in a local energy minimum and the PMI state is located in a global minimum of free energy, when the applied field is switched off. Next, we examine the giant pressure effect observed on the relaxation time of remanent magnetostriction. Furthermore, we compare the present results with the magnetic relaxation data on the $z=0.6$ crystal.

II. EXPERIMENT

Single crystals of $(La_{0.4},Pr_{0.6})_{1.2}Sr_{1.8}Mn_2O_7$ were grown by the floating-zone method using a mirror furnace. The calculated lattice parameters of the tetragonal crystal structure of the crystals used here were shown in a previous report.[14] The dimensions of the $z=0.6$ sample are 3.4×3 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. First, the sample was cooled down to the selected temperatures in zero field and we then started measuring the isothermal magnetostriction upon increasing (or decreasing) the applied fields at a sweep rate of 0.2 T/min. Finally, we recorded the isothermal remanent magnetostriction as a function of time just after switching off the field. Hydrostatic pressures in both magnetostriction and magnetization experiments were applied by a clamp-type cell using Fluorinert as a pressure-transmitting medium. The pressure was calibrated by the critical temperature of lead. The magnetization measurements were made with a superconducting quantum-interference device magnetometer both at Iwate University and NIMS.

III. RESULTS AND DISCUSSION

Figure 2 shows the *ab*-plane and *c*-axis magnetostrictions, $dL_a(H)/L_a(0)$ and $dL_c(H)/L_c(0)$, both under ambient pressure and a hydrostatic pressure of 0.8 GPa at different temperatures, where the applied field is parallel to the *c* axis $(H\parallel c)$. Here, the value of $dL_i(H)$ is defined as $L_i(H)-L_i(0)$. First, we observe that the *c* axis rapidly shrinks near 2 T upon applying the field, but the *a* axis expands at the same field value. The field-induced IM transition accompanies a stable decrease of the *c* axis by ~0.2%, in contrast with a small increase of the *a* axis by ~0.06%, resulting in a volume shrinkage of ~−0.08% $[dV(H)/V=2dL_a(H)/L_a(0)+dL_c(H)/L_c(0)]$ (Ref. 15). This value is in good agreement with the volume striction $dV/V\sim-0.09\%$ associated with the spontaneous IM transition of the parent compound $La_{1.2}Sr_{1.8}Mn_2O_7$, indicating that a volume shrinkage in the

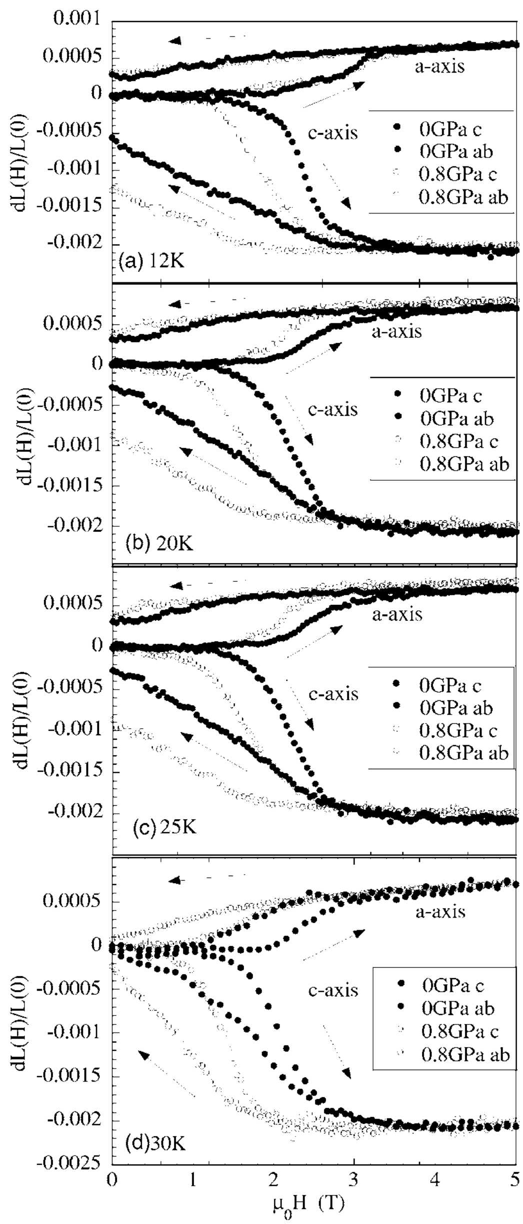

FIG. 2. The *ab*-plane and *c*-axis magnetostrictions $dL_a(H)/L_a(0)$ and $dL_c(H)/L_c(0)$, both under ambient pressure and a hydrostatic pressure of 0.8 GPa, at different temperatures (a) 12 K, (b) 20 K, (c) 25 K, and (d) 30 K. The applied field is parallel to the *c* axis $(H\parallel c)$.

metallic state is a consequence of the charge delocalization.[16,17] Here, the variations of the lattice parameters of the parent manganite, $\Delta a/a\sim-0.08\%$ and $\Delta c/c\sim0.07\%$, are estimated from neutron-diffraction measurements.[18] At selected temperatures, a clear hysteresis in the magnetostriction curves was observed, which has a close relationship to a memory effect in magnetoresistance, magnetization, and magnetothermal conduction of the $z=0.6$ crystal.[14,19,20] Next, the application of pressure to the magnetostrictions reduces the critical fields and also enhances the remanent magnetostriction just after removing the applied field. We show in Fig. 3 the pressure effect on the *ab*-plane and *c*-axis magnetostrictions $dL_a(H)/L_a(0)$ and

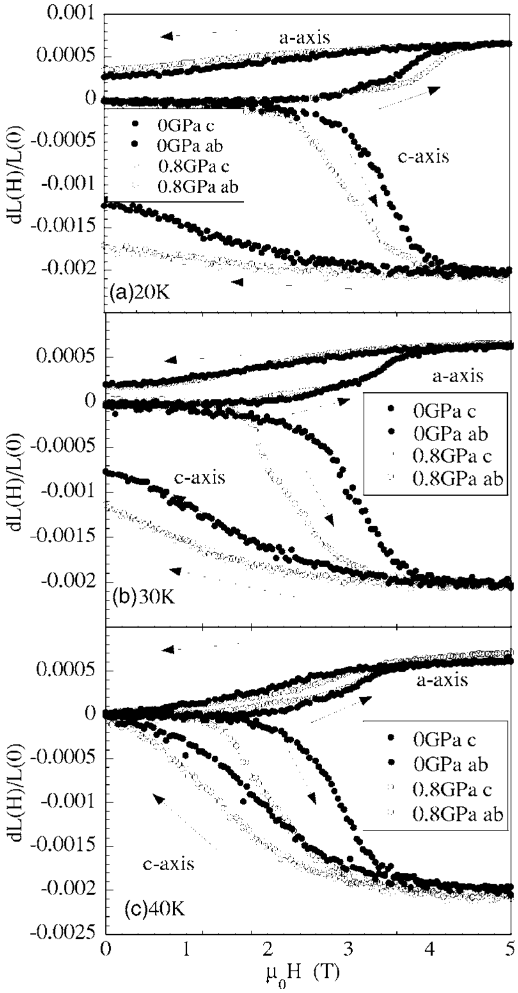

FIG. 3. The *ab*-plane and *c*-axis magnetostrictions $dL_a(H)/L_a(0)$ and $dL_c(H)/L_c(0)$, both under ambient pressure and a hydrostatic pressure of 0.8 GPa, at different temperatures (a) 20 K, (b) 30 K, and (c) 40 K. The applied field is parallel to the *ab* plane ($H \| ab$).

$dL_c(H)/L_c(0)$ in the case of the field applied in the *ab* plane ($H \| ab$). The magnetostriction behavior in $H \| ab$ is qualitatively similar to that in $H \| c$. We note that quantitative differences in dL/L between $H \| ab$ and $H \| c$ are higher critical fields and larger hysteresis regions in the former case. This finding is probably related to the easy axis of magnetization through the orbital occupation of the e_g electron between the $d_{x^2-y^2}$ and $d_{3z^2-r^2}$ states as mentioned below.

Now, we examine the *c*-axis magnetization under different pressures (0, 0.5, and 1.0 GPa) as shown in Fig. 4. For comparison, the *ab*-plane magnetization data are given in Fig. 5. Here, the value of $M_c(5\ \mathrm{T})$ reaches the $\sim 3.5\mu_B$/Mn site, close to the full moment of the $3.6\mu_B$/Mn site while the value of M_{ab} is $\sim 3.0\mu_B$/Mn site, which is by about 20% smaller than the ideal value.[14] The difference in the saturated magnetizations between M_{ab} and M_c at 5 T probably arises from the easy axis of magnetization along the *c* axis associated with the rise of the $d_{3z^2-r^2}$ orbital occupancy at $z=0.6$.[21] This finding is naturally understood by considering the importance of the spin-orbit interaction in the bilayered manganite system.[22] We note that the *ab*-plane magnetization exhibits no remanent value at all temperatures, in striking contrast with the substantial values of remanent magnetostriction, remanent magnetoresistance, and remanent magnetothermal conduction.[14,20] This discrepancy is not the central issue discussed here, but it has a close relationship with the formation of magnetic domains, keeping their local moments.[23] First, the application of pressure on magnetization suppresses the critical field inducing the PMI to FMM transition to the system, as well as the pressure effect on magnetostriction. The phase transition lines in Fig. 1 are defined as a maximum of dM/dH because the MH curves show metamagnetic behavior. At higher temperatures (30 K), it is true that the remanent magnetization is also enhanced upon increasing the pressure. However, at lower temperatures (15 and 20 K) the remanent M_c shows a rapid decrease with the

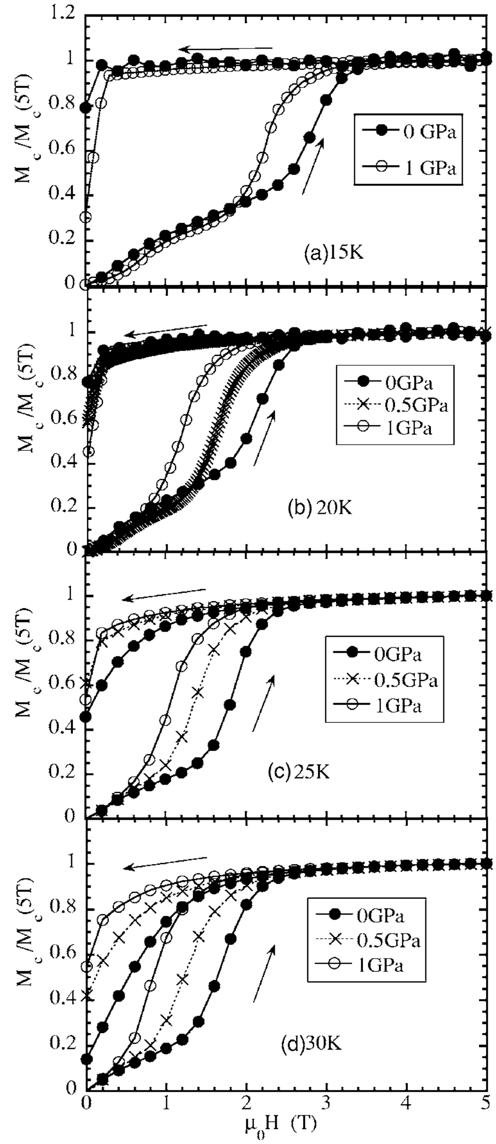

FIG. 4. *c*-axis magnetization under different pressures (0, 0.5, and 1.0 GPa) at selected temperatures (a) 15 K, (b) 20 K, (c) 25 K and (d) 30 K. The data are normalized by the saturated magnetization at 5 T.

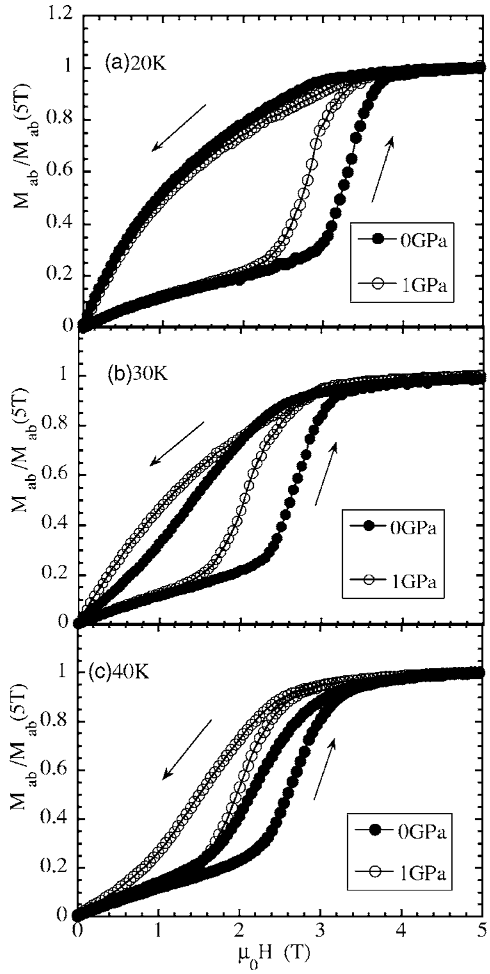

FIG. 5. *ab*-plane magnetization under different pressures (0 and 1.0 GPa) at selected temperatures (a) 20 K, (b) 30 K, and (c) 40 K. The data are normalized by the value of M_{ab} at 5 T.

applied pressures. The negative-pressure effect on remanent M_c, at lower T, contrasts with the positive-pressure dependence of remanent magnetostriction at the corresponding T. Recently, neutron-diffraction studies under pressure on bilayered manganite crystal with the same composition have shown similar results for the pressure influence on magnetization.[24] We comment now on the pressure effect on the MnO_6 octahedron sites in the parent bilayered manganite $La_{1.2}Sr_{1.8}Mn_2O_7$. Argyriou *et al.* have reported that upon increasing pressure up to 0.6 GPa, in the PMI state, both the Mn-O(1) and Mn-O(3) bond lengths shrink but the Mn-O(2) bond length elongates.[9] Here, the O(1) and O(2) oxygen atoms are located at the apical site along the c axis, where O(1) lies between two MnO_2 layers and O(2) within a rocksalt-type La/Sr-O layer. The O(3) oxygen atom is within the MnO_2 layer. Assuming such a variation of the MnO_6 octahedron in the PMI state of Pr-substituted $La_{1.2}Sr_{1.8}Mn_2O_7$, the Mn-O(1)-Mn interactions along the c-axis and the Mn-O(3)-Mn interactions in the ab plane are expected to be strengthened upon application of pressure. In other words, the double-exchange (DE) interaction between Mn^{3+} and Mn^{4+} within the MnO_2 single layer and the DE interaction along the c axis within the bilayer are enhanced and this is closely related to the decrease of the critical fields of the FMM state with pressure. On the other hand, the reciprocal response of Mn-O(2) to the applied pressures causes an elongation of the distance between adjacent bilayers, and it thus weakens the superexchange interactions between adjacent bilayers, keeping the DE interaction in the bilayer. This assumption describes well the negative-pressure effect on the remanent M_c at lower temperatures and is consistent with the positive effect on the remanent magnetostriction. Accordingly, the DE interaction-driven FMM state within a bilayer is enhanced with pressure but the SE interaction-driven ferromagnetic state between adjacent bilayers is suppressed.[24]

Finally, we examine the pressure effect on the relaxation time of remanent magnetostriction. Just after the removal of the applied fields, both the ab-plane and the c-axis remanent magnetostrictions are recorded as a function of time. The normalized remanent magnetostriction $d(c/a)/(c/a)$ is shown as a function of time in Fig. 6 where the anisotropic lattice striction $d(c/a)/(c/a)$ is estimated from the ab-plane and c-axis remanent data using $dL_c/L_c - dL_a/L_a$. The data points are normalized in such a manner that we make the initial value, just after the removal of field, to be zero, while the virgin value before the application of field becomes unity. In our case, the lattice variation along the c axis is by a factor of ~3 as large as the value in the ab plane and the value of $d(c/a)/(c/a)$ is almost taken as dL_c/L_c. As reported in previous papers,[19,25,26] the temporal profile of the remanent magnetization and magnetostriction follows a stretched exponential function $1-\exp[-(t/\tau)^{\beta}]$, where τ and β represent the characteristic relaxation time and exponent, respectively. The stretched exponential behavior of the relaxation curves indicates the existence of frustrations among competing interactions such as the FM and AFM interactions in spin-glass systems. We believe that the remanent magnetostriction shows a stretched exponential decay because of the competition between the double-exchange and JT-type lattice-orbital interactions.[25] The former interaction causes a suppression of the local lattice distortion along the c axis through the itinerant state, but the latter favors lattice deformation through the local JT effect.[17] Neutron-scattering measurements on the parent bilayered manganite $La_{2-2x}Sr_{1+2x}Mn_2O_7$ $(x=0.38)$ have revealed the presence of an orbital frustration in the paramagnetic insulating state, which prevents the system from the formation of a long-range orbital- and charge-ordered state.[13] We expect that this type of frustration survives for the Pr-substituted crystal with nearly the same doping and is relevant to the phenomenon of slow relaxation observed in remanent striction.

For a stretched exponential fit to the data points at 12 K, we get $\tau = 4.0 \times 10^5$ s under ambient pressure. Under a pressure of 0.8 GPa, τ becomes 3.7×10^7 s, which is much longer than the relaxation time without pressure. As a result, in Fig. 7, we summarize the relaxation time of both the remanent striction and remanent magnetization as a function of $1/T$. The application of pressures up to ~0.8 GPa to the system increases the relaxation time of the lattice by about two orders of magnitude, giving a more stable metallic state coupled with a suppression of MnO bond lengths in the MnO_6 local lattice. The relaxation data of remanent magne-

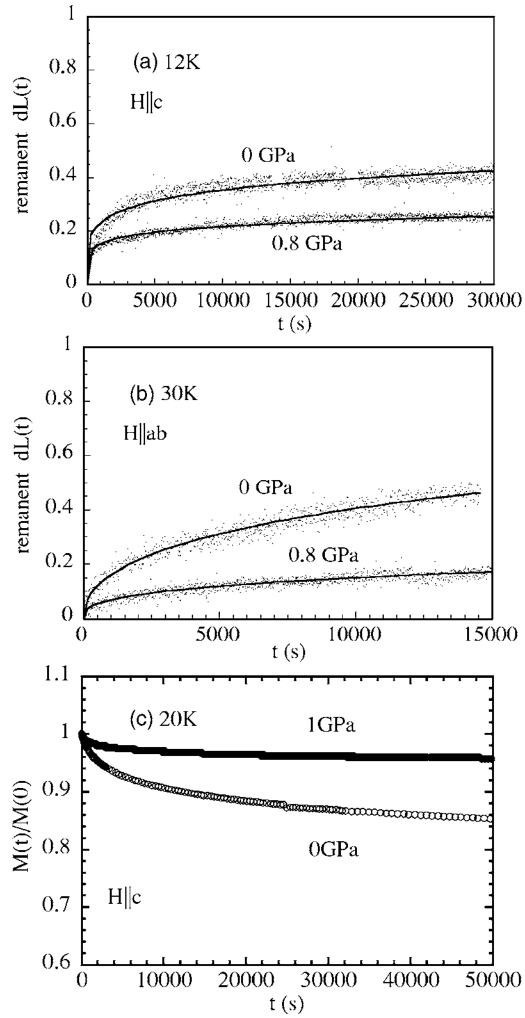

FIG. 6. (a) Normalized remanent magnetostriction $d(c/a)/(c/a)$ as a function of time at 12 K, just after the field along the c axis was switched off ($H \| c$). The solid lines correspond to a fit to the data points by a stretched exponential function $1-\exp[-(t/\tau)^{\beta}]$, where τ and β represent the characteristic relaxation time and exponent, respectively. In this case, we get $\tau=4.0\times10^5$ s with $\beta=0.22$ and $\tau=3.7\times10^7$ s with $\beta=0.17$, at 0 and 0.8 GPa, respectively. (b) The normalized remanent magnetostriction at 30 K, just after the field in the ab plane was switched off ($H \| ab$). $\tau=4.0\times10^4$ s with $\beta=0.47$ at 0 GPa and $\tau=1.4\times10^6$ s with $\beta=0.37$ at 0.8 GPa. (c) The normalized remanent magnetization at 20 K, just after the field along the c axis was switched off ($H \| c$) $\tau=3.1\times10^6$ s with $\beta=0.16$ at 0 GPa and $\tau=3.4\times10^9$ s with $\beta=0.1$ at 1.0 GPa. The data points are fitted using $M(t)/M(0)=1-\exp[-(\tau/t)^{\beta}]$.

tization under a pressure of 1 GPa at 20 K are presented in Fig. 6(c). In spite of the negative-pressure effect on the remanent M_c at lower T, as mentioned before [Figs. 4(a) and 4(b)], the relaxation time is strongly enhanced at 1 GPa by more than two orders of magnitude and shows a positive-pressure dependence. The difference in the relaxation time between remanent magnetization and magnetostriction can probably be explained by the fact that the magnetostriction observed here is not associated with a long-range order parameter such as the long-range orbital order reported in the

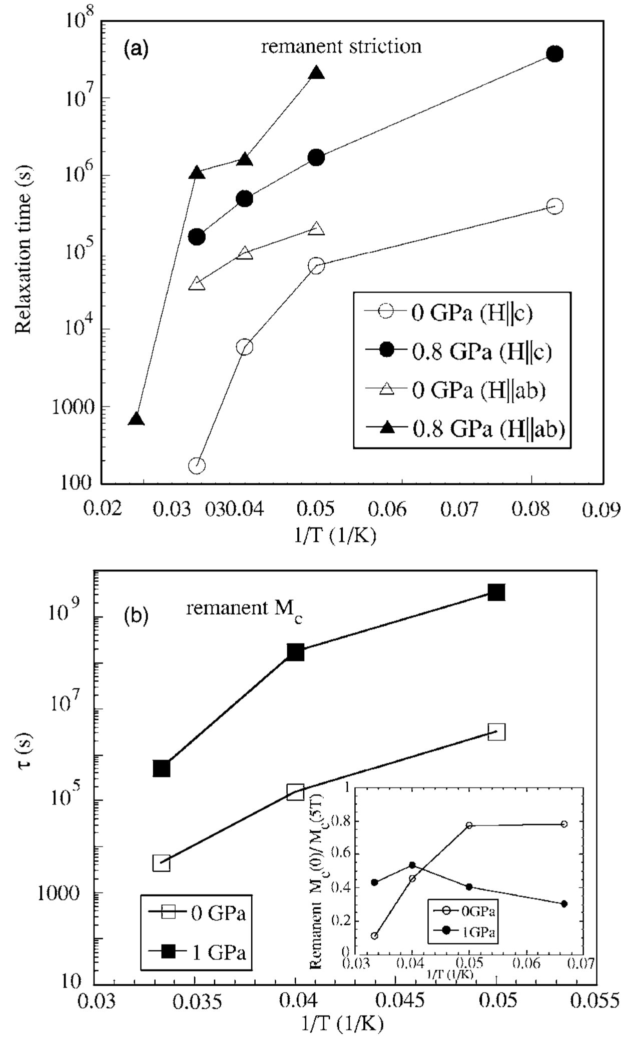

FIG. 7. (a) Pressure effect on the relaxation time of the remanent lattice striction as a function of $1/T$. (b) Pressure effect on the relaxation time of the remanent c-axis magnetization. For comparison, the inset represents the remanent M_c versus $1/T$ taken from the data in Fig. 4.

$x=0.5$ crystal. On the other hand, the magnetization is closely related to it.[26]

IV. SUMMARY

We have demonstrated the effect of pressure on magnetostriction, both in the ab plane and along the c axis, in a bilayered manganite single crystal of $(La,Pr)_{1.2}Sr_{1.8}Mn_2O_7$ over the temperature region where the FMM transition takes place. For comparison, we have examined the pressure dependence of the magnetization curves at the corresponding temperatures. The applied pressure reduces the critical field of the FMM transition and it also enhances the remanent magnetostriction. The quantitative differences observed in dL/L between $H \| ab$ and $H \| c$ are higher critical fields and larger hysteresis regions in the former case, which is probably related to the easy axis of magnetization along the c axis through the orbital occupation of the e_g electron between the

$d_{x^2-y^2}$ and $d_{3z^2-r^2}$ states. An anomalous pressure effect on the remanent lattice relaxation is observed to be similar to the pressure effect on the remanent magnetization along the c axis. In spite of the negative-pressure effect on remanent M_c at lower T, the relaxation time is strongly enhanced at 1 GPa by more than two orders of magnitude and shows a positive pressure dependence. These findings are understood considering that the double-exchange interaction-driven FMM state within a bilayer is strengthened upon application of pressure. Application of pressure reinforces the DE interaction, both in the *ab* plane and along the c axis, through a shrinkage of Mn-O(1) and Mn-O(3) bond lengths, resulting in a giant effect on the relaxation time of remanent striction and remanent magnetization. We believe that the slow relaxation observed here correlates to the orbital frustration existing in the paramagnetic insulating state of parent bilayered manganite.

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