

Elastic and magnetic properties of the bilayer manganese oxide (Pr_{0.6}La_{0.4})_{1.2}Sr_{1.8}Mn₂O₇

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The elastic and magnetic properties of a single crystal of the bilayer manganese oxide (Pr_{0.6}La_{0.4})_{1.2}Sr_{1.8}Mn₂O₇ have been investigated by means of ultrasonic and high-field magnetization measurements. Remarkable changes in the elastic constants C_{11} , C_{33} , C_{44} , and C_{66} as a function of temperature and magnetic field have been observed. In particular, a distinct elastic anomaly was observed at low temperatures and in magnetic fields when crossing the phase boundary between the paramagnetic insulating and the field-induced ferromagnetic metallic state. A pronounced elastic softening as a function of magnetic field (H) appears across the boundary of the low-temperature magnetic phase below around 40 K, accompanied by a distinct hysteresis. In the high-field region, however, these elastic constants exhibit a monotonic increase upon increasing the magnetic field. The high-field magnetization measurements characterizing the magnetic state point out a strong coupling between the elastic strain and the magnetic moment. The data can be described reasonably well considering a strong coupling between elastic strain and magnetic susceptibility $\chi_m = \partial M / \partial H$.

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I. INTRODUCTION

Manganites exhibit interesting original properties such as colossal magnetoresistance (CMR), giant magnetostriction, etc., which are thought to originate from a competition between charge, lattice, and spin degrees of freedom. Even though the fundamental mechanism of CMR is reasonably accounted for within the framework of the double exchange arising from the hopping of mobile e_g electrons between Mn³⁺ and Mn⁴⁺ ions,^{1,2} several other properties such as insulating ferromagnetic ground state, giant magnetostriction, magneto-thermal conductivity, etc., are not well understood. Among manganites, the double layer compound La_{2-2x}Sr_{1+2x}Mn₂O₇ has attracted considerable attention due to the enhanced CMR effect observed for $x=0.4$ and attributed normally to its quasi-two-dimensional structure,^{3,4} made of two MnO₆ layers alternately stacked with (La, Sr) O₂ layers along the c axis of the structure. Furthermore, it was shown that substitution of Pr³⁺ for La³⁺ at the concentration of $z=0.6$ in (La_{1-z}Pr_z)_{1.2}Sr_{1.8}Mn₂O₇ (hereafter denoted as PLSMO) resulted in a small contraction of the a parameter from $a=3.875$ Å to $a=3.863$ Å and an expansion of the c parameter from $c=20.13$ Å to $c=20.15$ Å. This gave rise to unexpected interesting properties. The ferromagnetic transition at $T_c=125$ K, accompanied by an insulator to metal transition observed in the compound with $z(\text{Pr})=0$, was completely suppressed in the compound with $z(\text{Pr})=0.6$. However, a magnetic field could induce a field-induced first-order

paramagnetic insulator to ferromagnetic metal transition in this latter compound. In addition, a huge decrease of the c -axis resistivity by a factor of 1×10^6 was observed at $T=5$ K in the presence of a magnetic field of 5 T.^{5,6} The first-order transition was also accompanied by a remarkable negative magnetoresistance when the field was oriented along the c axis, indicating the presence of an important interplay between spin, carrier, and orbital degrees of freedom.^{7,8} A polarized neutron study of this compound revealed an increase in the population of the $d(3z^2-r^2)$ orbitals of Mn³⁺ in the field-induced ferromagnetic state when the field was along the c axis.^{9,10} In order to investigate further this interplay, we have studied the elastic properties of this compound as a function of temperature and magnetic field. Ultrasonic sound velocity measurements were used to determine the elastic constants. We expected indeed that near such a field-induced phase transition, the elastic strain would couple to the fluctuation which, in the case of magnetic transitions, would produce noticeable changes in the sound velocity that would imply changes in the elastic constants. A preliminary report of our first results has been published in Ref. 11.

II. EXPERIMENT

The single crystal of PLSMO used for our experiments was grown by the floating-zone method using a mirror furnace and cut to a size of 5×4 mm² in the ab plane and

1 mm along the c axis.⁵ The sound velocity (v) was measured by the pulse echo method with carrier frequencies of 5 and 10 MHz for longitudinal and transverse waves, respectively. For precise measurement of the sound velocity, the constant phase method was introduced. The echo signals were multiplied by the in phase and quadrature phase references using a double-balanced mixer (DBM), and the two outputs of DBMs through a low pass filter were detected independently. The frequency was controlled to maintain a constant phase between the two outputs. The sound velocity change is thus obtained from the frequency change (phase comparison method). The resolution in the relative velocity was 10^{-6} in the present work. Plates of LiNbO_3 transducers were used for the generation and detection of the sound waves. The transducers were glued on the parallel faces of the sample by the elastic polymer Thiokol. We have performed, in addition, high-field magnetization (M) measurements with the help of a superconducting quantum interference device magnetometer operating between 1.8 and 300 K in magnetic fields up to 5 T and a vibrating sample magnetometer in magnetic fields up to 12 T and at temperatures down to 4.2 K.

III. EXPERIMENTAL RESULTS

A. Magnetization measurements

In this section, we briefly recall the magnetic properties of our sample and present a few additional measurements. In an earlier study, we have reported a cusp in the zero-field-cooled (ZFC)–field cooled curve around 40 K, in fields up to 5 T, that can be attributed to a spin-glass-like behavior.⁵ Significant differences were observed in the resistivity when it was measured along the c axis or in the ab plane, and the magnetization data revealed that the c axis was the easy axis of the magnetization. Figure 1 shows the magnetization of our sample at several temperatures ranging from 10 to 200 K as a function of magnetic field applied along the c axis. The data clearly indicate that, for $T < 100$ K, the magnetization shows an increase for $H > 4$ T, along the c axis. A sharp transition with a pronounced hysteresis is clearly visible at $T = 3.7$ K for a critical field of about 5 T, as shown in Fig. 2. These data clearly indicate a metamagnetic transition to a field-induced ferromagnetic state. The transition is observed at higher fields and becomes smeared at higher temperatures. It can also be noted that the transition is no longer accompanied by hysteresis above 50 K. Furthermore, the metamagnetic transition was undetectable at 200 K.

B. Ultrasonic measurements

In this section, we report the elastic property measurements of PLSMO. Figure 3 shows the temperature dependence of the relative changes in the longitudinal elastic constants C_{11} and C_{33} and the transverse elastic constants C_{44} and C_{66} in zero field. In order to avoid overlap and for clarity, the curves are shifted arbitrarily. We recall that C_{11} and C_{33} are the elastic constants determined by a longitudinal sound wave propagating along the a and c axes, respectively, and C_{44} and C_{66} are those determined by a transverse one

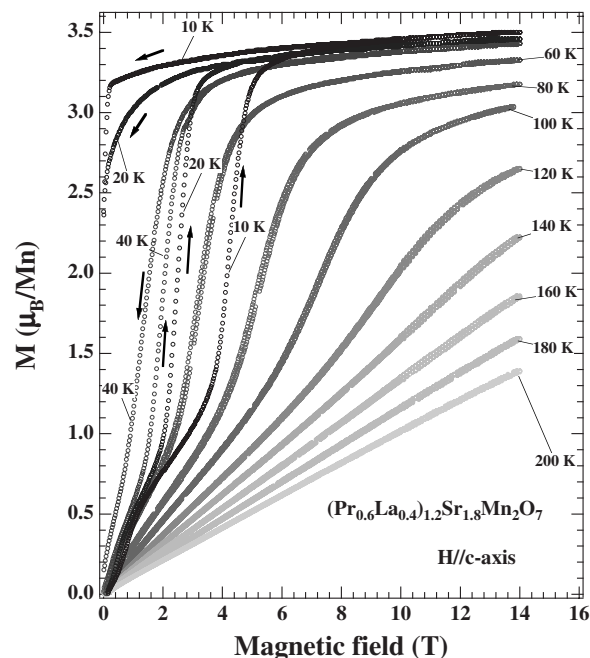


FIG. 1. Field dependence of the magnetization for a field applied along the c axis above 10 K.

propagating along the a axis with polarization along the c axes and propagating along the c axes with polarization along the a axes, respectively. Both increase monotonically with decreasing temperature. A slight anomaly is observed around $T^* = 40$ K and coincides with the earlier reported cusp in the magnetization data in low fields. However, a pronounced elastic anomaly was induced by application of magnetic fields. Figure 4 shows the field dependence of $\Delta C_{33}/C_{33}$ at selected temperatures. At 4.2 K, the value of $\Delta C_{33}/C_{33}$ decreases sharply at 6 T, and this is followed by a small increase for a further increase of the field. A strong

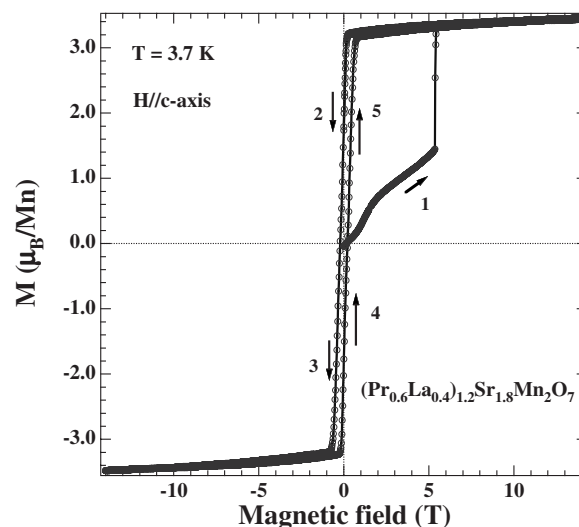


FIG. 2. Field dependence of the ZFC magnetization for a field applied along the c axis at 3.7 K. The label against each curve indicates the order of the magnetic field.

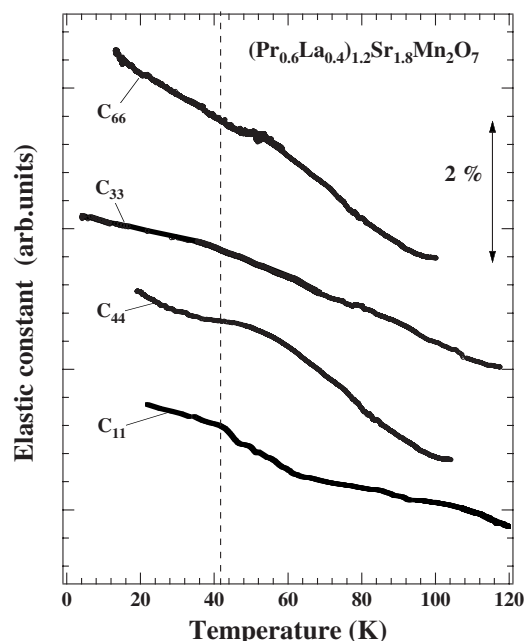


FIG. 3. Temperature dependence of elastic constants C_{11} , C_{33} , C_{44} , and C_{66} of LPSMO for $z=0.6$ in the absence of magnetic field. The dotted vertical line indicates the transition point determined by the resistivity measurements.

hysteresis, quite similar to that of the magnetic transition and indicative of a first-order transition, was observed when the field was decreased from 10 T to zero. The hysteresis was still observed for $T=30$ K. However, at $T=55$ K, no more

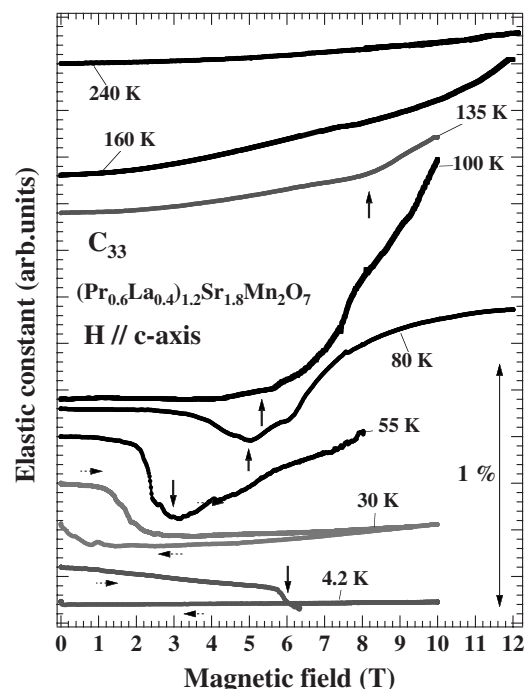


FIG. 4. Magnetic field dependence of the elastic constant C_{33} of PLSMO at selected temperatures. The vertical arrows indicate a transition point. The horizontal ones indicate the direction of the applied fields.

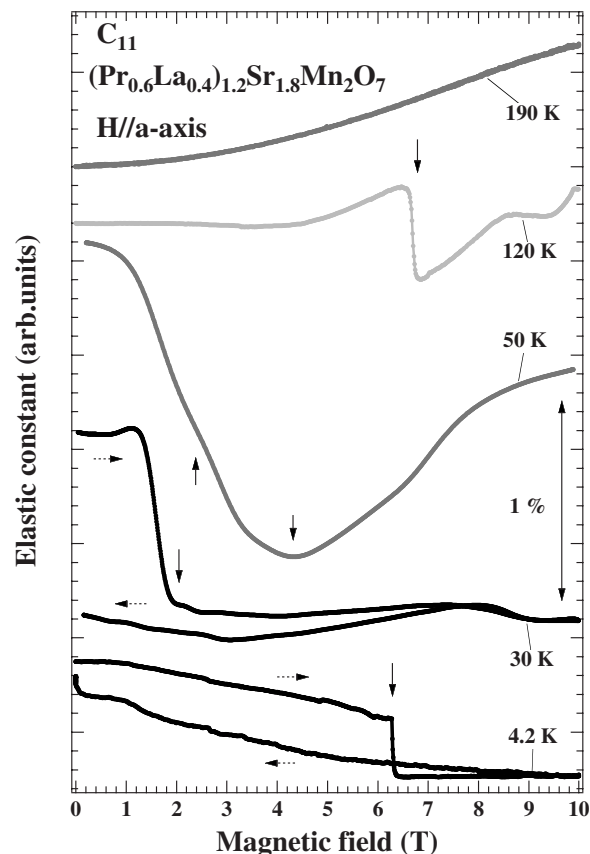


FIG. 5. Magnetic field dependence of the elastic constant C_{11} of PLSMO at selected temperatures. The vertical arrows indicate a transition point. The horizontal ones indicate the direction of the applied fields.

hysteresis was observed and the value of $\Delta C_{33}/C_{33}$ increased abruptly for $H < 3$ T. This increase was observed for $T < 135$ K. Figure 5 shows the field dependence of $\Delta C_{11}/C_{11}$ at selected temperatures. Although the general behavior was similar to that of $\Delta C_{33}/C_{33}$, additional anomalies were observed at 120 K and at 160 K. Figures 6 and 7 show the field dependence of $\Delta C_{44}/C_{44}$ and of $\Delta C_{66}/C_{66}$ at selected temperatures, respectively. The distinct elastic anomalies are also observed in the transverse elastic constant measurements. It is noted that C_{44} exhibits a sharp jump at the transition field, at 4.2 K, unlike C_{11} and C_{33} . Hysteresis effects are also observed. If we exclude these effects, we can say that the whole behavior around the transition field and also the elastic constants are qualitatively similar to those of longitudinal C_{11} and C_{33} .

C. Magnetic phase diagram

From these elastic and magnetic data, we can construct an (H - T) phase diagram for PLSMO, shown in Fig. 8. The transition points were determined by the inflection points at the M - H curves and H - T ones reported previously by our group. It is inferred from the observed features that there are, at least, three different phases I, II, and (III, III'). Regions I and II represent paramagnetic and field-induced ferromagnetic

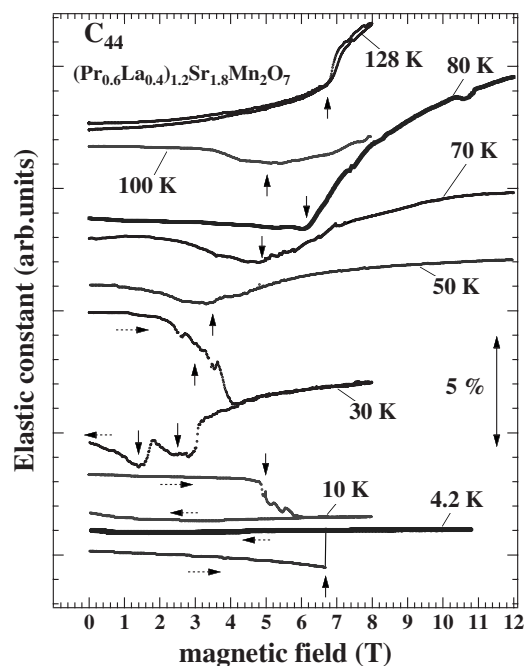


FIG. 6. Magnetic field dependence of elastic constant C_{44} of PLSMO at selected temperatures. The vertical arrows indicate a transition point. The horizontal ones indicate the direction of the applied fields.

phases, respectively. An almost full magnetic moment of $3.0 \mu_B/\text{Mn}$ is induced in phase II, where the system exhibits a metallic behavior of the resistivity. Thus, phase II is fully field-induced ferromagnetic phase. On the other hand, phase III is also considered to be a partial field-induced ferromag-

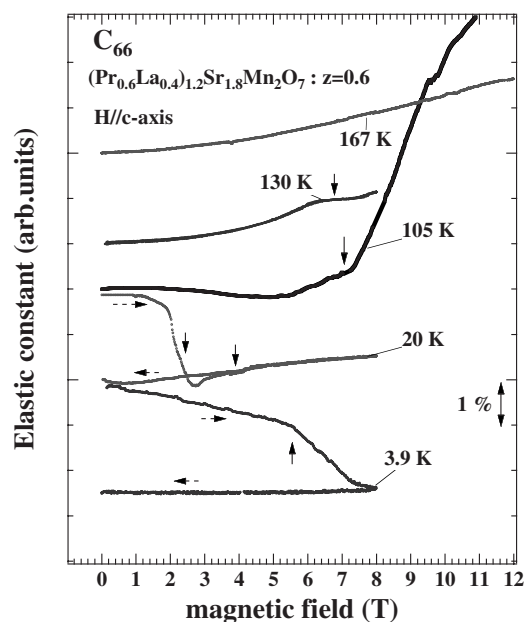


FIG. 7. Magnetic field dependence of elastic constant C_{66} of PLSMO at selected temperatures. The vertical arrows indicate a transition point. The horizontal ones indicate the direction of the applied fields.

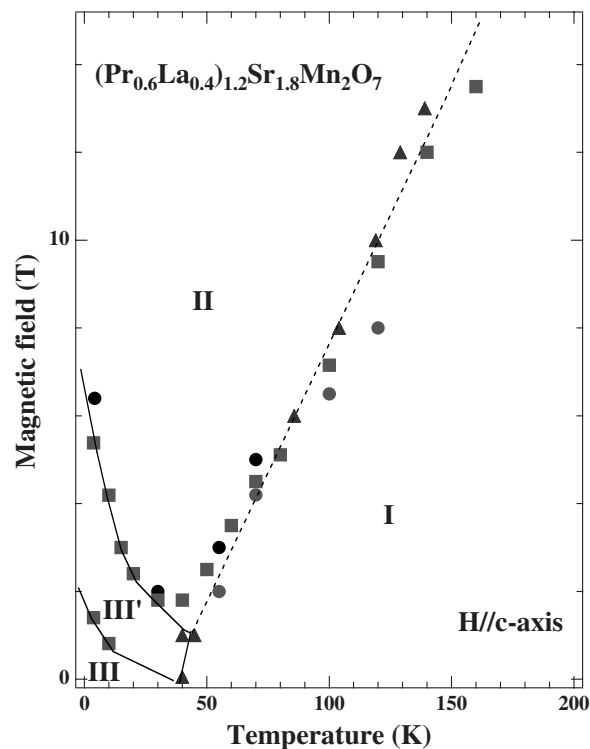


FIG. 8. $(H-T)$ magnetic phase diagram determined by the present results. The squares, triangles, and circles denote the transition points determined by the $M-H$ curves, $M-T$ curves, and elastic constant measurement, respectively. The solid and dotted lines are guides to the eye. The solid and dotted lines denote a phase boundary with or without hysteresis effect, respectively.

netic phase. There are two subregions in phase III where the boundary is defined only by the anomaly in the virgin $M-H$ and $C-H$ curves. It seems that this boundary may be due to domain rotation. It is noted that the boundary between I and III is poorly defined in zero field, whereas it becomes distinct, gradually with increasing field, indicating that phase III appears only under magnetic field. The transition is accompanied by a remarkable hysteresis in both magnetization and elastic constants at the boundary between phases III and II, indicating a first-order transition. In contrast, no hysteresis is observed between phases I and II, and given that the boundary between phases I and II ends at around 150 K, the transition is a first-order type. Although there is an insulator-metal transition on crossing the phase boundary between phases I and II, the symmetry of the system remains unchanged. Only on the zero magnetic field axis of the phase diagram there is a second-order phase transition, but that is between phases I and III. The qualitative features of the elastic constants can be summarized as follows. The elastic constant decreases when crossing the phase boundary from phase III to phase II, and a remarkable hysteresis is observed only in the virgin run upon increasing the field. However, it decreases gradually with increasing field in phase I and increases gradually after crossing the boundary from phase I to phase II, without any hysteresis. The temperature of $T^* = 40 \text{ K}$ is likely to be the critical temperature distinguishing these phases, although T^* is much less distinct in zero field.

Strong short-range-ordered spin correlations may arise around T^* . However, it seems that field application causes the stabilization of ferromagnetic long-range order, as will be discussed in detail below.

IV. DISCUSSION

The major findings of this work are the observation of the strong magnetic field dependence of the elastic constants C_{ii} for both longitudinal and transverse waves. This shows that the ordering of the magnetic moments clearly affects the elastic properties of the magnetic materials. A sudden transition is also observed in the longitudinal elastic constant C_{11} and in the transverse elastic constant C_{44} for field values around 7 T, at 4.2 K. Another interesting feature of these constants is the observation of an hysteresis as a function of applied magnetic field for temperatures less than or equal to 40 K. The magnetic field value at which the hysteresis occurs depends on the elastic constant and temperature. However, the magnetic moments M_c and M_{ab} did not show any transition as a function of applied magnetic field where hysteresis or a sudden softening of the elastic constants was observed. This could be due to the very different time scales of the excitation of the lattice and magnetic moments of the systems. In general, the elastic constants vary with magnetic field, suggesting appreciable interaction between magnetic field and lattice. It is worthwhile discussing the behavior of the magnetic susceptibility $\chi_m = \partial M / \partial H$, a quantity which is intimately related to a change in the value of the lattice constant with magnetic field, according to Refs. 12 and 13. For heavy fermion systems, it has been observed that the magnetic susceptibility $\chi_m = \partial M / \partial H$ is directly related to the softening of the longitudinal modes according to the relation

$$\Delta C_\Gamma = -\Gamma_B^2 H^2 \chi_m, \quad (1)$$

where $\Gamma_B = -\partial \ln Hc / \partial \epsilon_\Gamma$ is the Grüneisen parameter and ϵ_Γ is the elastic strain with Γ symmetry. We verify this relation by estimating Γ_B^2 from the plots of $\chi_m H^2$ and $\Delta C_{33} / C_{33}$ as a function of magnetic field as shown in Figs. 9 and 10, respectively, for different values of temperature. We find that the values of Γ_B^2 calculated at the critical field is almost constant or varies over a very small range for all the temperatures. Although some of their quantitative aspects do not agree well between the experimental results and the calculated ones, we emphasize that this agreement gives evidence for a strong spin-lattice coupling in PLSMO above $T^* = 40$ K. The expected upturn of the elastic constants by $-\chi_m H^2$ above H_c , below 40 K, does not appear in the measured elastic constants, which is expected from Eq. (1). One scenario to explain the discrepancies is that they result from a change of the carrier number in the field-induced metallic phase, as will be discussed in detail below.

Obviously, the physics of these materials is dominated by the double-exchange process, electron-phonon coupling, and the electron kinetic energy represented by the bandwidth of the e_g band. The hopping of the e_g electrons, which promotes the ferromagnetic order, is affected by the values of the lattice constants in addition to the relative alignment of the core spins. The Jahn-Teller effect involves the simultaneous split-

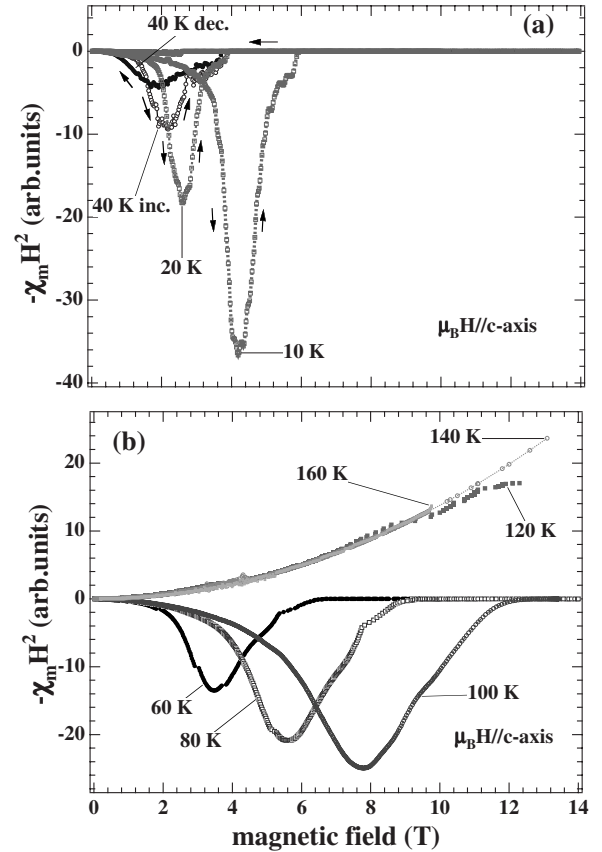


FIG. 9. Field dependence of $-\chi_m H^2$ (a) below the critical temperature of 40 K and (b) above 40 K. The dotted arrows indicate the direction of the applied fields. 40 K inc. and 40 K dec. denote a result at 40 K in the increase and decrease processes of the magnetic field, respectively.

ting of the electronic states and the symmetry lowering distortion of the lattice. The distortion of the MnO_6 octahedra couples the magnetic and lattice vibrations, resulting in the fact that electronic, lattice, and magnetic degrees of freedom are intimately connected. This affects both the exchange interaction and the crystal field potential. The coupling changes the single particle electron state energy E_k^0 , and, according to the deformation potential coupling theory, the modified energy is given by

$$E_k(\epsilon) = E_k^0 + d_\Gamma(k)\epsilon_\Gamma, \quad (2)$$

where d_Γ is the deformation potential coupling constant with Γ symmetry.^{14–17} The index Γ denotes the irreducible representation in the tetragonal symmetry.¹⁸ The free energy F_{el} for the mobile electrons can be written as

$$F_{el} = nE_F - k_B T \sum_k \ln \left(1 + \exp \left(\frac{(E_F - E_k)}{k_B T} \right) \right). \quad (3)$$

Here, E_F and n denote the Fermi energy and the number of the mobile electrons in the e_g band, respectively.

Using Eqs. (2) and (3), the elastic constant $C_\Gamma = \partial^2 F_{el} / \partial \epsilon_\Gamma^2$ for a wave vector k -dependent deformation potential coupling constant can be written as

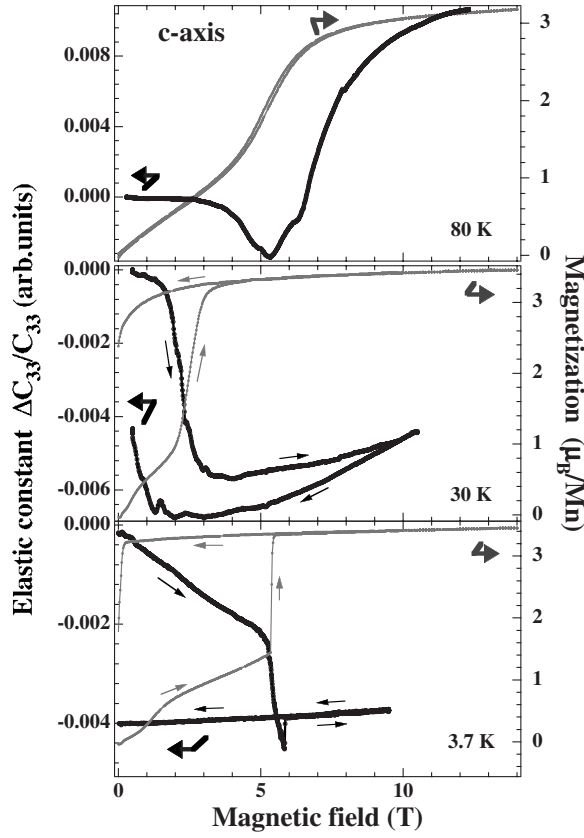


FIG. 10. Comparison of the obtained elastic constants with the magnetization at 3.7, 30, and 80 K, respectively.

$$\Delta C = -\frac{1}{k_B T} \sum_k d_\Gamma(k)^2 f_k (1 - f_k) + \frac{1}{k_B T} \left(\sum_k d_\Gamma(k)^2 f_k (1 - f_k) \right)^2 / \sum_k f_k (1 - f_k), \quad (4)$$

where $\Delta C_\Gamma = C_\Gamma - C_\Gamma^0$ and C_Γ^0 is the elastic constant without magnetic field. In the expression of $\partial^2 F_{el} / \partial \epsilon_\Gamma^2$, the $\sum_k (\partial F_{el} / \partial \epsilon_\Gamma)^2 f_k$ term is zero due to the linear dependence of the dispersion $E_k(\epsilon) = E_k^0 + d_\Gamma(k) \epsilon_\Gamma$ on ϵ_Γ . For a single band system, no changes in the elastic constants will be observed for a k -independent $d_\Gamma(k)$. However, for a two-band model, if $d_\Gamma(k)$ is independent of k [i.e., $d_\Gamma(k) = d_\Gamma$], ΔC_Γ is nonzero only if each band has a different value of d_Γ . From the general equation (4), it is clear that for a dispersive $d_\Gamma(k)$, which is the case with $(\text{Pr}_{0.6}\text{La}_{0.4})_{1.2}\text{Sr}_{1.8}\text{Mn}_2\text{O}_7$, the sudden changes in the elastic constants arise from the occupation probability functions which are affected when d band splits into two bands due to the Jahn-Teller interaction. The electron-phonon coupling originates from the Jahn-Teller splitting of the d levels of the Mn ions whose electronically active orbitals are $d(x^2 - y^2)$ and $d_2(3z^2 - r^2)$, and we assume that these orbitals form a two-band model leading to the “band-Jahn-Teller effect.” A sudden change in the occupancy level of electrons in these bands upon application of a magnetic field could lead to a sudden change in the values of the elastic constants. The resistivity of these materials did not show any

sudden transition¹⁹ with magnetic field, suggesting that the sudden transition in the elastic constants may not be the result of e_g electron interaction with the lattice. The decrease of the resistivity upon increasing the magnetic field leads to the decrease of the elastic constant values as described by Eq. (4). It is inferred that this may suppress the upturn in the field dependence of the elastic constant just above H_c , expected from Eq. (1), as shown in Fig. 9. Unfortunately, we cannot discuss these changes quantitatively at present since the absolute values of the elastic constants were not determined. Nevertheless, we suggest that the elastic anomaly and the absence of the upturn of the elastic constant above H_c , below T^* , are ascribable to the coupling effect between the relevant elastic strain and the magnetic moment and between the elastic strain and the mobile Mn e_g electrons.

Next, we would like to comment on the low-temperature and low-field magnetic phase III, shown in Fig. 8. The characteristic features observed may suggest that this phase is fairly metastable. Figures 4–6 give typical examples to describe this situation. Actually, oscillatory phenomena are observed for C_{33} at 30 K, C_{11} at 30 and 50 K, and C_{44} at 10 and 30 K, below H_c . The oscillations disappear above H_c and at higher temperatures. We conjecture that these oscillations can be related to the vicinity of a magnetic instability, such as a spin-glass state, since the transition is accompanied by a prominent hysteresis in both the ultrasonic and magnetization measurements. However, this is somehow unusual, because a slight anomaly is observed in zero field. The spin-glass-like behavior shows up only by applying magnetic field. The critical temperature in zero field seems to be around $T^* = 40$ K by interpolation of the boundary between phases I and III if we assume that the transition line continues linearly with magnetic field. A slight anomaly was observed in both the temperature dependence of the elastic constant and magnetization curve at T^* . It is interesting to compare the magnetic properties of the compounds $(\text{La}_{1-z}\text{Nd}_z)_{1.2}\text{Sr}_{1.8}\text{Mn}_2\text{O}_7$ and $(\text{La}_{0.8}\text{Gd}_{0.2})_{1.4}\text{Sr}_{1.6}\text{Mn}_2\text{O}_7$, reported previously by Moritomo *et al.*²⁰ and Dho *et al.*,²¹ respectively. The prominent features of their phase diagrams correspond to those obtained in $(\text{La}_{1-z}\text{Nd}_z)_{1.2}\text{Sr}_{1.8}\text{Mn}_2\text{O}_7$ ($z = 0.4$) at low temperature. In comparison, phase III is presumably due to a metastable region where the parain insulator and ferrometal states can coexist. That is to say ferromagnetic clusters can be grown by application of a magnetic field. This phase was understood within the framework of spin degree of freedom, so far. Our previous results for PLSMO have indicated that the magnetostriction (MS) along the c axis and in the ab plane showed a sudden decrease and increase at the transition field, respectively.^{7,8} The formation of domains due to bound carriers is also suggested by recent optical conductivity measurements on the compound $(\text{Pr}_{0.6}\text{La}_{0.4})_{1.2}\text{Sr}_{1.8}\text{Mn}_2\text{O}_7$.²² The measurements did not show any metallic behavior even for magnetic fields up to 10 T in the ferromagnetic metallic state, but, instead, a far-infrared bound carrier excitation was inferred.

Furthermore, it should be noted that the transition is accompanied by a prominent hysteresis in MS. This implies that an orbital degree of freedom plays a crucial role in this ordered phase III as well. The inflection points in the elastic constant for field-increasing and field-decreasing runs shift

toward low fields with increasing temperature in phase III, leading to a reduction of the field at which hysteresis sets in. Classical thermodynamics indicates that the first-order phase transition of the metastable state can occur in the condition that the potential barrier energy between two states becomes comparable with the temperature.²⁰ Thus, it becomes difficult to shift from the metastable state to the other upon decreasing the temperature. This energy difference may correspond to $T^* = 40 \text{ K} = 3.3 \text{ meV}$. The data relative to the elastic constants and MS as a function of field imply that the paramagnetic and ferromagnetic states are different. As was seen in $\text{La}_{1.2}\text{Sr}_{1.8}\text{Mn}_2\text{O}_7$, the e_g state is dominated by $d(x^2 - y^2)$ in the ferromagnetic phase, whereas some of the $d(3z^2 - r^2)$ population mixes with the $d(x^2 - y^2)$ component in the paramagnetic phase. Recently, this behavior was confirmed microscopically by neutron scattering measurements.^{23,24} It seems that a spatial disorder of the orbital state may arise as well, in addition to this magnetic disorder in this system.

V. CONCLUDING REMARKS

We have performed magnetization and ultrasonic measurements on a single crystal of $(\text{Pr}_{0.6}\text{La}_{0.4})_{1.2}\text{Sr}_{1.8}\text{Mn}_2\text{O}_7$ as a function of temperature and magnetic field. A pronounced

elastic anomaly was observed when crossing the boundaries of the phase diagram. In the absence of a microscopic theoretical descriptions, we proposed a phenomenological interpretation of our data in terms of the coupling between elastic strain and $\chi_m = \partial M / \partial H$. Various phases in the $(H-T)$ magnetic phase diagram of this system were discussed, and we observed a field-induced magnetic phase which exhibited hysteresis. Characteristic features of this phase suggest a strong coupling between the spin, orbital, and lattice of Mn ions degrees of freedom. It is noteworthy that the remarkable hysteresis in the elastic constants and also in magnetostriction was observed in the vicinity of the boundary between phases III' and II.

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