FIELD DEPENDENCE OF ELASTIC CONSTANTS IN THE BILAYER MANGANITE: $(La_{1-z}Pr_z)_{1.2}Sr_{1.8}Mn_2O_7$ FOR $z = 0.6$*

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Elastic properties of the Pr-doped bilayer manganite: $(La_{1-z}Pr_z)_{1.2}Sr_{1.8}Mn_2O_7$ for z=0.6 was investigated by means of the ultrasonic measurement. No remarkable anomaly was observed around the transition temperature in the temperature dependence of C_{33} in zero field. A pronounced elastic anomaly, however, has been observed around the magnetic phase transition field Ht in the longitudinal elastic constants C_{11}, indicating the phase can be induced in magnetic fields. The transition accompanies a large hysteresis, implying the ordered state to be so-called "orbital-glass state". The origin of observed elastic anomalies are discussed in terms of the coupling between elastic strains and magnetic moments of Mn ions, and a change of carrier numbers.

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Recently much attention is being focused on the bilayer manganites with general formula $(La_{1-z}Pr_z)_{1.2}Sr_{1.8}Mn_2O_7$: abbreviated to LSMO(327), as well as other manganites [1–5]. Because the largest magnitude of Metal–Insulator (MI) transition was found in their family of the Ruddlesden–Popper series $(R, D)_{n+1}Mn_nO_{3n+1}$. The feature of LSMO(327) with n=2 is that the system has two-dimensional network in MnO_2 sheets with inserted blocking layers $(La_{1-x}Sr_x)_2O_2$ leading to significant magnetic fluctuations compared to three-dimensional one (n=∞). This two-dimensionality is believed to play an important role in the enhancement of the MI transition and the colossal magnetoresistance (CMR). In particular, LSMO(327) for x=0.4 exhibits the largest MI transition and CMR effect near T_c'125 K [1].

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Furthermore, it has been found that these effects were significantly enhanced in the bilayer manganites by the substitution of the smaller Pr atom at the La site. As a result the drastic change of the resistivity by a factor of one million can be seen in magnetic fields [6–7]. With increasing the Pr concentration z, the MnO_6 octahedra elongate along the c-axis both at room temperature and in the ground state, leading to a lowering T_c. This implies the increase of the $d(3z^2 - r^2)$ orbital polarization, and suppress the planar ferromagnetic interaction. The ferromagnetic phase seems to be vanished around $z = 0.5$. Thus, it is expected that a field-induced magnetic phase appears, and it seems to cause the extraordinarily large CMR effect. In this system, it is also pointed out that electron-lattice coupling is strong and plays an important role. This will cause a large elastic anomaly around the phase transition points. In this paper, we present the elastic properties of Pr-doped bilayer manganite : $(La_{1-z}Pr_z)_{1.2}Sr_{1.8}Mn_2O_7$ for z=0.6 in magnetic fields.

Single crystals of $(La_{1-z}Pr_z)_{1.2}Sr_{1.8}Mn_2O_7$ for z=0.6 were grown from sintened rods of the same nominal composition by the floating-zone technique by using a minor furnace. The dimension of the sample were 5×4 mm in the ab-plane and 1 mm along the c-axis. In order to observe the ultrasonic pulse echoes with an exponential decay, we polished carefully the surfaces of the sample to be parallel. The $LiNbO_3$ transducers for the generation and detection of the sound waves with frequencies 5'10 MHz were bonded on the surfaces of the sample by an elastic polymer Thiokol. The sound-wave velocity v was detected by an ultrasonic apparatus based on the phase-comparison method. The magnetic field up to 12 T was generated by a superconducting magnet.

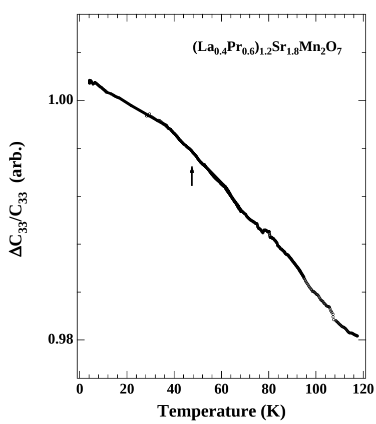

Fig. 1. Temperature dependence of the longitudinal elastic constant C_{33} of $(La_{1-z}Pr_z)_{1.2}Sr_{1.8}Mn_2O_7$ for z=0.6 in zero field. An arrow indicates the temperature where the resistivity shows the kink.

The selected longitudinal elastic constant C_{33} in the present study, which propagates along c-axis, exhibited a much more remarkable anomaly than that of other elastic constants in our previous studies of LSMO(327) for x=0.35 and 0.4 [8]. Figure 1 shows the temperature dependence of longitudinal elastic constant C_{33} in zero field. They increase monotonically with decreasing the temperature. No anomaly was observed around 120 K at which the resistivity exhibits a kink implying appearance of a kind of phase transition [6–7]. Figure 2 shows the magnetic field dependence of C_{33} at the selected temperatures. The remarkable elastic anomalies appeared in magnetic fields. At 4.2 K, C_{33} exhibits an abrupt decrease around 6 T, where a meta-magnetic transition occurs in the magnetization curve [6–7]. This anomaly becomes larger and shifts to lower magnetic fields with increasing temperatures up to 50 K. However, it becomes smaller and shifts to higher magnetic fields with increasing temperatures above 50 K by contrast. It is noteworthy that the field-induced transition accompanies a large hysteresis below 50 K.

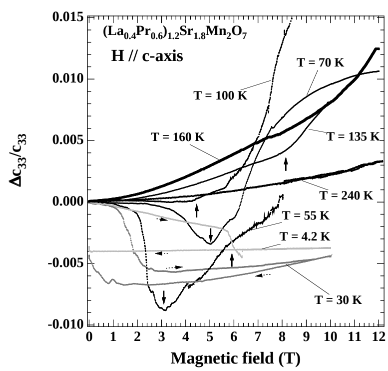

Fig. 2. Magnetic field dependence of C_{33} in $(La_{1-z}Pr_z)_{1.2}Sr_{1.8}Mn_2O_7$ for z=0.6 at selected temperatures. A solid arrow indicates the phase transition point. A dotted arrow indicates process of the measurement.

Here, we discuss the present results. The fact, that no elastic anomaly has been observed at 50 K in zero field, is consistent with a result of the magnetic susceptibility [9]. No or quite small magnetic anomaly has been observed around 50 K in the magnetic susceptibility. However, the anomaly became gradually larger by applying magnetic fields, and came to accompany a large hysteresis. The behavior of the field-cooled magnetization below the transition temperature is quite different from that of zero-field-cooled one, possibly indicating the presence of a spin-glass-like state or the coexistence of antiferromagnetic and ferromagnetic cluster [6–7]. Simultaneously, the resistivity showed the semiconductor like behavior with decreasing temperature in zero field, however, the drastic decrease appeared around the field-induced transition temperature [6–7]. These previous results indicate phenomenologically that the field-induced magnetic moment can decrease the resistivity, significantly, as being seen also in LSMO(327) for x=0.35 and 0.4. In fact, the huge negative magnetoresistance was observed at the temperature below 50 K with a large hysteresis [6–7]. As pointed out in our previous paper of LSMO(327), a coupling between elastic stress and magnetic moment causes the elastic hardening [8]. Thus, the observed hardening of C_{33} as a function of magnetic fields above the transition field at the temperature below 50 K is ascribable to the coupling between elastic stress and field-induced magnetic moment. On the other hand, an increase of the carrier number can bring about elastic softening. It is because that the effective charge of ions can be decreased more by the screening effect of additional conduction electrons [8].

Finally, we comment on the accompanying large hysteresis. A large hysteresis observed in elastic constant C_{33} possibly indicates a orbital-glass like state below 50 K. Since the orbital ordering of 3d-levels in Mn ions restricts the magnetic degree of freedom, the spin glass-like behavior can result. Actually, the magneto-striction as a function of magnetic fields below 50 K also shows a remarkable anomaly with a large hysteresis [6–7]. As considering this fact and our results, we can suggest the presence of orbital glass-like state below 50 K.

In summary, we performed the ultrasonic measurement on $(La_{1-z}Pr_z)_{1.2}Sr_{1.8}Mn_2O_7$ for z=0.6. A remarkable anomaly was observed in C_{33} as a function of magnetic fields below 50 K. A large softening/hardening is considered to be due to the coupling between the elastic stress and carriers/magnetic moment, respectively. The same study with different elastic constants for z=0.6 and different concentration for z=0.2 and 0.4 is currently in progress.

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