Ultrasonic Study of Orbital and Charge Orderings in $La_{1-x}Sr_xMnO_3 (x = 1/8)$

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Abstract

In order to investigate orbital and charge orderings in perovskite manganite $La_{1-x}Sr_xMnO_3$ with $x = 1/8$ (Mn^{3+} : $Mn^{4+} = 7 : 1$), we have measured elastic constants by ultrasonic experiments. $La_{0.875}Sr_{0.125}MnO_3$ shows successive structural phase transitions at $T_s = 275$ K and $T_{co} = 150$ K. Above T_s, the elastic constant $(C_{11} - C_{12})/2$ exhibits a remarkable softening, while C_{44} shows a monotonous increase with decreasing temperature. The softening of $(C_{11}-C_{12})/2$ arises from the coupling of quadrupole moment of e_g orbital in Mn^{3+} ion to elastic strain. Furthermore, the $(C_{11} - C_{12})/2$ and C_{44} exhibit a considerable softening above T_{co}, which is caused by the coupling of charge fluctuation associated with the distribution of Mn^{3+} : $Mn^{4+} = 7 : 1$ to elastic strain.

Key words: quadrupole ordering, charge ordering, elastic constant

Perovskite manganite $La_{1-x}Sr_xMnO_3$ ($x = 1/8$) shows successive structural phase transitions at $T_s = 275$ K and $T_{co} = 150$ K and a ferromagnetic phase transition at $T_C = 200$ K [1]. These orderings originate in the spin, charge (Mn^{3+}, Mn^{4+}) and orbital ($d(3z^2 - r^2)$, $d(x^2 - y^2)$) degrees of freedom in $La_{0.875}Sr_{0.125}MnO_3$. The structural phase transition at T_s is accompanied by the Jahn-Teller distortion from pseudo cubic to pseudo tetragonal due to the e_g orbital in Mn^{3+} ion. While the structural phase transition at T_{co} changes the perovskite structure, which is warped to pseudo tetragonal, to pseudo cubic. It has been pointed out that the structural phase transition at T_{co} is charge ordering characterized by the coexistence ratio $Mn^{3+} : Mn^{4+} = 7 : 1$ [2]. We paid attention to the coupling of e_g orbital or charge fluctuation of Mn ions to lattice in $La_{0.875}Sr_{0.125}MnO_3$ and measured the elastic constants by ultrasonic experiments.

Fig. 1 shows the temperature dependence of the elastic constants in $La_{0.875}Sr_{0.125}MnO_3$. Above T_s, the $(C_{11} - C_{12})/2$ shows a remarkable softening, while the C_{44} exhibits a monotonous increase in lowering temperature. These obsevations are similar to those of $La_{1-x}Sr_xMnO_3 (x = 0.12, 0.165)$ and explained by the quadrupolar susceptibility of a $3d$ electron in e_g orbital doublet[3]. The temperature dependence of the elastic constant $C_{\Gamma3} = (C_{11} - C_{12})/2$ is described by

$$C_{\Gamma3}(T) = C^0_{\Gamma3} - N g^2_{\Gamma3} \frac{\chi_{\Gamma3}(T)}{1 - g'_{\Gamma3}\chi_{\Gamma3}(T)}. \quad (1)$$

Here $\chi_{\Gamma3}$ is the quadrupolar susceptibility for O^0_2 or O^2_2 of a $3d$ electron in e_g orbital doublet, $C^0_{\Gamma3}$ is a background and N is the number of Mn^{3+} ions in unit volume. The $g_{\Gamma3}$ is a coupling constant of quadrupole-strain interaction as $H_{QS} = -\sum_i g_{\Gamma3}(O^0_2(i)\varepsilon_u + O^2_2(i)\varepsilon_v)$. The O^0_2 and O^2_2 are

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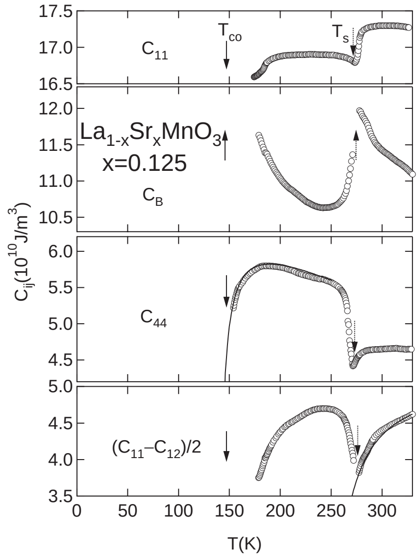

Fig. 1. Temperature dependence of the elastic constants in $La_{0.875}Sr_{0.125}MnO_3$.

quadrupolar operators on $3d$ electron and ε_u and ε_v are elastic strains with Γ_3 symmetry. The $g'_{\Gamma3}$ is a coupling constant for the inter-site quadrupolar interaction as $H_{QQ} = -\sum_i g'_{\Gamma3}(\langle O_2^0\rangle O_2^0(i) + \langle O_2^2\rangle O_2^2(i))$. The solid line on the $(C_{11}-C_{12})/2$ above T_s in Fig.1 is the calculated curve with Eq. (1) and we obtained $|g_{\Gamma3}| = 462$ K and $g'_{\Gamma3} = 20$ K. These coupling constants are comparable with those of $La_{1-x}Sr_xMnO_3(x = 0.12, 0.165)$ [3].

The other important finding is that both of $(C_{11} - C_{12})/2$ and C_{44} show a softening above T_{co} definitely. These softenings appear around $x = 1/8$, for instance $La_{0.88}Sr_{0.12}MnO_3$ [3]. The quadrupolar susceptibility of an electron in e_g orbital doublet could not lead to the softening of C_{44} above T_{co}. Therefore, the softening of C_{44} above T_{co} originates from the coupling of charge fluctuation of Mn ions associated with the coexistence ratio of $Mn^{3+} : Mn^{4+} = 7 : 1$ to elastic strain with Γ_5 symmetry such as $H_{CS} = -\delta Q_{\Gamma5}\varepsilon_{\Gamma5}$. Here $Q_{\Gamma5}$ is the order parameter of the charge ordering. The softening of $(C_{11} - C_{12})/2$ probably arises from the anharmonic coupling of elastic strain with Γ_3 symmetry to the order parameter fluctuation. In the case of charge ordering such as Yb_4As_3 [4], the softening of the elastic constant owing to the charge fluctuation of Yb^{2+} and Yb^{3+} ions is written as $C_\Gamma(T) = C_\Gamma^0(T - T_c^0)/(T - \Theta)$. The equation can be applied to the softening of C_{44} above T_{co} in the present case. The solid line on the C_{44} above T_{co} in Fig. 1 is the calculated curve with this equation and we obtained the characteristic temperatures $T_c^0 = 140$ K, $\Theta = 137$ K of C_{44}.

Colossal magnetoresistance around T_c is also observed in $La_{0.875}Sr_{0.125}MnO_3$[5]. Therefore, we have

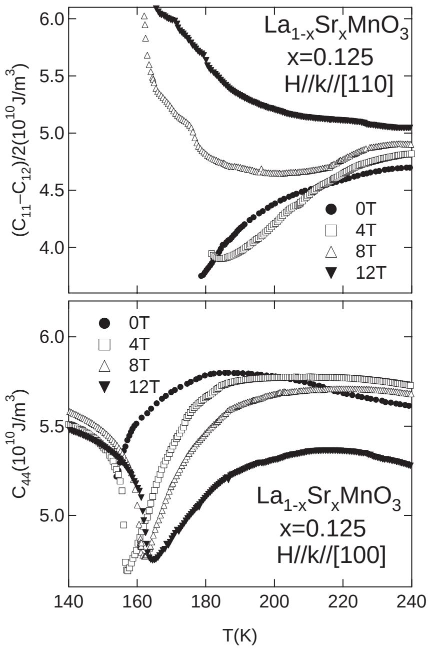

Fig. 2. Temperature dependence of the elastic constants $(C_{11} - C_{12})/2$ and C_{44} around T_{co} in $La_{0.875}Sr_{0.125}MnO_3$ under several magnetic fields along [110] and [100], respectively.

measured the elastic constants in $La_{0.875}Sr_{0.125}MnO_3$ under several magnetic fields. Fig. 2 shows the temperature dependence of the elastic constants $(C_{11}-C_{12})/2$ and C_{44} around T_{co} in $La_{0.875}Sr_{0.125}MnO_3$ under several magnetic fields along [110] and [100], respectively. The C_{44} remains to show a pronounced softening about 10 % under 12 T. This means the charge fluctuation of Mn ions characterized by $Q_{\Gamma5}$ is still relevant even in fields up to 12 T. In contrast, the softening of $(C_{11} - C_{12})/2$ above T_{co} becomes small gradually with increasing magnetic field. The charge ordering point corresponding to the minimum of C_{44} moves to high temperatures with increasing magnetic field rapidly. The charge ordering of Mn ions in $La_{0.875}Sr_{0.125}MnO_3$ becomes stable under magnetic fields.

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