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Characteristics of Short-ranged Stripe Order in the Bilayer Manganite $La_{2-2x}Sr_{1+2x}Mn_2O_7$

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The A-type antiferromagnetism in the two-dimensional $La_{2-2x}Sr_{1+2x}Mn_2O_7$ crystal is accompanied with short-ranged stripe order. In the c-plane, holes form a vertical stripe with periodicity of $\sim 3.3a$. Within each bilayer, two stripes overlap and form a bi-stripe structure. The bi-stripes form a body centred structure in the bc plane. We argue that the stripe order causes insulating behavior of the resistivity over a whole temperature region. Comparing with the two-dimensional (2D) high T_C cuprate superconductor and isomorphic 2D nickelate, we examine characteristics of the stripe ordering in the present 2D bilayer system $La_{2-2x}Sr_{1+2x}Mn_2O_7$.

KEYWORDS: colossal magnetoresistance (CMR), bilayer manganite $La_{2-2x}Sr_{1+2x}Mn_2O_7$, stripe order, A-type antiferromagnet

§1. Introduction

Charge ordering is an ubiquitous phenomenon for perovskite transition-metal oxides especially in the vicinity of either the metal-insulator transition, or the insulator to superconductor transition. Depending on detailed electron-electron or electron-phonon interactions, the charge ordering exhibits a variety of charge ordering patterns. A stripe order appears in high T_C cuprate superconductors[1] and isomorphic nickelates,[2,3] while a so-called "CE"-type charge/orbital order is frequently observed in hole-doped manganites such as $La_{1-x}Ca_xMnO_3$[4,5] and $La_{0.5}Sr_{1.5}MnO_4$.[6] A two-dimensional (2D) bilayer manganite $La_{2-2x}Sr_{1+2x}Mn_2O_7$ also exhibits characteristic charge ordering. Vasiliu-Doloc *et al.* reported an indication of the short-ranged charge ordering in the $x = 0.40$ sample.[7] Very recently, Kubota *et al.* have established that the diffuse scattering pattern of the possible charge ordering is consistent with the stripe order with the propagation vector parallel to the tetragonal [100] (*vertical*) direction, and that it persists for a surprisingly wide range of hole concentration ($0.30 \leq x \leq 0.50$).[8] In this report, we shall argue the structure of the stripe order in the $La_{2-2x}Sr_{1+2x}Mn_2O_7$ system, and compare with the stripe orders observed in cuprates and nickelates.

§2. Experimental

Neutron diffraction measurements were carried out on triple-axis spectrometers GPTAS and TOPAN in the JRR-3M of JAERI, Tokai, Japan. Several incident neutron momentums were utilized, depending on the necessity of intensity and instrumental resolution, along with various combinations of collimators with PG filters. The $(h0l)$ and $(hk0)$ reciprocal planes were aligned as the scattering planes, and the samples were set in aluminum capsules filled with helium gas and were attached to the cold head of a closed-cycle helium gas refrigerator.

§3. Results and Discussion

The A-type antiferromagnetic (AFM) spin ordering is observed near the hole concentration $x \sim 0.5$ in the three-dimensional manganites.[9] The A-type AFM spin structure consists of antiferromagnetically stacked ferromagnetic (FM) layers. One of important features of the A-type AFM state is metallic conductivity. For example, $Nd_{0.45}Sr_{0.55}MnO_3$ is known to exhibit the A-type AFM structure, and its resistivity is metallic below $T_N (= 220\ K)$.[10] Such a metallic character is ascribed to the interplay between the unique orbital ordering of the $d_{x^2-y^2}$ orbitals within FM MnO_2 layers and the double-exchange mechanism in doped manganites.

$La_{2-2x}Sr_{1+2x}Mn_2O_7$ shows the 2D character due to the crystal structure in which FM MnO_2 layers are well separated by insulating non-magnetic $(La_{1-x}Sr_x)_2O_2$ layers.[11,12] The $x = 0.48$ sample shows the onset of the A-type AFM spin order ($T_N = 205\ K$),[13] but its resistivity remains insulating even below T_N in distinct contrast to the metallic behavior of the resistivity of $Nd_{0.45}Sr_{0.55}MnO_3$. We have demonstrated that the $La_{2-2x}Sr_{1+2x}Mn_2O_7$ system for $0.30 \leq x \leq 0.50$ is characterized by the bi-stripe order in the paramagnetic phase as well as in the A-type AFM spin ordered phase, and that the stripe order gives rise to the insulating behavior of the resistivity.[8] Figure 1(a) shows the profile of the stripe order observed in the $(h0l)$ scattering plane. Throughout this report, we employ the unit cell of $a \times a \times c$ of the underlying tetragonal crystal structure of $La_{2-2x}Sr_{1+2x}Mn_2O_7$. The scan was made along the

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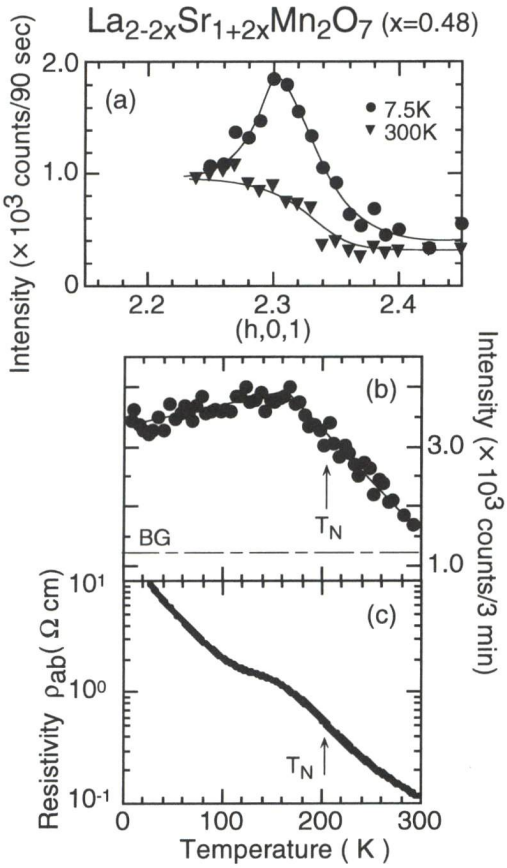

Fig. 1. (a) Profiles of the diffuse scattering from the stripe order in the A-type AFM sample $La_{2-2x}Sr_{1+2x}Mn_2O_7$ with $x = 0.48$ observed at T=300 K (background), and 7.5 K. Horizontal bar indicates the instrumental resolution. (b),(c) Temperature dependence of the diffuse intensity and the resistivity of the $x = 0.48$ sample.

[100] direction. This profile can be interpreted as follows. In the presence of the stripe ordering, Mn ions with two different valences may form a charge ordering pattern in the system. Since the radii of Mn^{+3} and Mn^{+4} ions are different, they cause a difference of the Mn-O bond length, yielding local distortions in the system. Consequently, such distortions may be observed as nuclear diffuse peaks in neutron diffraction experiments. In the present study, center positions of the observed diffuse signal are $(2 \pm 0.3\ 0\ 1)$, which indicates the stripe ordering enlarges the unit cell as $3.3a \times a \times c$. It can be viewed as a vertical stripe structure, where stripes run parallel to the a axis with the periodicity of holes of $3.3a$ within the c-plane. The periodicity of the stripe is incommensurate, and depends little on the hole concentration. Note that the line width of the profile is wider than the instrumental resolution, indicating that the stripe order remains short-ranged.

We have observed the temperature dependence of the diffuse signal for the bi-stripe order, and it is shown in Fig. 1(b). The strong diffuse intensity grows with decreasing temperature from 300 K, but levels off in the A-type AFM phase. The similar behavior was also observed for $x = 0.50$.[14)]

From the analysis of the observed data, we have determined the relative configuration of the stripes within and between bilayers, and their schematic pattern is depicted in Fig. 2. Detail discussions on the determination of the stripe ordering structure will be given elsewhere. As mentioned above, our analysis revealed that holes are aligned along the tetragonal [100] (vertical stripes). Furthermore, stripes are stacked in phase within the same bilayer, namely two stripes overlap and form a bi-stripe structure within each bilayers. (We tentatively call it as a *bi-stripe*.) On the other hand, a pair of bi-stripes are stacked out of phase between adjacent bilayers, thus bi-stripes form a body centred structure in the *bc* plane. This bi-stripe order pattern strongly indicates that the antiferromagnetic correlation is important within bilayers, but the Coulomb interaction is relevant outside bilayers.

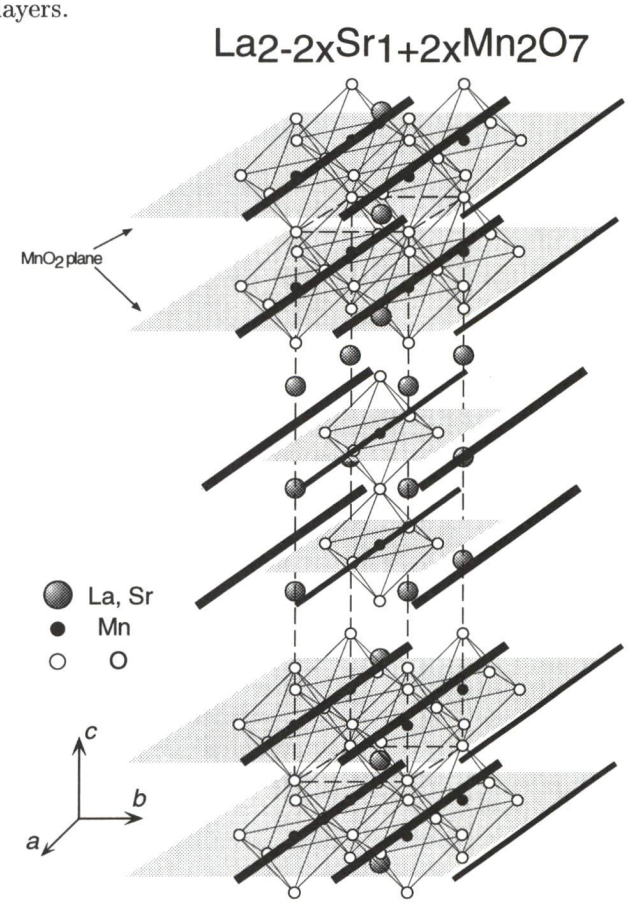

Fig. 2. Schematic pattern of the stripe order. The stripes form a body centred structure in the *bc* plane. Thick and thin line represent a larger and a smaller local lattice distortion, respectively.

The present bi-stripes in the bilayer manganite $La_{2-2x}Sr_{1+2x}Mn_2O_7$ have several novel characters comparing with those in the cuprates and the nickelates. Concerning the background spin pattern, the bi-stripes in the bilayer manganite subsist over the FM spin order, while it is well-known that the stripe order in the latter case emerges as an antiphase domain boundaries between the AFM spin correlation.[1,2)] In addition, the correlation length of the bi-stripe order is very anisotropic along and perpendicular to the stripe direction. Namely, the correlation length along the stripe direction is an order of magnitude shorter than that of the perpendicular direction. This implies either the hole density within

the stripes are strongly disturbed by Mn^{3+} ions, or the stripes are not straight, and consist of short segments of order of a few tens of Å.

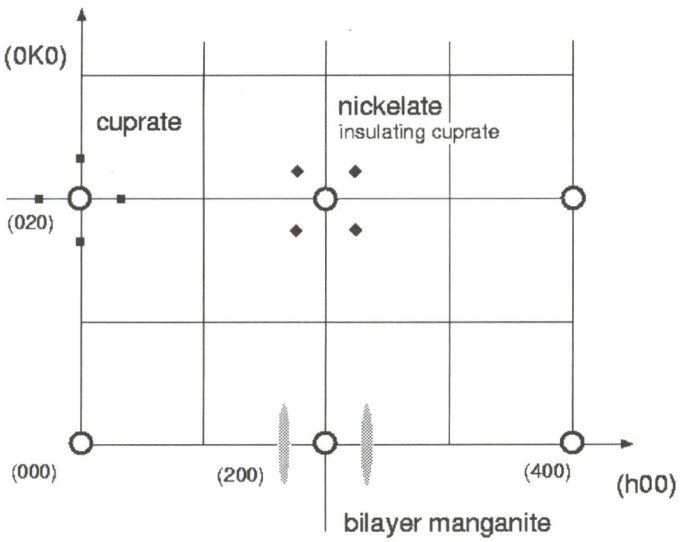

Fig. 3. The observed satellite position of the stripe order for the cuprate, the nickelate, and the manganite in the $(hk0)$ scattering plane.

Next, we shall examine the features of the diffuse scattering patterns of the stripe ordering within the *ab* plane for the cuprate ($La_{2-x}Sr_xCuO_4$), the nickelate ($La_{2-x}Sr_xNiO_4$), and the bilayer manganite ($La_{2-2x}Sr_{1+2x}Mn_2O_7$). Figure 3 illustrates the schematic positions of the satellites (diffuse scattering peaks) for the respective stripe orders on the $(hk0)$ scattering plane in the tetragonal symmetry. For the cuprates, two sets of paired satellites (total four spots) are observed along the vertical directions around each fundamental Bragg reflection as depicted at (020). This is consistent with the characteristics of the structure in which the tilting of the CuO_6 octahedra stabilizes the stripes along the [100] and [010] directions alternatively as going to the adjacent CuO_6 planes.[1,15,16]

On the other hand, in the insulating phase of cuprates and in the nickelates, a propagation vector of the stripe order is parallel to the [110] (*diagonal*) direction. In this case, four satellites were visible along the [110] and the $[1\bar{1}0]$ directions, as schematically depicted around (220).

Comparing these two cases, the diffuse pattern of the $La_{2-2x}Sr_{1+2x}Mn_2O_7$ system is very unique. First of all, there appear only two diffuse peaks in the longitudinal direction, as depicted around (200). They are peaked around $Q = (2 \pm q_o, 0, 0)$ (in the longitudinal direction) with $q_o \sim 0.3a^*$. It should be noted that the incommensurability (peak position) q_o is almost constant for $0.30 \leq x \leq 0.50$, in remarkable contrast with the linear relation with x in the case of the under-doped cuprate as well as nickelate systems.

As mentioned above, the width of diffuse signal is quite anisotropic, unlike those of the cuprate and the nickelate. To our knowledge, only the magnetic superlattice peak in the $x = 0.02$ Sr-doped spin-glass cuprate sample exhibits anisotropic diffuse scattering, but we caution that the underlying physics is totally different.[17]

§4. Conclusion

The short-ranged bi-stripe order exists in the $La_{2-2x}Sr_{1+2x}Mn_2O_7$ system for a surprisingly wide hole concentration region. Holes are aligned in line along the [100] direction in the tetragonal symmetry. The bi-stripe order dominates the transport properties and causes the insulating behavior of the resistivity.

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