Novel stripe-type charge ordering in the metallic A-type antiferromagnet $Pr_{0.5}Sr_{0.5}MnO_3$

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Abstract

A single crystal of $Pr_{0.5}Sr_{0.5}MnO_3$, which exhibits a collossal magnetoresistance phenomenon in its A-type antiferromagnetic state, was investigated by a neutron diffraction study. This material exhibits a stripe-type charge ordering with a wave vector $\boldsymbol{q} \sim (0, 0, 0.3)$, which controls the electron conductivity. This charge ordering is fundamentally different from a widely observed checkerboard (CE-type) charger ordering, and is specific to the conductive A-type AFM state.

Key words: colossal magnetoresistance; charge ordering; stripe order

A drastic increase of the conductivity accompanied by a ferromagnetic (FM) transition, or a colossal magnetoresistance (CMR) effect, in perovskite manganites has attracted enormous interests. Recent studies have revealed that the checkerboard-type (CE-type) charge ordering which appears at $x \sim 1/2$ is essential to this dramatic change of the transport property. The CE-type charge ordering is stabilized in systems with a narrow one-electron band width W such as $Pr_{1-x}Ca_xMnO_3$ and $La_{1-x}Ca_xMnO_3$. On the other hand, the CMR phenomenon is not limited to the narrow W manganites, and is indeed observed in systems with wider W such as $Pr_{1-x}Sr_xMnO_3$, $Nd_{1-x}Sr_xMnO_3$, and $La_{2-2x}Sr_{1+2x}Mn_2O_7$ with $x \sim 0.5$. An important characteristics of these compounds is they show a *highly conductive* A-type antiferromagnetic (AFM) state around $x = 1/2$, where a planar ordering of the $d(x^2 - y^2)$ orbitals is established. This orbital ordering mediates the FM coupling and fairly large mobility of carriers within the orbital-ordered planes [2,3]. The scenario for the CMR phenomenon based on the CE-type charge ordering is clearly irrelevant in this case, and another microscopic mechanism ought to be invoked.

To find what causes a drastic change in the transport property in wide W manganites with the A-type AFM state, we have performed a neutron diffraction study on a single crystal of a cubic A-type antiferromagnet $Pr_{0.5}Sr_{0.5}MnO_3$. This material exhibits a first-order phase transition from a FM metal to an A-type AFM less-conductive state at $T_N \sim 140$ K (Fig. 1(c)), and it exhibits a significant MR below T_N [1]. The neutron diffraction measurement was performed using the triple axis spectrometer GPTAS installed at the JRR-3M reactor in JAERI with an incident neutron momentum $k_i = 3.83$ Å^{-1}. The crystal has a distorted cubic structure with $a \sim 3.8$ Å, and we employ the cubic notation in indexing with the FM layers of the A-type AFM structure perpendicular to the [010] direction.

First, we surveyed the $(h, 0, l)$ scattering plane. Figure 1(a) presents a map of the scattering intensities around (002) at 7 K. Apart from an intense funda-

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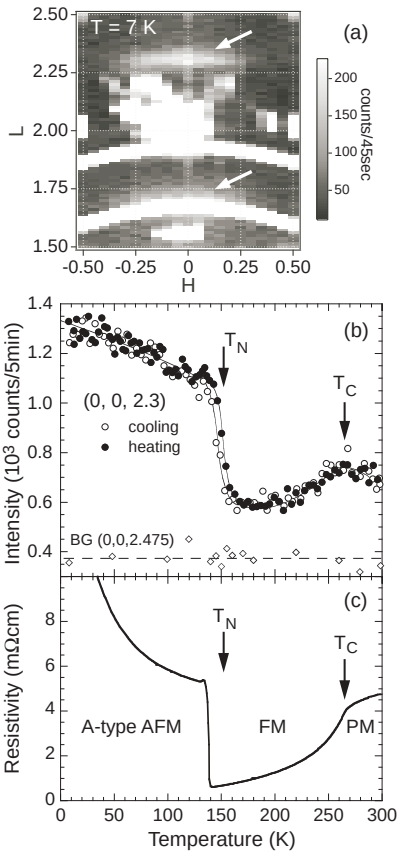

Fig. 1. (a) Intensity maps around $(0, 0, 2)$ at $T = 7$ K. Two ring-shaped scatterings are due to the Al sample cell. (b) Temperature dependence of the intensity at $(0, 0, 2.3)$. The background (BG) intensity measured at $(0, 0, 2.475)$ is also shown. Open symbols and closed symbols denote the data for cooling and for heating, respectively. (c) Temperature dependence of the resistivity.

mental nuclear Bragg reflection at (002) and A-type AFM Bragg reflections at $(\pm 0.5, 0, 2)$ and $(0, 0, 1.6)$, anisotropic diffuse scatterings are clearly observed around $(0, 0, 2 \pm 0.3)$ (white arrows). The profiles are elongated towards the [100] direction, indicating the correlation length along the modulation vector $\boldsymbol{q} = (0, 0, \pm 0.3)$ is much longer than that perpendicular to $\boldsymbol{q}$. By examining the $\boldsymbol{Q}$ dependence of the similar scatterings, we can conclude that these are attributable to longitudinal lattice modulations. A similar feature was also observed in the stripe-like charge ordering in a two-dimensional (2D) A-type AFM manganite $La_{2-2x}Sr_{1+2x}Mn_2O_7$ [4,5].

In order to investigate the influence of newly found diffuse scattering on the transport property, we measured the temperature (T) dependences of the intensity at $(0, 0, 2.3)$ (Fig. 1(b)). There is a strong correlation between the intensity and the resistivity (Fig. 1(c)), which establishes that the observed diffuse scattering

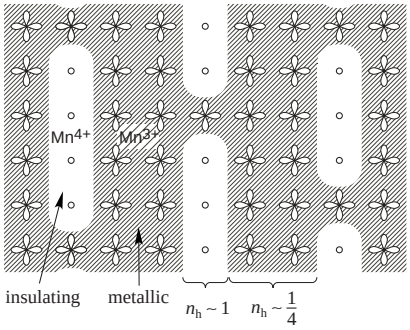

Fig. 2. Schematic illustration of the charge order and orbital order for $q = 1/3$ r.l.u. Cloverleaf symbols represent the $d(x^2-y^2)$ orbitals. $n_{\rm h}$ denotes the hole concentration within the Mn^{4+} stripe or the Mn^{3+}-like matrix.

arises from the charge ordering. Especially, the diffuse peak has large intensity in the A-type AFM state. In addition, we found that the correlation length of the charge ordering is well-developed below $T_{\rm N}$. These observations verify that the charge ordering is intrinsic to the A-type AFM state. Since a similar charge ordering was observed in the 2D A-type AFM manganites, the existence of such charge ordering must be specific to the wide W manganites independent of the spatial dimensionality.

The observed features of the charge ordering can be well explained by a model of the stripe-like charge ordering depicted in Fig. 2 for $q = 1/3$ r.l.u. In this model, Mn^{4+} ions segregate within the metallic matrix of Mn^{3+}-like sites with $d(x^2 - y^2)$ orbitals, and form stripe-like objects along the Mn-O-Mn bond direction. The insulating Mn^{4+} stripes block the hopping of the e_g electrons, whereas the Mn^{3+}-like matrix retains the mobility of carriers, producing the high conductivity in the AFM state. In Fig. 2, the hole concentration of the Mn^{4+} stripe is fixed to 1, while that of two Mn^{3+} lines be 1/4, resulting in the overall concentration $x = 1/2$. In reality, some e_g electrons may enter the line of Mn^{4+} ions as depicted in Fig. 2 to disturb the correlation along the stripes resulting in the observed anisotropy in the correlation lengths.

To summarize, we have performed a neutron diffraction study on a single crystal of $Pr_{0.5}Sr_{0.5}MnO_3$ and found an anisotropic charge ordering with a wave vector $q \sim 0.3$ r.l.u. The character of the charge ordering is consistent with the stripe-like charge ordering. This *"stripe-like charge ordering"* must be seriously considered to understand the transport properties in the materials with A-type AFM states.

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