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Rotation of orbital stripes in $Pr(Sr_{0.1}Ca_{0.9})_2Mn_2O_7$

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Introduction

In half-doped manganites, charge- and orbital- ordered (CO-OO) state is widely realized. Below the CO-OO transition temperature T_{CO}, charges are ordered in a checkerboard type, forming alternating Mn^{3+}/Mn^{4+} sites. Simultaneously, the e_g orbitals in Mn^{3+} sites are aligned in zigzag chain type. Because formations of orbital stripes and zigzag chains dominates the anisotropic charge dynamics in the CO-OO plane, the change in their direction connects to the control of the in-plane anisotropy of the optical and electrical conductivity. We have investigated the CO-OO states in a half-doped bilayered manganite $Pr(Sr_{0.1}Ca_{0.9})_2Mn_2O_7$.

Results

Through single-crystal X-ray diffraction measurements we have determined the lattice structures of $Pr(Sr_{0.1}Ca_{0.9})_2Mn_2O_7$ including superstructures affected by CO-OO: There exist two CO-OO phases in this material; the higher temperature phase (CO1) and the lower temperature one (CO2). Space groups are *Amam* (with a=5.410, b=5.462, c=19.277 Å at 405 K) for charge disordered phase ($T>T_{CO1}$), *Pbnm* (with a=5.412, b=10.921, c=19.234 Å at 330 K) for CO1 ($T_{CO1}>T>T_{CO2}$), and *Am2m* (with a=10.812, b=5.475, c=19.203 Å at 295 K) for CO2 ($T<T_{CO2}$), respectively. Note that the lattice structure is orthorhombic even above T_{CO1} because of tilt of MnO_6 octahedra around the a-axis. The orthorhombic distortion couples with the CO-OO to determine the direction of orbital stripes in the bilayeres. This is distinct from the case of the wide-bandwidth analogue, $LaSr_2Mn_2O_7$, which retains the tetragonal (*I4/mmm*) symmetry above the CO-OO transition temperature.

Figures 1(a) and (b) show the synchrotron X-ray oscillation photograph around the reflection 4 1 3 taken by using an imaging plate. Above $T_{CO1,}$ there is no additional spot except for fundamental reflections anticipated for orthorhombic *Amam* unit cell. In the higher temperature CO1 phase, superlattice spots appear at u v±1/2 w (u, v, w are integers) (Fig. 1(a)), indicating orbital zigzag-chains run along the b-axis, as depicted in Fig. 1(c). When the crystal is cooled below T_{CO2}, the superlattice spots along the b*-axis disappear and instead emerge along the a*-axis, i.e. at u±1/2 v w (Fig. 1(b)). This means that directions of orbital stripes and zigzag chains rotate by 90 degrees at T_{CO2} as depicted in Fig. 1(d), while retaining the underlying lattice orthorhombic cell essentially intact.

It is also revealed that the rotation of orbital stripes on the orthorhombically distorted lattice breaks the symmetry of the space inversion and gives rise to an electrically polarized state. The charge polarized state is also confirmed by the large second harmonic generation susceptibility in the low-temperature CO-OO state [2].

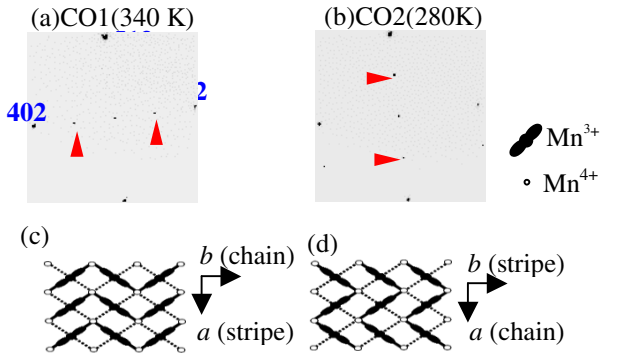

Fig. 1 X-ray oscillation photographs near the 4 1 3 reflection in $Pr(Sr_{0.1}Ca_{0.9})_2Mn_2O_7$ obtained by an imaging plate at 340 K (a), and 280 K (b), respectively. Red arrows indicate superlattice reflections. (c)(d) Schematics of charge and orbital ordering patterns at (c) $T_{CO1}>T>T_{CO2}$ (orbital stripes run along the a-axis) and (d) $T<T_{CO2}$ (orbital stripes run along the b-axis), respectively.

References

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