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Orbitally ordered state in $Y_{1-x}Ca_xTiO_3$ studied by resonant X-ray scattering

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Introduction

A perovskite-type transition metal oxide, $Y_{1-x}Ca_xTiO_3$, shows drastic Mott metal-insulator transition (MIT) induced by the substitution of Ca for Y [1]. This parent material $YTiO_3$ is a Mott-insulator and shows a ferromagnetic (FM)-insulator phase below Tc~30 K. The FM interaction is theoretically considered to be caused by the orbital ordering of Ti^{3+} t_{2g}-electron [2]. The theoretical predictions are consistent with the experimentally obtained orbital ordering [3]. By the substitution, the number of *3d*-electron on Ti ion can be controlled form 1 to 0. The FM ordering disappears at x_{FM}~0.15, while the insulating state is preserved up to x_{MI}~0.4. At the x_{MI} the drastic MIT occurs and the paramagnetic-metal phase is stabilized above x_{MI}. However, the origin of the respective phase transition is still unclear. The orbital degree of freedom in the transitions may play an important role. To make clear the orbital state in the respective phases, we have systematically investigated the orbitally ordered state in $Y_{1-x}Ca_xTiO_3$ using the resonant X-ray scattering (RXS) technique.

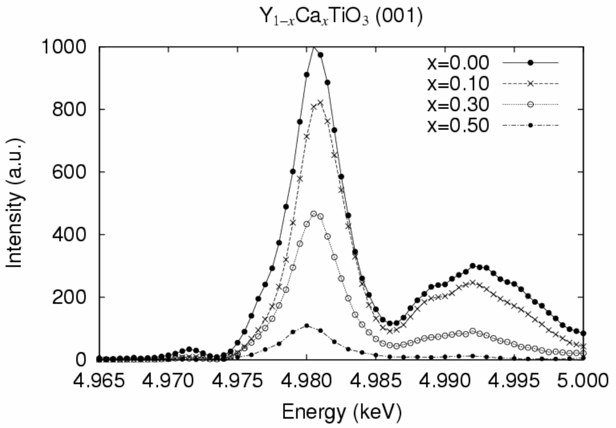
Experiment

The high quality single crystals of $Y_{1-x}Ca_xTiO_3$ were grown by a floating-zone technique. The powder x-ray diffraction at room temperature was performed to check the sample quality and the crystal structure at beam line(BL) 1B at Photon Factory. The RXS experiments were carried out by a four-circle diffractometer at BL-16A2. Polarizer with a PG(004) analyzer crystal was used in some experiments. The incident x-ray energy is about 4.97 keV of Ti *K*-edge and the energy resolution is about 1 eV.

Results

For the investigation of the orbitally ordered state of Ti *3d*-electron in $Y_{1-x}Ca_xTiO_3$, we have searched the RXS at the forbidden reflections, (001), (011) and (100), near Ti *K*-edge. The energy dependences of RXS intensity at (001) are shown in the figure as a function of Ca concentration. The RXS intensity is only a $\sigma \rightarrow \pi$ component. The energy spectra have two large peaks at *E*~4.980 (main-edge) and 4.994 keV. A small resonant peak is also observed at *E*~4.972 keV (pre-edge). The RXS intensities decrease monotonically and the energy spectra indicate no drastic change with increasing Ca concentration, x. The RXS at pre-edge (*1s->3d* transition energy) is caused by the hybridization between the Ti *4p* and the neighboring orbitally ordered Ti *3d*s. Therefore, the RXS intensity at the pre-edge is proper for the measurement of the order parameter of ordered *3d*-orbital. The RXS intensities at pre-edge rapidly decrease in 0<x<0.15 and barely survives at x=0.30 in the insulator phase before x_{MI}. In the metal phase, the intensity almost disappears. Namely, the orbital ordering is strongly suppressed toward x_{FM} and almost vanishes near x_{MI}. The hole concentration dependence of Jahn-Teller distortion determined by the x-ray structural analysis is also consistent with that of the orbitally ordered state.

On the other hand, the RXS at main-edge in metal phase may arise from the tilts of the neighboring TiO_6 octahedra as theoretically suggested [4].



References

[1] Y. Taguchi et al., Phys. Rev. B **48**, 511 (1993); F. Iga et al., Physica B **206-207**, 859 (1995).
[2] T. Mizokawa et al., Phys. Rev. B 60, 7309 (1999); M. Mochizuki and M. Imada, J. Phys. Soc. Jpn. **70**, 3475 (2000).
[3] J. Akimitsu et al., J. Phys. Soc. Jpn. **70**, 3475 (2001); M. Ito et al., J. Phys. Soc. Jpn. **68**, 2783 (1999); H. Nakao et al., Phys. Rev. B **66**, 184419 (2002).
[4] M. Takahashi et al., Phys. Rev. B **64**, 075110 (2001).

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