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Fabrication and electronic states of $Sr_2IrO_{4-x}F_{2x}$ thin films by topotactic fluorination

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1 Introduction

Iridium (Ir) oxides, being typical 5*d* transition metal oxides, have attracted much attention because of their unique physical properties arising from large spin-orbit interaction (SOI). For example, a layered perovskite Sr_2IrO_4 is a $J_{eff} = 1/2$ Mott insulator due to the combination of SOI and crystal field [1]. Recently, several studies have revealed that the electronic properties of layered perovskite oxides, such as Sr_2RuO_4, and $Sr_3Ir_2O_7$, can be altered by anion insertion into the SrO block with rock-salt structure or anion substitution for the O sites [2,3]. In this study, we succeeded in synthesizing new layered-perovskite $Sr_2IrO_{4-x}F_{2x}$ thin films by combining pulsed-laser deposition and topotactic fluorination methods and investigated their electronic states by synchrotron-radiation photoemission spectroscopy (PES).

2 Experiment

Sr_2IrO_4 epitaxial thin films were deposited on $SrTiO_3$ (001) substrates (STO) by pulsed laser deposition. The obtained precursor thin film was reacted with polyvinylidene fluoride (PVDF) for 3 hours at 250 °C in Ar gas flow. Crystal structure was investigated by X-ray diffraction (XRD), and chemical composition was evaluated by energy dispersive X-ray analysis (EDS). PES measurements were conducted at KEK-PF BL-2A at an incident energy of 1200 eV.

3 Results and Discussion

Figure 1 shows out-of-plane XRD patterns of the precursor thin film and the thin film reacted with PVDF. In the latter, several peaks from the precursor disappeared, and new peaks evolved. The lattice constant in the out-of-plane direction was expanded from 25.73 Å to 35.78 Å. In addition, EDS measurements confirmed the decrease in oxygen content and increase in fluorine content, accompanied with the reaction with PVDF. These results suggest that fluoride ion was inserted into the $(SrO)_2$ blocks of Sr_2IrO_4 with partial removal of oxide ion.

Figure 2 shows valence band spectra of the Sr_2IrO_4 and $Sr_2IrO_{4-x}F_{2x}$ thin films. The spectrum of the Sr_2IrO_4 film exhibited a peak located at ~1 eV (Peak A), which is assignable to the O 2*p* state hybridized with Ir 5*d* antibonding states. Notably, the peak was shifted towards higher binding energy side in the fluorinated film (Peak A'). Because fluorine has larger electronegativity than oxygen, the binding energy of F 2*p* state hybridized with a metal is generally higher than that of O 2*p*. Therefore, the peak A' can be assigned be a F 2*p*-Ir 5*d* hybridized state.

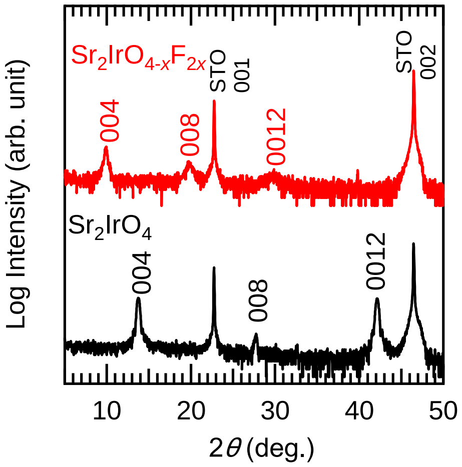

Fig. 1. XRD patterns of the Sr_2IrO_4 and $Sr_2IrO_{4-x}F_{2x}$ thin films.

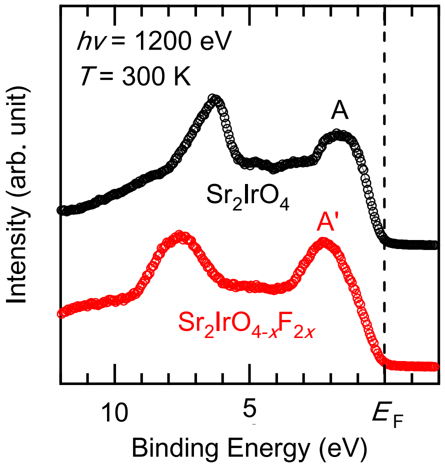

Fig. 2. Valence band spectra of the Sr_2IrO_4 and $Sr_2IrO_{4-x}F_{2x}$ thin films.

References

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