Electronic structure of infinite-layer $SrFeO_2$ film prepared via topotactic reduction

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In this research, we tried to investigate electronic structure of infinite-layer $SrFeO_2$ single crystalline film by using angle-resolved photoemission spectroscopy (ARPES). Because ARPES measurement requires clean surface of samples, we performed surface cleaning of the film via ultra-high vacuum annealing method. The annealing resulted in partially clean surface of the $SrFeO_2$ single crystalline film, however, a clear ARPES image was still difficult to be obtained.

0. About the authors

T. Katayama : He specializes in synthesis of anion doped oxide thin films with different anion orientations via pulsed laser deposition (PLD) and topotactic reactions. In this study, he synthesized thin-film samples by combining PLD and low-temperature topotactic reduction using CaH_2.

T. Sonobe : T. S. specializes in the angle-resorlved photoemission spectroscopy (ARPES) study on the electronic structure of the iron-based superconductors. His research interest is on the electronic structure of $SrFeO_2$ including the square planar Fe^{2+} layer like the iron-based superconductors. He performed the annealing process for surface cleaning and ARPES.

1. Introduction

Transition metal oxides are widely studied due to their intriguing features such as photocatalysis, ferroelectric, and colossal magnetoresistance. The properties of the oxides are strongly associated with the coordination of transition metal centers. Recently, low-temperature topotactic technique using metal hydrides enables to fabricate unique coordinations. For example, $SrFeO_2$, which is obtained by heating $SrFeO_{3-\delta}$ with CaH_2, possesses FeO_4 square-planer coordination as shown in Fig. 1, in contrast to conventional iron oxides which have three dimensionally FeO_6 octahedral, FeO_5 pyramidal and/or FeO_4 octahedral coordinations [1]. On the other hand, heating $SrVO_3$ with CaH_2 yields oxyhydride $SrVO_2H$ with VO_4H_2 octahedral coordination, where hydride ions locate at apical sites [2].

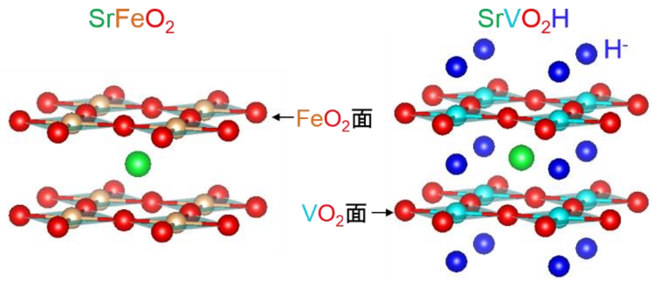

Fig. 1. Crystal structure of $SrFeO_2$ and $SrVO_2H$.

$SrFeO_2$ has attracted great interest because of the strong antiferromagnetic interaction with high Néel temperature of 473 K [1] and high electron mobility in a form of thin film [3]. So far, the electronic density states and electron configuration of iron in $SrFeO_2$ have been investigated by both theoretical calculation [4] and experimental X-ray photoemission and absorption spectroscopy methods [5]. However, the electronic structure has not been completely studied, and the origin of the high electron mobility is still unclear. Thus, we attempted further investigation of electronic structure of $SrFeO_2$ by using ARPES.

The ARPES measurement requires clean surface of single crystalline sample. In $SrFeO_2$, the single crystalline phase is only obtained in a form of film [6]. Therefore, we tried to perform ARPES measurement on single crystalline $SrFeO_2$ film, where its surface is cleaned through ultra-high vacuum annealing method.

2. Experiment

This work contains two steps; firstly, we checked crystal structure stability of $SrFeO_2$ films against ultra-high vacuum annealing by using X-ray diffraction (XRD) measurements, and then we performed ARPES on the films.

The $SrFeO_2$ epitaxial thin films with infinite-layer structure were prepared on 0.5 wt% Nb-doped STO (Nb:STO) substrates by combining the pulsed laser deposition (PLD) method with topotactic reduction using CaH_2, as reported in Ref. 3. The conductive Nb:STO substrates were used so as to reduce the charge-up effect. The thickness of the films was set as ~50 nm. The crystal structure stability of the $SrFeO_2$ films were examined by annealing at 220 and 400°C under vacuum pressure of 1×10^{-7} Torr. Crystal structures of the films were evaluated by using XRD with Cu *Kα* radiation.

For ARPES measurements, we used ARPES apparatus with R4000 analyser and light from a helium discharge tube (HeIIα = 40.814 eV). In this apparatus, samples can be introduced in ultra-high vacuum pressure after annealing process ($\leq 1*10^{-8}$ Torr). We performed ARPES measurements for the $SrFeO_2$ film and films obtained by annealing the $SrFeO_2$ films at 200 and 400°C for 2 h in ultra-high vacuum pressure. The all measurements were conducted at 300 K.

3. Results and discussion

3.1. Crystal structure

Figure 2 shows XRD patterns of the $SrFeO_2$ film and films obtained by annealing the $SrFeO_2$ film at 220 and 400°C for 2 h under vacuum pressure of 1×10^{-7} Torr. The $SrFeO_2$ film shows 002 XRD diffraction peak, probing that the film has *c*-axis infinite-layer structure. After annealing at 220°C, the 002 peak position did not shift, indicating that the single crystalline $SrFeO_2$ film has crystal structure stability against ultra-high vacuum annealing at 220°C. On the other hand, after annealing at 400°C, the 002 peak intensity decreased a little.

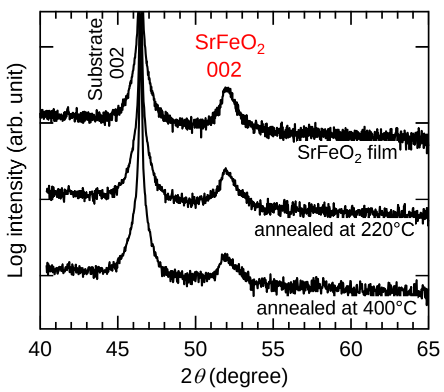

Fig. 2. XRD patterns for the $SrFeO_2$ film and films obtained by annealing the $SrFeO_2$ film at 220 and 400°C for 2 h under vacuum pressure of 1×10^{-7} Torr.

3.2. Attempt of electronic structure investigation of the $SrFeO_2$ films

ARPES measurements were conducted for the films. Figure 3(a) plots ARPES image of the $SrFeO_2$ film before annealing. Though some bands show 1eV-order band width in the band calculation[4], the spectrum does not show any angle dependence. It indicates that the measurement failed probably due to surface adsorption impurities. Figure 3(b) shows ARPES image of the film after annealing at 400°C for 2 h. The spectrum also does not show any angle dependence. Thus, in this study, we could not obtained good ARPES image of $SrFeO_2$.

Figure 3(c) shows valence band spectra for the $SrFeO_2$ films before and after annealing at 200 and 400°C for 2 h. The spectra have peaks at binding energy (E_B) of 5.5 eV and ~8 eV, which agrees well with the previous PES result[5] and the band calculation[4] considering charge-up effects. The peak at E_B = 5.5 eV does not change after annealing. On the other hand, the peak intensity at $E_B \sim$ 8 eV increases with annealing temperature. This suggests that the surface of the $SrFeO_2$ films was partially cleaned via ultra-high vacuum annealing method. Thus, it is suggested that it would become possible to obtain clear ARPES image of $SrFeO_2$ by using higher and longer annealing process.

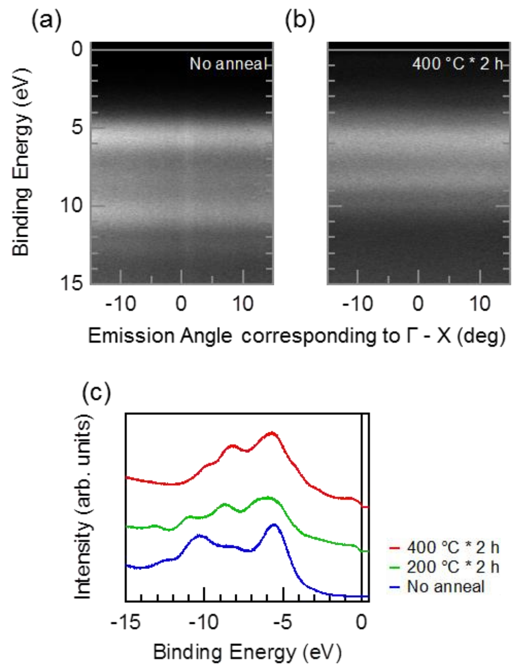

Fig. 3. ARPES images for the $SrFeO_2$ films (a) before and (b) after annealing at 400°C for 2 h. (c) Valence band spectra for the $SrFeO_2$ films before and after annealing.

4. Conclusion

We fabricated infinite-layer $SrFeO_2$ thin films and researched crystal structure stability of the $SrFeO_2$ films against ultra-high vacuum annealing and performed ARPES on the films. The annealing resulted in partially clean surface of the $SrFeO_2$ single crystalline film, however, a clear ARPES image was still difficult to be obtained.

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Reference

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