Thickness Dependence and Critical Thickness of Charge and Magnetic Orderings in Antiferromagnetic Perovskite $La_{1/3}Sr_{2/3}FeO_3$ Thin Films

Critical thickness is a characteristic parameter of thin films. There are some reports on the thickness dependence of ferromagnetic oxide thin films. In this study, we examined antiferromagnetic perovskite thin films and evaluated the spatial structure of ordered states. We used perovskite $La_{1/3}Sr_{2/3}FeO_3$ thin film and systematically varied the thickness. We used the X-ray diffraction technique and observed the change of correlation length around the critical thickness. Our results show that the correlation lengths of charge and magnetic orderings have different dependencies on thickness and the orderings disappear when two correlation lengths take similar values. These results suggest that spatial correlation is closely related to critical thickness.

Transition metal oxides exhibit various phenomena including giant magnetic resistance or superconductivity. These functionalities of oxides suggest that transition metal oxides have the potential to be used for electronic devices. Accordingly, oxide thin films should be studied intensively. The geometrical limitation originating from the thickness of a thin film affects the nature of the thin film and we examined the effect in this study. The critical thickness of ordered states is one of the most characteristic parameters of thin films. Antiferromagnetic perovskite $La_{1/3}Sr_{2/3}FeO_3$ thin film includes high-valent Fe^{5+} ($d^5\underline{L}^2$) and there are many previous reports on spectroscopic [2] and diffraction [3] studies. We examined the effects of thickness on charge and magnetic ordered states when varying the thickness of the samples.

The orderings are shown in Fig. 1(a). $La_{1/3}Sr_{2/3}FeO_3$ thin film shows charge ordering of Fe^{3+}-Fe^{5+}-Fe^{3+} along the [111] direction below the transition temperature of ~190 K. At the same temperature, long-range antiferromagnetic ordering of $Fe^{3+}\downarrow$-$Fe^{5+}\downarrow$-$Fe^{3+}\downarrow$- $Fe^{3+}\uparrow$-$Fe^{5+}\uparrow$-$Fe^{3+}\uparrow$ is realized due to antiferromagnetic coupling between Fe^{3+}-Fe^{3+} and ferromagnetic coupling between Fe^{3+}-Fe^{5+}. This transition is accompanied by a resistivity jump. The thickness effect of the resistivity jump was clarified by Minohara et al. and they showed that the resistivity jump remains around 13 nm and there is less dependence on substrate direction [4].

We used the X-ray diffraction technique in order to capture spatial information on the orderings in ultra-thin films. Hard X-ray diffraction was carried out at BL-4C for charge ordering. The X-ray energy was 12.4 keV and the samples were $La_{1/3}Sr_{2/3}FeO_3$ (5, 15, 17, 78 nm)/$SrTiO_3$(111) thin film. Resonant soft X-ray diffraction was used for detecting magnetic ordering at BL-19B. The X-ray energy was tuned to the Fe 2p absorption edge and the samples were $La_{1/3}Sr_{2/3}FeO_3$ (5, 15, 34, 78 nm)/$SrTiO_3$ (111) thin films.

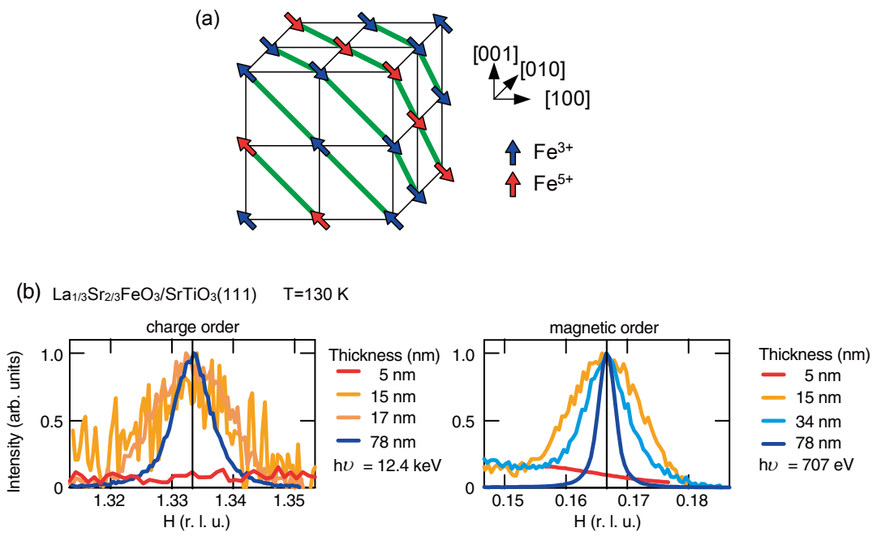

Figure 1: (a) Charge and magnetic ordering in $La_{1/3}Sr_{2/3}FeO_3$. (b) Diffraction peak from charge ordering around [4/3 4/3 4/3] and magnetic ordering around [1/6 1/6 1/6].

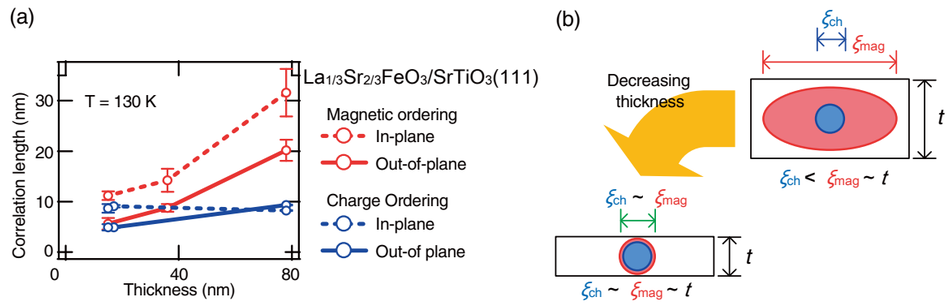

Figure 2: (a) Correlation lengths of charge (ξ_{ch}) and magnetic (ξ_{mag}) ordering in $La_{1/3}Sr_{2/3}FeO_3$ are plotted as a function of thickness. (b) Thickness dependence of correlation lengths is depicted schematically.

The charge ordering peak disappeared around 15 nm and the magnetic ordering peak also disappeared in the range of 5–15 nm as shown in Fig. 1(b). These results are consistent with previous reports of resistivity measurements. We examined the correlation length, which is defined as the inverse of peak width. The obtained correlation length is plotted as a function of thickness (t) in Fig. 2(a). The correlation length of magnetic order (ξ_{mag}) is proportional to thickness; however, the correlation length of charge orderings (ξ_{ch}) is more robust to change of thickness. This relationship is schematically depicted in Fig. 2(b). The correlation length of magnetic ordering is limited by thickness and charge, and magnetic orderings disappear when the correlation length of magnetic orderings reaches that of charge ordering. The thickness at that time gives the critical thickness.

We examined surface states because critical thickness sometimes arises from the surface state. We employed surface sensitive grazing incident resonant soft X-ray diffraction for studying the surface states of $La_{1/3}Sr_{2/3}FeO_3$ thin film. The grazing incident measurement was performed at BL-19B under experimental conditions similar to those of normal resonant soft X-ray diffraction as described above except for the orientation of the sample. The samples were $La_{1/3}Sr_{2/3}FeO_3$/$SrTiO_3$(110) thin films and the modulation vector of magnetization ordering lies in-plane. X-ray probing depth was around 2 nm due to the small incident angle. Transition temperature and correlation length are of the same order as the abovementioned results. This indicates that the picture of critical thickness obtained from the thickness dependence of the diffraction peak comes from the intrinsic geometrical limitations of thickness.

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BEAMLINES

BL-19B and BL-4C

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