X-ray spectroscopic study of BaFeO$_3$ thin films; an Fe$^{4+}$ ferromagnetic insulator

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We investigated the electronic and magnetic properties of fully oxidized BaFeO$_3$ thin films, which show ferromagnetic-insulating properties with cubic crystal structure, by hard x-ray photoemission spectroscopy (HAXPES), x-ray absorption spectroscopy (XAS) and soft x-ray magnetic circular dichroism (XMCD). We analyzed the results with configuration-interaction (CI) cluster-model calculations for Fe$^{4+}$, which showed good agreement with the experimental results. We also studied SrFeO$_3$ thin films, which have an Fe$^{3+}$ ion helical magnetism in cubic crystal structure, but are metallic at all temperatures. We found that BaFeO$_3$ thin films are insulating with large magnetization (2.1µ$_B$/formula unit) under ~ 1 T, using valence-band HAXPES and Fe 2p XMCD, which is consistent with the previously reported resistivity and magnetization measurements. Although Fe 2p core-level HAXPES and Fe 2p XAS spectra of BaFeO$_3$ and SrFeO$_3$ thin films are quite similar, we compared the insulating BaFeO$_3$ to metallic SrFeO$_3$ thin films with valence-band HAXPES. The CI cluster-model analysis indicates that the ground state of BaFeO$_3$ is dominated by $d^5L$ (L: ligand hole) configuration due to the negative charge transfer energy, and that the band gap has significant O 2p character. We revealed that the differences of the electronic and magnetic properties between BaFeO$_3$ and SrFeO$_3$ arise from the differences in their lattice constants, through affecting the strength of hybridization and bandwidth.

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I. INTRODUCTION

Strongly correlated transition-metal oxides show interesting physical properties such as colossal magnetoresistance, metal-insulator transitions, and the ordering of charge, orbital and spin [1]. Among this class of materials, high-valent metal compounds, such as SrFeO$_3$ and CaFeO$_3$, have attracted great attention due to their rich and anomalous physical properties [2]–[11]. CaFeO$_3$ shows charge ordering below 290 K with the charge disproportion of $d^5L + d^5L \rightarrow d^3 + d^5L^2$ ($L$: ligand hole), inducing monoclinic crystal distortion [3, 4]. The ground state of SrFeO$_3$ is dominated by the $d^5L$ configuration and is metallic with cubic crystal structure. The $d^5L$ electronic configuration gives rise to features in SrFeO$_3$, such as the helicoidal magnetic order, metallicity and absence of Jahn-Teller effect, which cannot be expected in the $d^4$ electronic configuration [2, 3]. Mostovoy theoretically revealed that the helicoidal magnetic order of perovskite Fe$^{4+}$ compound is attributed to the O 2p hole, rather than the competition between ferromagnetic double-exchange and antiferromagnetic superexchange interaction [2]. Furthermore, the reports of topological Hall effect suggest many possibilities for complex spin textures [5, 8].

BaFeO$_3$ is also one of those Fe$^{4+}$ series, and Hayashi et al. reported the first success in synthesis of polycrystalline cubic BaFeO$_3$ with the lattice constant of 3.97 Å [14]. A-type helicoidal magnetic state with $q = (0 0 0 06) \frac{2\pi}{a}$ ($a = 3.97$ Å) was found, but it becomes ferromagnetic under a small magnetic field of ~ 0.3 T with a saturation magnetization of 3.5 µ$_B$/formula unit (f.u.), and transition temperature of 111 K, respectively [14]. Li et al. theoretically showed that the energy difference between the A-type helicoidal magnetic and ferromagnetic phase is quite small, and can be switched by small magnetic field in BaFeO$_3$ unlike SrFeO$_3$ [12, 16]. The difference in their magnetic properties is explained by the difference in their lattice constant: the larger Ba stretches out the lattice and decreases the strength of Fe 3d - O 2p hybridization, which closely relates to the magnetic phases [12, 15, 16].

There have been several reports on BaFeO$_3$ thin films on SrTiO$_3$ (001) substrates prior to the successful fabrication of bulk BaFeO$_3$, but all of them showed much lower magnetization and larger lattice constant than bulk BaFeO$_3$, due to oxygen vacancies in the thin films [17–20]. Chakraverty et al. reported the synthesis of fully
oxidized single crystalline BaFeO$_3$ thin film which shows large saturation magnetization (in-plane: 3.2$\mu_B$/f.u., out-of-plane: 2.7$\mu_B$/f.u.) with the bulk lattice constant of 3.97 Å in cubic crystal structure [21]. Although the saturation magnetization and lattice constant of the recently reported thin film were quite close to that of bulk BaFeO$_3$, no signature of helicoidal magnetic structure has been observed [14, 21]. Transport and optical properties indicate that the fully oxidized film is an insulator with band gap $\sim$ 1.8 eV. The absence of the helicoidal magnetic order in BaFeO$_3$ film is likely due to the small energy barrier between the A-type helicoidal magnetic and the ferromagnetic phases, so that residual strain, or reduced dimensionality, stabilizes the uniform ferromagnet [16].

Given these exciting preliminary results on newly available single crystalline BaFeO$_3$ films, spectroscopic studies of the electronic and magnetic structures would be invaluable. For this purpose, we carried out hard x-ray photoemission spectroscopy (HAXPES) for Fe 2$p$ core level and valence band, Fe 2$p$ and O 1$s$ x-ray absorption spectroscopy (XAS) and x-ray magnetic circular dichroism (XMCD). The results were analyzed with configuration-interaction (CI) cluster-model calculations. These experimental and calculated results accurately explain that the difference of the valence-band spectra between BaFeO$_3$ and SrFeO$_3$ originates from the differences in their strength of Fe 3$d$ - O 2$p$ hybridization due to the difference in their atomic distances $(R)$ [the atomic distance of BaFeO$_3$ $(R_{BFO})$ is $\sim$ 1.99 Å and that of SrFeO$_3$ $(R_{SFO})$ is $\sim$ 1.93 Å, respectively].

II. EXPERIMENTAL AND CALCULATIONS

A single crystalline, cubic, fully oxidized 50 nm thin film of BaFeO$_3$ (with lattice constant 3.97 Å) was grown on SrTiO$_3$ (001) substrate, using pulsed laser deposition. The details of fabrication are described in Ref. [21]. HAXPES measurements were performed at BL47XU in SPring-8 at room temperature with the photon energy of 7940 eV (probing depth is $\sim$ 5 nm) [22] for both BaFeO$_3$ thin films and 50 nm thin films of SrFeO$_3$ on (LaAlO$_3$)$_{0.3}$-(SrAl$_{0.5}$Ta$_{0.5}$O$_3$)$_{0.7}$ (LSAT) (001) substrates to compare their electronic states [8]. The Fermi energy $(E_F)$ position and the energy resolution of $\sim$ 250 meV were determined by measuring the Fermi edge of gold which has electrical contact with the samples. XAS and XMCD were carried out at BL-16A in Photon Factory at a temperature of 20 K. The magnetic field of $\sim$ 1 T was applied perpendicular to the thin film surface, strong enough to saturate the out-of-plane magnetization [21]. XMCD was measured by switching the applied magnetic field under a fixed circular polarization of the incident beam incident on the sample perpendicular to its plane. The XAS and XMCD spectra were detected with the total-electron-yield mode with the probing depth of $\sim$ 2 nm [23, 24].

| TABLE I: Parameters used for the CI Cluster-model calculations of HAXPES in Fig.1 and Fig.2 and XAS and XMCD spectra in Fig.3. See text for details. |
|----------------------------------|
| $\Delta$ $(pds\bar{r})$ | $U_{dd}$ | $\phi$ [eV] |
|--------------------------|---------|-------|
| BaFeO$_3$ | $-0.9$ | $-1.5$ | $7.1$ |

The CI cluster-model calculation has been prevalently utilized to describe the physical properties of correlated materials since it can treat the electronic correlation in a cluster precisely [11, 10, 22, 20]. The details of the CI cluster-model calculations are given in Refs. [10, 25, 26]. In this model, multiplet states including the charge transfer state $\Psi_{\alpha}$, Eq. (1)) and for the final state of Fe 2$p$ core-level HAXPES $(\Psi_{fj},$ Eq. (2)), of valence-band HAXPES $(\Psi_{fj},$ Eq. (3)) and of Fe 2$p$ XAS $(\Psi_{fj},$ Eq. (4)) are described as

$$|\Psi_{\alpha}\rangle = |\alpha_1 \rangle |d^4\rangle + \alpha_2 |d^5L\rangle + \alpha_3 |d^6L^2\rangle + \cdots$$ (1)

$$|\Psi_{f1}\rangle = |\beta_1 \rangle |d^4\rangle + |\beta_2 \rangle |d^5L\rangle + |\beta_3 \rangle |d^6L^2\rangle + \cdots$$ (2)

$$|\Psi_{f2}\rangle = |\gamma_1 \rangle |d^4\rangle + |\gamma_2 \rangle |d^5L\rangle + |\gamma_3 \rangle |d^6L^2\rangle + \cdots$$ (3)

$$|\Psi_{f3}\rangle = |\delta_1 \rangle |d^4\rangle + |\delta_2 \rangle |d^5L\rangle + |\delta_3 \rangle |d^6L^2\rangle + \cdots$$ (4)

respectively. Here, $\zeta$ denotes the Fe 2$p$ core hole. Fitting parameters in this model are the charge transfer energy from the O 2$p$ orbitals to the empty Fe 3$d$ orbitals denoted by $\Delta$, the strength of Fe 3$d$ - O 2$p$ hybridization denoted by Slater-Koster parameters (p$ds\bar{r}$), the on-site 3$d$ - 3$d$ Coulomb interaction energy denoted by $U_{dd}$. The parameters used in this work are summarized in TABLE 1 which were adjusted near the phase boundary of A-type helicoidal magnetic and ferromagnetic phases to explain the magnetic ground state of BaFeO$_3$ thin films [12]. The CI cluster-model calculation with our set of parameters revealed that the ground state is dominated by $d^5L^2$ configuration (17% $d^5L$, 65% $d^5L^2$, 18% $d^6L^2$) due to the negative charge transfer energy.

III. RESULTS AND DISCUSSION

Figure 1 shows the Fe 2$p$ core-level HAXPES spectra for SrFeO$_3$ (top), BaFeO$_3$ (middle) and CI cluster-model calculation (bottom). The experimental spectra have main peaks (feature A and A’') and weak satellite structures (feature B, C, B’ and C’') for both the 2$p_{3/2}$ and 2$p_{1/2}$ transition as indicated in the top and middle spectra of Fig.1. Since Fe$^{3+}$ would greatly enhance the feature C, the weak intensities of the feature C in our results suggest that the formal valence of Fe is not 3+ but 4+ and that both the sample SrFeO$_3$ and BaFeO$_3$ thin films were fully oxidized (probing depth; $\sim$ 5 nm from the surface) [10, 21, 26]. The CI cluster-model calculation indicates that the main peaks (feature A and A’’)
The slight lowering in the binding energy of feature C in SrFeO$_3$ originates from the reduction of Fe 3d - O 2p hybridization due to the smaller lattice constant, the structure of feature A shifts toward higher binding energy, and its bandwidth becomes broader, as we will discuss later. These features are reproduced by local-spin-density-approximation calculations in Ref. [16], although they predicted that BaFeO$_3$ becomes half-metallic. The multiplet states calculated by cluster-model calculations were decomposed into $d^5$, $d^5 L$ and $d^6 L^2$ components. The feature B and C in the experimental spectrum for BaFeO$_3$ consist of strongly hybridized states between $d^5 L$ and $d^6 L^2$ configurations. The discrete state composing the feature A, which locates the binding energy of $\sim 1.8$ eV, is created by the strong Fe 3d - O 2p hybridization. The results of cluster-model calculations showed that the $d^6 L^2$ configuration dominates at the discrete first ionization state (2 % $d^5$, 40 % $d^5 L$, 58 % $d^6 L^2$), indicating the heavy O 2p character at the state. Therefore, the dominant transition from the ground state to first ionization state is $d^6 L^2 \rightarrow d^5 L$, indicating the extra oxygen hole in this transition.

The Fe 2p XAS (top) and XMCD (bottom) spectra are shown in Fig. 3(a). The broad 2p$_{3/2}$ XAS peak without shoulder structures reflects the heavily mixed state due to the strong hybridization in BaFeO$_3$ which is quite similar to that in SrFeO$_3$. These broad peaks seen in BaFeO$_3$ and SrFeO$_3$ suggests that the valence of Fe is not 3+ but 4+. Fe 2p XMCD shows a large peak intensity of $\sim 18$ % of the XAS peak intensity. The XMCD intensity of BaFeO$_3$ thin film is approximately twice as large as that of bulk BaFeO$_3$. To obtain the orbital moment ($M_{\text{orb}}$) and spin moment ($M_{\text{spin}}$), we applied XMCD sum rules in Eq. (5) and (6) [31, 32].

$$M_{\text{orb}} = \frac{2n\hbar}{3N} \int_{2p_{1/2}} \Delta \mu \, dE$$  

$$M_{\text{spin}} = \frac{7}{2} M_T = \frac{n\hbar}{N} \left( \int_{2p_{3/2}} \Delta \mu \, dE - 2 \int_{2p_{1/2}} \Delta \mu \, dE \right)$$

$\Delta \mu = \mu^+ - \mu^-$ and $\frac{1}{2}(\mu^+ + \mu^-)$ corresponds to the...
The XMCD and XAS intensity defining the expression for
\[ N = \frac{1}{\pi} \int_{2p_{u/\sigma}+2p_{\pi}} (\mu^+ + \mu^-) \, dE. \]
Here, \( \mu^+ \) and \( \mu^- \) denote the absorption intensities for left-handed and right-handed circularly polarized incident photons. \( n_k \) denotes the number of holes in the \( d \) orbital, and we assumed \( n_k = 6 \) for this analysis due to the formal valence of \( Fe^{4+} \).

By applying the XMCD sum rules, we obtained a large magnetic moment of \( M_{total} = 2.1 \pm 0.3 \mu_B/f.u., \) composed of \( M_{spin} = 1.8 \pm 0.2 \mu_B/f.u. \) and \( M_{orb} = 0.3 \pm 0.1 \mu_B/f.u. \). Note that we considered the correction coefficient of 1/0.58 for \( M_{spin} \), due to strong electronic correlations. We also considered corrections of 1/0.88 for \( M_{spin} \) and 1/0.49 for \( M_{orb} \) to compensate for the saturation effect in total-electron-yield mode. Here, we assumed that the x-ray attenuation length at the 2p3/2 is \( \approx 2 \) nm. Since both the \( M_{spin} (\geq 0) \) and \( M_{orb} (\geq 0) \) have the same sign in BaFeO3, it suggests the substantial weight of the \( d^\pi L^2 \) configuration in the ground state of BaFeO3, by taking Hund’s rule into account. Since BaFeO3 thin films have negative charge transfer energy, it is deduced that the \( d^\pi L^2 \) configuration is stabilized some extent besides the main component of the \( d^\pi L \) configuration, which is consistent with the CI cluster-model calculations. The magnetic moment obtained by the experimental XMCD and superconducting quantum interference device (SQUID) measurements are shown in TABLE. The XMCD result gave a smaller total magnetic moment than the value obtained by SQUID measurement. The origin of the deviation is not clear at this moment, but there is a possibility that the magnetic moment on the oxygen site might cause the underestimation based on the total magnetic moment in the iron site.

In Fig. 1(b), the calculated Fe 2p XAS (top) and XMCD (bottom) spectra are presented. The cluster-model spectrum reproduces the features of the experimental results very well, as opposed to the atomic multiplet calculation for \( d^\pi \) in Ref. 11, suggesting the importance of charge transfer effects in this system.

The O 1s XAS (top) and XMCD (bottom) spectra are presented in Fig. 4. O 1s XAS spectra of correlated compounds originate from transitions into unoccupied states with O 2p character hybridized with transition metal 3d states. Therefore, the structure of the spectrum is qualitatively related to empty bands of primarily Fe 3d weight. The structure from 528 eV to 534 eV, mainly Fe 3d-related, is similar to that of SrFeO3. The O 1s XAS of La1−xSr2FeO3 shows that a new structure below the threshold of LaFeO3 grows rapidly with increasing Sr, and dominates the spectra at high concentrations, corresponding to the Fe-related main structure in our spectrum. Those main features seen in SrFeO3 and BaFeO3 are attributed strong hybridization between Fe 3d - O 2p with \( e_g^+ \) symmetry. Such effective states are also seen in La2−xSr2CuO4 and Li2-xNi1-xO due to the anomalously strong antiferromagnetic coupling between Cu or Ni 3d and O 2p.

The O 1s XMCD spectrum directly shows that the effective state in BaFeO3 has O 2p character with the peak intensity of \( \approx 4 \) % versus that of XAS. This is the evidence of the significant O 2p hole character in the ground states due to the negative charge transfer energy and strong Fe 3d - O 2p hybridization. Since the threshold of XAS is dominated by the transition of \( d^\pi L \rightarrow 1s \), the strong intensity of XMCD was observed at the XAS structure of the lowest energy even in the usually non-magnetic oxygen. Here, 1s denotes the O 1s core hole created by the incident x-ray. Thus, when an extra electron is added to the ground state, the electron will be introduced into the ligand hole at the affinity level.

We showed that the experimental spectra of BaFeO3 are reproduced quite well by the results of CI cluster-model calculations with a common set of parameters, which can explain the magnetic ground state of BaFeO3 by comparing to the phase diagram in Ref. 12. The CI cluster-model calculations clarified that the ground state, the first affinity level and first ionization level are dominated by \( d^\pi L \) (17 % \( d^4, 65 % \), \( d^\pi L, 18 % \)) by \( d^\pi \) (71
% \text{d}^3, 26 \% \text{d}^6\text{L}, 3 \% \text{d}^1\text{L}^2) \text{ and by } \text{d}^5\text{L}^2 (2 \% \text{d}^3, 40 \% \text{d}^1\text{L}, 58 \% \text{d}^6\text{L}^2). \text{ Therefore, the band gap is characterized by } \text{d}^5\text{L} + \text{d}^5\text{L} \to \text{d}^5 + \text{d}^6\text{L}^2 \text{ and has significant O 2p character, which is schematically shown in the right panel of Fig.\textsc{[a]}}. \text{ From those results and the set of the parameters for the CI cluster-model calculation, BaFeO}_3 \text{ is classified as a covalent insulator (negative-charge-transfer-energy insulator) as proposed by Sarma \textit{et al}. (Mizokawa \textit{et al}. in the modified Zaanen-Sawatzky-Allen diagram. [1, 39–43]. Zaanen-Sawatzky-Allen diagram, which classifies the electronic properties of correlated materials systematically, was modified to consider the insulating phase due to the strong covalency within the negative-charge-transfer-energy region [40 [12].}

The \text{d}^5\text{L} configuration composing the ground state is originally a continuum and thus it tends to be metallic. The strong Fe 3d - O 2p hybridization, however, creates the \text{d}^5\text{L}-like split-off state below the continuum \text{d}^5\text{L} \text{ structure with significant stabilization, as schematically shown in the left panel in Fig.\textsc{[a]}. As a result, the oxygen hole is strongly localized despite the weak electronic correlation on the oxygen site.}

Due to the localized oxygen hole, the system becomes an insulator and the band gap opens, as shown in the right panel of Fig.\textsc{[a]}. NaCuO$_3$ (Cu$^{3+}$) has also localized oxygen holes due to anomalously strong antifer-

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**FIG. 3**: (Color online) Fe 2p XAS and XMCD spectra of BaFeO$_3$ thin films are shown in panels (a) and (b) obtained by experiment and CI cluster-model calculation, respectively.

**FIG. 4**: (Color online) O 1s XAS and XMCD spectra of BaFeO$_3$ thin films are presented.

**TABLE II**: Magnetization obtained by XMCD and SQUID. We applied the XMCD sum rules to obtain spin, magnetic and total magnetic moments from Fe 2p XMCD. The SQUID measurement is taken from Ref. [21].

|         | $M_{\text{spin}}$ | $M_{\text{orb}}$ | $M_{\text{total}}$ | $[\mu_B/f.u.]$ |
|---------|------------------|------------------|-------------------|---------------|
| XMCD    | 1.8 ± 0.2        | 0.3 ± 0.1        | 2.1 ± 0.3         |               |
| SQUID   | —                | —                | —                 | 2.7           |
romagnetic Cu 3d - O 2p hole coupling, whose split-off \( d^{5/2} \) like state with the \( ^1A_1 \) symmetry is denoted as the Zhang-Rice singlet [43].

Although the discrete split-off state is stable within a single cluster, the band gap tends to become unstable and collapse in a periodic lattice, unless the interactions between the clusters are weak, such as for the distorted bond angle structure in CaFeO\(_3\), or orthogonality of the orbitals between the clusters in NaCuO\(_2\) [42]. However, the small band gap in CaFeO\(_3\) by small distortions, unlike the strong one in NaCuO\(_2\), induces charge disproportionation by overcoming the band gap of \( d^{5/2} + d^{3/2} \rightarrow d^{5/2} + d^{3/2} \) [3, 42]. On the other hand, the non-distorted structure in SrFeO\(_3\), with the Fe-O-Fe bond angle of \( \sim 180^\circ \), completely closes the band gap to form a metal in a periodic lattice. According to the modified Zaanen-Sawatzky-Allen diagram, the boundary of the metal-insulator transition is sensitive not only to \( \Delta \) and \( U \), but also \( (pd\sigma) \) and O 2p bandwidth denoted by \( W_p \), within the negative-charge-transfer-energy regime [40, 42]. In this regime, the band gap tends to be larger as \( (pd\sigma) \) increases and as \( W_p \) decreases [42].

In the case of BaFeO\(_3\) and SrFeO\(_3\), the metal-insulator transition occurs due to the competition of the parameters of \( (pd\sigma) \) and \( W_p \). We should note that the effective one-electron bandwidth is approximately related to \( (pd\sigma) \) by \( W_p \propto (pd\sigma)^2 \) via the second-order process. Since the ratio of atomic distances in BaFeO\(_3\) to SrFeO\(_3\) is \( \left( \frac{R_{BFO}}{R_{SFO}} \right) \sim 1.03 \), we obtained the rough estimation of the ratio of the bandwidth \( (W_p)_{BFO} \) to \( (W_p)_{SFO} \) as \( \sim 0.82 \), from Harrison’s rule [29]. The band gap in BaFeO\(_3\) is explained by the significant reduction of the bandwidth compared to the relatively weaker reduction of the Fe 3d - O 2p hybridization. We can see directly the differences of those parameters in their valence-band spectra in Fig. [2]. The feature A of SrFeO\(_3\) is located at a higher binding-energy position than that of BaFeO\(_3\). Since the Fe 3d - O 2p hybridization is stronger in SrFeO\(_3\), the split-off discrete state of 4-electron systems (see left panel in Fig [5]) is more stable than that of BaFeO\(_3\). As a result, the energy difference of the ground state in 4-electron systems and feature A in 3-electron systems for SrFeO\(_3\) tends to be larger than that for BaFeO\(_3\). The broader bandwidth of feature A in SrFeO\(_3\) valence-band spectrum compared to that in BaFeO\(_3\) also suggests the stronger Fe 3d - O 2p hybridization in SrFeO\(_3\). Even in the non-distorted structure in BaFeO\(_3\) and in a periodic lattice, the band gap remains open since the interactions between the clusters are weak due to the larger lattice constant.

To summarize, as the lattice constant increases from CaFeO\(_3\) to SrFeO\(_3\), the Fe-O-Fe bond angle straightens to \( \sim 180^\circ \) and the O 2p bandwidth increases. Therefore, SrFeO\(_3\) is metallic. When the lattice constant is further increased from SrFeO\(_3\) to BaFeO\(_3\), the effect of narrowing bandwidth causes a further transition to an insulator.

The coefficient of antiferromagnetic superexchange interaction \( (J) \) greatly reduces as the lattice constant increases. By considering Harrison’s rule, we can roughly estimate the ratio of \( J \) between BaFeO\(_3\) and SrFeO\(_3\) as \( \sim \left( \frac{(J)_{BFO}}{(J)_{SFO}} \right) \sim 0.67 \) [29]. This reduction of \( J \) in BaFeO\(_3\) induces stabilization of ferromagnetic or A-type helicoidal magnetic phases, compared to SrFeO\(_3\) [12]. Mostovoy explained that the ferromagnetic phase in Fe\(^{4+}\) perovskite crystal structure with negative charge transfer energy is unstable, due to the high density of spin-flip excitations of holes from the spin-down Fermi sea to pure oxygen spin-up bands [12]. On the other hand, if the spin-flip excitation is suppressed by opening a band gap in the insulating phase, the ferromagnetic phase can be stable. Thus, the reduction of antiferromagnetic superexchange interaction and the insulating phase of BaFeO\(_3\) together stabilize the ferromagnetic phase.

IV. CONCLUSION

We investigated the electronic and magnetic properties of BaFeO\(_3\) thin films by HAXPES, XAS, XMCD and Cl cluster-model analysis. We employed the parameters for Cl cluster-model calculations on the phase boundary of ferromagnetic and A-type helicoidal magnetic phase in order to explain the magnetic properties of BaFeO\(_3\). The calculated results reproduced all of the experimental spectra quite well. The Cl cluster-model calculation suggests that BaFeO\(_3\) thin film has negative charge transfer...
energy, and its ground state is dominated by the $d^5L$ electronic configuration. The valence-band HAXPES showed the insulating property of BaFeO$_3$ thin films. The band gap has heavy O 2p character, suggesting that BaFeO$_3$ is a covalent insulator. We obtained the saturation magnetic moment of $2.1 \pm 0.3 \mu_B/\text{f.u.}$ from Fe 2p XMCD and found that it is composed of large spin magnetic moment and small orbital magnetic moment. Such a stable ferromagnetic phase under small magnetic field derives from the reduction of the antiferromagnetic superexchange interaction and the suppression of the spin-flip excitation due to the band gap in BaFeO$_3$ thin films. Thus, the metal-insulator transition and magnetic phase transition between SrFeO$_3$ and BaFeO$_3$ derive from the difference of their lattice constants, and the resultant bandwidth tuning.

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