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Probing the Antiferromagnetic-Ferromagnetic Phase Transition in $GdBaCo_2O_{5.5}$ Thin Films by Soft X-ray Magnetic Circular Dichroism

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1 Introduction

Entanglement of charge, spin, lattice and orbital degrees of freedom in transition metal oxide (TMO) systems have attracted much attention recently [1]. The electric and magnetic properties of TMOs, which are dominated by the partially filled *d* orbitals, could be strongly affected by the crystal field effect from the surrounding lattice and corresponding *d*-orbital occupation. With a strongly localized feature of *d* electrons in TMO systems (especially 3*d* TMO systems), variety of ordering phenomena like magnetic ordering, charge ordering and ferroelectricity can coexist and significantly couple with each other. Tantalizing physical phenomena with prospective applications, such as high-T_c superconductivity and colossal magnetoresistance, have emerged during the investigation of strongly correlated TMO systems.

Among 3*d* TMO systems, except the famous maganites and cuprates, cobaltites have also drawn much research attention due to their specific spin-state transition of Co ions. In many cobaltite systems, for instance the most simple case, perovskite $LaCoO_3$, the competition between crystal field splitting and Hund coupling can lead to various spin states of Co ions[2]. Take Co^{3+} as an example, with a strong octahedral crystal field, low-spin (LS, $t_{2g}^6e_g^0$, S=0) state is energetically preferable and high-spin (HS, $t_{2g}^4e_g^2$, S=2) state with occupied e_g orbital is preferred when crystal field is weak. In spite of LS and HS states, there is also debate that intermediate-spin (IS, $t_{2g}^5e_g^1$, S=1) state can also be realized in various cobaltites. The spin state of Co ions is very sensitive to the external condition such as temperature, pressure, magnetic field and substrate strain, etc. It is intensely reported that the magnetic properties of cobatites are dominated by the spin state of Co ions and complicated magnetic structures could be observed in quite a few cobaltite systems.

Recently, $REBaCo_2O_x$(5<x<6, *RE* is rare earth element) systems have been intensely investigated [3-5] for their intriguing physical properties such as high oxygen conductivity, metal-insulator transiton, giant magneto-resistance and spin-state ordering, etc.. The oxygen concentration in these systems is always variable, which leads to the high oxygen mobility within the lattice. When x=6, the cystal structure is standard double perovskite with *RE* and Ba sitting at A site, while Co sitting at B site. The nominal valence state of Co is +3.5. Due to the different ionic size of *RE* and Ba ions, A site is often ordered into layers with alternating *RE* and Ba layers. When x=5, a structure with ordered oxygen vacancy layers can be formed. When x=5.5, the oxygen vacancies order into columns along a axis, resulting in 4 non-equivalent Co sites coordinated by either normal oxygen octahedra or oxygen pyramids. With this oxygen concentration, the nominal valence state of Co is +3. In this case spin-state ordering is frequently observed. For example it is reported that in $PrBaCo_2O_{5.5+x}$ samples HS, IS and LS coexist and contribute to a complicated magnetic structure and magnetic phase transition [4].

On the other hand, phase transition with large resistivity change entangled with antiferromagnetic(AFM)-ferromagnetic (FM) transition and spin-state transition were also explored in $GdBaCo_2O_{5.5-x}$ system[5]. Magnetic ordering with an index of (0 0 1/2) was clearly observed. However, systematic investigation of Co moments and spin-states, as well as how they changes across the AFM-FM transition (~230 K) is still absent. To achieve this, soft x-ray magnetic circular dichroism (XMCD) characterization is necessary.

Despite the report of rough XMCD measurement in Ref. [5], systematic XMCD characterization of this AFM-FM transition is still absent for $GdBaCo_2O_{5.5}$ system, and even absent for all the similar materials systems. Detailed temperature/field dependent XMCD characterization will be greatly helpful to understand the complicated spin-state configuration and magnetic structure. For this purpose, we prepared high-quality epitaxial $GdBaCo_2O_{5.5}$ (GBCO) thin film sample on $SrTiO_3$(001) (STO) and $LaSrAlO_4$ (LSAO) substrates and conducted the soft x-ray XMCD measurement to investigate the phase transition behavior.

2 Experiment

We measured XAS/XMCD spectra for $GdBaCo_2O_{5.5}$ thin film samples at BL-16A of Photon Factory. Since the samples have in-plane magnetic anisotropy, a 30^o

incidence of Co *L* edge soft x-ray was used. A magnetic field of 5 T was applied during the measurement and total electron yield was detected to obtain the XAS spectra.

3 Results and Discussion

Fig. 1 shows the full Co *L* edge XAS of GBCO thin films. Intense Ba *M* edge signal can also be observed near the Co absorption edges.

Fig. 2 shows expanded Co *L* edge XAS of GBCO thin films. One can clearly see the different spectral shape of samples on different substrates at Co L_3 edge, which should originate from the different electronic structure of Co 3*d* band induced by different substrate strain.

Fig. 3 shows the Co *L* edge XMCD spectra of GBCO thin films. One can easily observe a larger XMCD signal at 250 K than that of 200 K for both samples, which shows clear sign of the AFM-FM phase transition. In addition, in the FM phase, film on STO has larger XMCD than the film on LSAO. This difference may originate from the different spin state and local environment of magnetic Co ions.

Although we successfully conducted the XMCD measurement for GBCO samples, it is hard to conduct sum-rules analysis for these data due to the overlap between Co *L* edge and Ba *M* edge.

Our experimental results confirmed the AFM-FM phase transition behavior in these GBCO thin film samples. These results are important for discussion about the electronic structure and substrate strain effect in GBCO thin films.

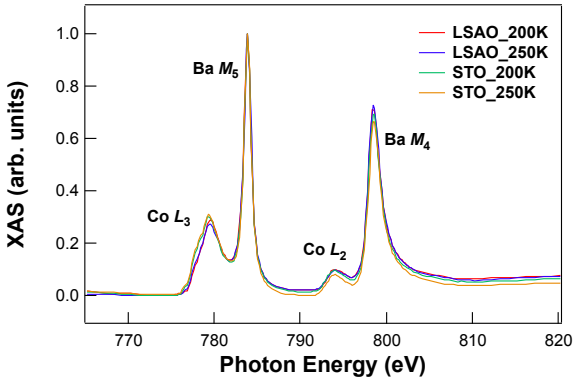

Fig. 1: Full Co *L* edge XAS of GBCO thin films.

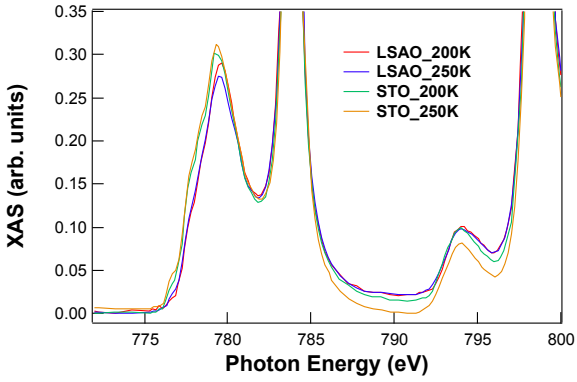

Fig. 2: Expanded Co *L* edge XAS of GBCO thin films.

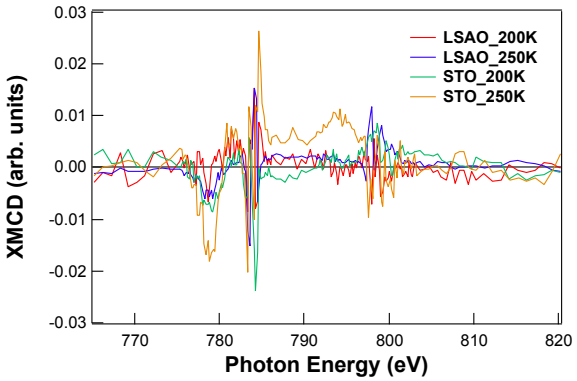

Fig. 3: Co *L* edge XMCD spectra of GBCO thin films.

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