Electronic structure and magnetic properties of magnetic dead layers in epitaxial $CoFe_2O_4/Al_2O_3/Si(111)$ films studied by X-ray magnetic circular dichroism (XMCD)

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The spin filter effect through a $CoFe_2O_4$ tunnel barrier has a potential ability for generating a highly spin-polarized electron current. Nevertheless, the experimental spin-polarization value estimated from tunnel magnetoresistance has been much less than expectation [1, 2], and many researchers have begun to point out the degradation of the electronic structure and magnetism due to spontaneously-formed structural disorder. It is plausible from the general recognition that magnetic dead layers formed at heterointerfaces cause to degrade ferrimagnetic ordering further with decreasing thickness, however, the magnetism of $CoFe_2O_4$ layers with various thicknesses have not yet systematically investigated. In this study, we present the electronic structure and magnetic properties of $CoFe_2O_4(111)$ layers with various thicknesses using XAS and XMCD.

Epitaxial $CoFe_2O_4(111)$ layers (thickness d = 1.5, 2.5, 4.3, and 12 nm) were grown on $Al_2O_3(111)/Si(111)$ using the same procedure as in the previous study [3]. In all the measurements, external field H was applied perpendicular to the film surface. Figure 1 shows the XMCD-H curves for d = 12 (blue curve) and 1.5 (red curve) nm at 300 K. The clear hysteresis for d = 12 nm indicates that the T_C of $CoFe_2O_4$ is higher than 300 K, whereas the linear line for d = 1.5 nm indicates the formation of a magnetic dead layer at the $CoFe_2O_4/Al_2O_3$ interface. Further measurement of XMCD for d = 1.5 nm revealed that the intensity at 7 T was increased by only 2.5 times when temperature was decreased from 300 to 6 K. This indicates frustration of magnetic interactions in the magnetic dead layer.

We analyzed the sites and valences of Fe cations using the experimental XMCD spectra and theoretical calculation based on the configuration-interaction (CI) cluster model [4]. Figure 2 shows the experimental Fe L_3-edge spectra (black curve) for d = 12 nm, and the corresponding best fit (red curve) with the weighted sum of the calculated spectra for Fe^{3+} (O_h), Fe^{3+} (T_d), and Fe^{2+} (O_h) sites which are plotted by blue, yellow, and green curves, respectively. Here, O_h and T_d denote octahedral and tetrahedral sites, respectively. The experimental spectra are well reproduced by the weighted sum of the calculated spectra, even the characteristic kink structure at around 708 eV. These results give strong evidence that the Fe cations only occupy the Fe^{3+} (O_h), Fe^{3+} (T_d), and Fe^{2+} (O_h) sites.

Figure 3 shows the d dependence of lattice site occupancies for Fe^{3+} (O_h), Fe^{3+} (T_d), and Fe^{2+} (O_h) estimated from the same fitting procedure. The inversion parameter y, which is defined by the chemical formula $[Co_{1-y}Fe_y]_{Td}[Fe_{2-y}Co_y]_{Oh}O_4$, gradually decreases with decreasing d and finally drops to 0.54 at d = 1.5 nm. Another striking feature is that the site occupancy for Fe^{2+} (O_h) in the sample with d = 1.5 nm is twice as large as those in the thicker samples. Thus, the decrease of d leads to the decreases in the fraction of the inverse/normal spinel structures and in the valence of the Fe cations at the O_h sites. Since the linearity of the XMCD-H curve increased with decreasing d and there is the frustration of magnetic interactions in the magnetic dead layer, the magnetic dead layer at the $CoFe_2O_4/Al_2O_3$ interface probably originates from various complex networks of superexchange interactions through the change in the crystalline and electronic structures.

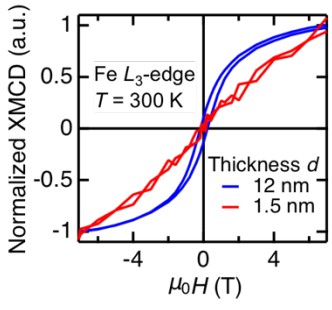

Fig. 1. XMCD-H curves of the $CoFe_2O_4$ film with d = 12 (blue curve) and 1.5 (red curve) nm.

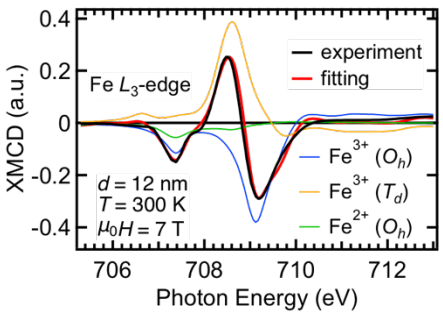

Fig. 2. XMCD spectra for the $CoFe_2O_4$ film with d = 12 nm, and the corresponding best fit from calculation.

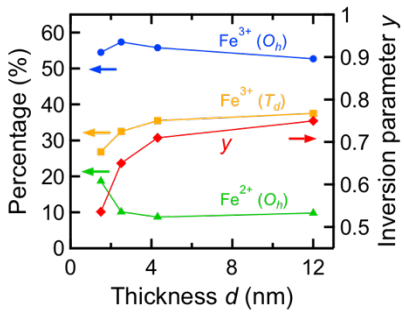

Fig. 3. d dependence of the lattice site occupancies, and inversion parameter y.

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