X-ray magnetic circular dichroism studies for Fe/Si interfaces using standing waves

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Abstract. Depth-resolved magnetic circular dichroism (MCD) spectra were measured for ferromagnetically and antiferromagnetically-coupled Fe/Si/Fe trilayer samples at the Fe L$_{2,3}$ edges using X-ray standing wave technique. Physicochemical state of the interdiffused layer of the samples was also estimated using soft X-ray fluorescence (SXF) spectroscopy. It was found for both the trilayers that enhancement in the orbital moment due to the interface anisotropy occurs at the Fe$_3$Si layer in the interdiffused layer. This means that SXF measurement enables us to predict to some extent magnetic state of buried interfaces in terms of the chemical state.

1. Introduction
Magnetic multilayers have been widely studied from the viewpoint of the controllability of the interlayer exchange coupling without magnetic field. Particularly the Fe/Si system has attracted much interest for the strong antiferromagnetic (AF) coupling across thin Si layers, because it stimulates future applications to AF coupled storage media for hard disks. For the origin of the interaction, several models such as phonon-assisted hopping electrons in the localized states of amorphous Si (a-Si) [1], RKKY interaction [2-5], and thermally excited carriers in the narrow-gap semiconducting spacer [6] have been proposed. Thus the models so far proposed for Fe/Si multilayers have been quite controversial. The key point of the problem is the severe interdiffusion at the interface and thus the complex diffused structure of the Si spacer. Interdiffusion at the interface throughout layer deposition is to some extent avoidable by high-speed deposition. Endo, Kitakami, and Shimada [7] deposited Fe/Si multilayers at a high rate, and achieved a drastic enhancement in the interlayer exchange coupling. They confirmed that the spacer layer is insulating from the temperature dependence of the electrical resistivity, and thus explained the interlayer coupling using the quantum interference model formalized by Bruno [8]. However, they have not clarified as to which material remains to mediate the interlayer coupling and thus the origin of the enhancement. Their result really motivated us, and in the previous study [9], we studied the interfacial layers of the Fe/Si multilayers nondestructively using soft-X-ray fluorescence (SXF) spectroscopy, and have made clear for the highly AF coupled multilayer that the amorphous Si (a-Si) layer of 1.3 nm thick changed in its middle region into a-FeSi$_2$ of 0.7 nm thick and no a-Si remained. The a-FeSi$_2$ middle layer was sandwiched with a-Fe$_3$Si layers of 0.5 nm thick. It is quite reasonable that these synthesized layers should be referred to thin film materials rather than bulk one. According to the data for thin films [10] Fe$_3$Si is metallic and ferromagnetic (F), while FeSi$_2$ is insulating and nonmagnetic. Therefore, the a-FeSi$_2$ spacer mediates
the strong AF exchange coupling in the Fe/Si multilayer. The FeSi
layer is thought to be active in the magnetic coupling in terms of a quantum interference model. In the study, the layer structure of the interface is deduced from the physicochemical view point. However, it has not yet been confirmed that the local magnetic state in the interdiffused layer is consistent with the result of the SXF studies. As a next step depth-resolved magnetic property of the interdiffused layer should be investigated. For the studies X-ray magnetic circular dichroism (MCD) spectroscopy combined with soft X-ray standing wave (SW) is promising [11]. Actually, Kim and Kortright used the SW technique in conjunction with MCD for a Pd/Co/Pd trilayer, and found that large enhancements of the number of Co d holes, and of in-plane orbital and spin magnetic moments, are strongly localized at a thin, chemically modified interface layer. However, they have not yet resolved the chemical state of the interface layer responsible for the magnetic anisotropy. In this paper we describe the consistency of physicochemical structure of the Fe/Si interface determined using SXF spectroscopy and its magnetic property resolved using MCD spectroscopy combined with soft X-ray SW technique, and also discuss the model of the magnetic interlayer coupling.

2. Experimental

SW is formed by interference effect between incident and reflected X-ray waves in a multilayer. The effect is the most remarkable when the Bragg condition \( \lambda = 2d \sin \theta \) is satisfied, where \( \lambda \) is the wavelength of the incident X-ray, \( \theta \) the grazing angle of incidence and \( d \) the multilayer periodicity (d-spacing). From a simulation study it was confirmed that the SW suffers merely a slight perturbation when the multilayer is capped with thin layers. That is, the SW depends almost on the multilayer itself and a little on additional thin layers. Thus we can deposit thin layers on a multilayer as a sample of interest to investigate its depth profile. In this case, the multilayer acts as a standing wave generator (SWG). SWG should be designed to have high reflectance for the incident X-rays because a strong SW field is formed in the entire sample/SWG structure. In this study W/B\textsubscript{4}C multilayer was chosen as a SWG because of high reflectance at the Fe L\textsubscript{2,3} absorption thresholds. The SWG was designed to satisfy the Bragg condition at a grazing angle of about 15° because in-plane magnetization is concerned with the interlayer coupling in this study.

W/B\textsubscript{4}C multilayers were fabricated on Si wafers at ambient temperature using a magnetron sputter system. The base pressure was lower than 1.3×10\textsuperscript{-4} Pa (1.0×10\textsuperscript{-6} Torr) and the Ar gas pressure was 0.27 Pa (2.0 mTorr). Their d-spacing was examined to be 3.53 nm using small-angle X-ray diffraction. Fe/Si(1.35nm)/Fe(2.94nm) and Fe/Si(0.85nm)/Fe(3.43nm) trilayers were prepared on the W/B\textsubscript{4}C multilayers. As is noticed, sum of thickness of Si layer and that of the second Fe layer is approximately 4.3 nm so that the SW antinode would stay around the first Fe/Si interface. Thickness of the topmost Fe layer is 0.98 nm to prevent the photoelectron emission from being suppressed. The trilayer samples were deposited using the magnetron sputter system, where the film thickness was controlled with the deposition time based on deposition rates of 0.041 nm/s and 0.028 nm/s for Fe and Si, respectively. All the samples were covered with a 2.0 nm thick C film without breaking the vacuum of the sputter system. Thickness of 2.0 nm for the C film was confirmed to be enough to prevent the topmost Fe layer from oxidation using X-ray photoelectron spectroscopy.

Fe(2.94nm)/Si(t) (t = 0.85, 1.35 nm) multilayers were also prepared using the same sputter system to examine the Si-layer dependence of the interlayer magnetic coupling. Using a vibrating sample magnetometer, it was found that the Fe/Si(0.85nm) multilayer was AF and the Fe/Si(1.35nm) multilayer was F coupled. From the results it is reasonable to conclude that the coupling mode of the Fe/Si(0.85nm)/Fe and the Fe/Si(1.35nm)/Fe trilayers are AF and F coupled, respectively. Physicochemical state of the interdiffused layer was also analyzed using SXF spectroscopy. Si layer thickness of the Fe/Si multilayer in AF coupling mode is 0.85 nm as thin as compared with 1.3 nm in the previous study [9]. A possible reason for it is the difference in the conditions such as deposition rate and Ar gas pressure due to the different sputter systems between the studies, but has not been resolved exactly.
Fe $L_{2,3}$ absorption measurement for the samples C/Fe/Si(1.35nm)/Fe/SWG was carried out for circularly-polarized soft X-rays from an undulator at AR-NE1B [12] of the Photon Factory, where monochromatized light of 0.35 eV in energy width at 700 eV with a degree of circular polarization of 98% is available. Magnetic field of $\pm$1.06 T oriented along the incident beam was applied to the sample using an assembly of permanent magnet units antiparallel to each other. X-ray MCD is defined as difference between absorptions measured when the helicity of the incident beam (photon spin) and the sample magnetization are parallel and antiparallel, respectively. An MCD spectrum measured with reversed helicity as well as magnetization is equivalent to that as measured with their initial orientations. Therefore, MCD spectra are obtained by reversing the helicity with a constant magnetization and vice versa. In this study the latter measurement was employed. Helicity-dependent absorption spectra were measured under opposite magnetic fields for a fixed circular polarization by total electron yield via sample drain current from 690 to 750 eV. They were actually recorded at each photon energy $h\nu$ for the alternative applied fields by switching the magnet units. Absorption spectra were measured for grazing angles of incidence from 12.0° through to 16.0° (18.0°) at a 0.2° interval. An angle of incidence of 0° was settled when incident direct beam passing by the sample surface was the highest in intensity.

Absorption spectra are normalized to obtain magnetic moments per atom so that the absorption intensity far above the absorption edge should be higher by one than just below it. Figure 1 illustrates twice the helicity-averaged Fe $L_{2,3}$ absorption spectra on a ($\theta$, $h\nu$) surface measured for the Fe/Si(1.35nm)/Fe sample, where each spectrum is sum of the helicity-dependent spectra, and the step is twice. Familiar Fe $L_3$ and $L_2$ peaks are observed at 710 and 722 eV, respectively. As is noticed, the absorption spectrum suffers from no oxidation. Fe $L_{2,3}$ MCD spectra on a ($\theta$, $h\nu$) surface obtained from the helicity-dependent spectra are illustrated in Fig. 2. A deep dip and a peak appear at the Fe $L_3$ and $L_2$ edges, respectively.

The SW antinode is not stationary during $h\nu$ scan to record an absorption spectrum for a value of $q$. From a simulation study it is also confirmed that for a scattering vector $q = 4\pi \sin \theta / \lambda$, the SW antinode is tuned to a layer at an almost constant depth. The scattering vector is essential to obtain information of a layer at a constant depth. From the surface of the Fe $L_{2,3}$ MCD spectra on a ($\theta$, $h\nu$) surface as shown in Fig. 2, we calculated MCD spectra on a ($q$, $h\nu$) surface by interpolation based on the scattering vector. Figure 3 shows the MCD spectra on a ($q$, $h\nu$) surface obtained for the

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Fig. 1. Helicity-averaged Fe $L_{2,3}$ absorption spectra plotted on a ($\theta$, $h\nu$) plane for the Fe/Si(1.35nm)/Fe trilayer.

Fig. 2. Fe $L_{2,3}$ MCD spectra plotted on a ($\theta$, $h\nu$) surface. The insertion illustrates the MCD surface viewed from another side.

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Fig. 3. Fe $L_{2,3}$ MCD spectra plotted on a $(q, h\nu)$ surface for the Fe/Si(1.35nm)/Fe sample. The insertion illustrates the MCD surface viewed from another side.

Fe/Si(1.35nm)/Fe sample, where the insertion illustrates the MCD surface viewed from another side. For these MCD spectra we obtained depth resolved magnetic moments $m_{\text{orb}}$ and $m_{\text{spin}}$ by applying the magnetic sum rules [13]:

$$m_{\text{orb}} = -2(A + B)/C$$
$$m_{\text{spin}} = -3(A - 2B)/C,$$

where $A$ and $B$ are the areas of the MCD peaks and $C$ is a constant.

3. Results and discussion

Orbital moments thus obtained for the Fe/Si(1.35nm)/Fe sample are plotted versus depth from the surface $z$ in the graph in Fig. 4, where design depths of center of the Si layer and the Fe/Si interfaces are indicated with lines at $z = 3.7$ nm, and at $z = 3.0$ and 4.4 nm, respectively. The orbital moment keeps a constant about 0.04 $\mu_B$ between $z = 3.0$ and 4.2 nm, while it increases to 0.07 $\mu_B$ and 0.13 $\mu_B$ around $z = 2.6$ nm and 4.3 nm, respectively. The orbital moment of 0.07 $\mu_B$ at $z = 2.6$ nm is lower than 0.12 $\mu_B$, which was obtained for an Fe thin film of 9.8 nm capped with a C film in this study. The enhancement around $z = 4.3$ nm is not understood because the SW antinode is not dominant here and it is too deep for photoelectrons to escape from surface. Because of the SW configuration, contribution to the moment from layers covered with the SW node is also counted. Therefore, relative change in the moment should be taken into account rather than the absolute value. Interdiffused layer of the Fe/Si(1.35nm)/Fe sample resolved using SXF technique in this study is also illustrated for comparison in Fig. 4, where the center of the Si layer is adjusted to that of the upper graph. As is shown, initial Si layer has so much changed into silicides that the Si layer is not remained and an FeSi$_2$ layer of 0.5 nm

Fig. 4. Orbital moment vs. depth $z$ obtained for the Fe/Si(1.35nm)/Fe sample (top), and its interdiffused layer analyzed using SXF technique (bottom).
thick is sandwiched with FeSi and Fe\textsubscript{3}Si layers of 0.1 nm and 0.7 nm thick, respectively. The result for the silicides in the interdiffused layer is almost consistent with the previous study except for the contribution of FeSi layer. As is noticed in Fig. 4, the orbital moment is low over the FeSi\textsubscript{2} and FeSi\textsubscript{2} layers, while it shows peaks inside the Fe\textsubscript{3}Si layers. Fe\textsubscript{2}Si film is F, while FeSi\textsubscript{2} and FeSi films are both nonmagnetic [10]. Therefore, the result is quite reasonable because magnetization occurs inside the Fe\textsubscript{3}Si layers as well as in the Fe layers. The enhancement of the moment is interpreted to localize strongly at the Fe\textsubscript{3}Si layer due to the interface anisotropy. Depth-resolved orbital moment and interdiffused layer obtained for the Fe/Si(0.85 nm)/Fe sample are shown in Fig. 5. The orbital moment is 0.02 $\mu_B$ near the center of the Si layer and increases in both the Fe\textsubscript{3}Si layers to 0.05 and 0.07 $\mu_B$ at $z = 2.7$ nm and $z = 3.9$ nm, respectively. The moment is low inside the nonmagnetic spacer layers and increases in both the Fe\textsubscript{3}Si layers, which is quite similar to the Fe/Si(1.35 nm)/Fe sample. However, absolute value of the moment is obviously smaller than that of the Fe/Si(1.35 nm)/Fe sample. The reason for this is that magnetization of the Fe/Si(0.85 nm)/Fe trilayer is not saturated with the applied field in this study due to its AF coupling, while the Fe/Si(1.35 nm)/Fe sample is saturated easily owing to the F coupling.

Enhancement of the orbital moment occurs at the Fe\textsubscript{3}Si layers in the Fe/Si interfaces, which suggests strongly that magnetization occurs outside the nonmagnetic silicides such as FeSi and FeSi\textsubscript{2} layers. Moreover, respective magnetization occurs outside the nonmagnetic silicide layers for the AF and F-coupled Fe/Si/Fe trilayers. Magnetized layers assigned by the depth-resolved MCD study correspond well to the interdiffused layer analyzed using SXF technique. This means that we can predict depth-resolved magnetic state of buried interfaces by means of SXF measurements to some extent without X-ray MCD study, which needs circularly-polarized X-rays.

Concerning the AF-coupled trilayer, there seems to occur no particular magnetization such as site shift toward the nonmagnetic silicic layers, which was thought to be a possible reason for the strong interlayer coupling. With respect to the model of the magnetic interlayer coupling, quantum interference model is reasonable as discussed in the previous study that the FeSi\textsubscript{2} spacer mediates the strong AF exchange coupling in the Fe/Si multilayer [9]. The FeSi\textsubscript{2} layer is thought to be active in the interlayer coupling of F layers consisting of Fe\textsubscript{3}Si and Fe in terms of a quantum interference model.

4. Conclusions

Depth-resolved Fe $L_{2,3}$ MCD spectra were measured for an AF-coupled Fe/Si(0.85 nm)/Fe trilayer and a F-coupled Fe/Si(1.35 nm)/Fe sample using X-ray SW technique. Physicochemical state of the interdiffused layer of the samples was also estimated using SXF spectroscopy. It was found for both the trilayers that enhancement in the orbital moment due to the interface anisotropy occurs at the Fe\textsubscript{3}Si layer in the interdiffused layer. This result suggests strongly that we can get information about depth-resolved magnetic state of buried interfaces from the physicochemical state obtained by means of SXF.
measurements to some extent. As for the AF-coupled sample, there seems to occur no particular magnetization different from that for the F-coupled one.

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