5 Surface/Interface Magnetism Project

– Crystalline, magnetic and electronic structures at the surface and interface of magnetic thin films and multilayers –

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5-1 Introduction

The surface and interface of magnetic thin films play essential roles in the appearance of extraordinary magnetic properties such as perpendicular magnetic anisotropy (PMA) and the giant magnetoresistance effect. We are investigating the crystalline, magnetic and electronic structures at the surface and interface of magnetic thin films and multilayers, in order to reveal the origin of fascinating magnetic properties that cannot be realized in bulk materials. For example, we have studied magnetic anisotropy of Fe/Ni multilayers [1-4], magnetic structures at the interface between antiferromagnetic FeMn and ferromagnetic Ni [5], effects of ion irradiation on ultrathin films [6,7], and a voltage-induced change in magnetic anisotropy of FeCo thin films grown on a ferroelectric substrate, by means of X-ray magnetic circular dichroism (XMCD), extended X-ray absorption fine structure (EXAFS), and polarized neutron reflectivity (PNR) techniques. We also plan to perform muon spin rotation experiments using an ultra-slow muon source.

5-2 Scientific topics: Anomalous magnetic structure at the interface between ferromagnetic Ni and antiferromagnetic FeMn [5]

Magnetic interactions between antiferromagnetic (AFM) and ferromagnetic (FM) materials have attracted much interest owing to the extraordinary effects observed at their interface. Among such interactions, the magnetic anisotropy interaction can be used to control the magnetic direction of ultrathin films. It has been reported that the magnetic easy axis of Ni films in FeMn/Ni/Cu(100) bilayers switches from the perpendicular to in-plane directions as the FeMn layer exhibits the AFM feature [8]. The FeMn-induced change in magnetic anisotropy was attributed to the interfacial magnetic frustration between Ni and FeMn as follows: Each FeMn plane has a net magnetic moment in the perpendicular direction, and those in adjacent FeMn planes are aligned antiparallel to each other. When the FeMn/Ni interface has a monolayer step structure, the magnetic moments in the Ni layer are parallel or antiparallel to those in the adjacent FeMn plane, depending on the lateral position. This induces a frustration, leading to in-plane magnetization in the FM Ni layer.

Therefore, some noncollinear magnetic structure in the FM Ni layer might be expected when the Ni layer undergoes the magnetic anisotropy interaction at the interface with the AFM FeMn layer. For instance, if the Ni film itself exhibits perpendicular magnetization whereas the magnetic anisotropy interaction with the FeMn layer encourages in-plane magnetization, the spin moment in the Ni layer might be canted to the in-plane direction around the interface with the FeMn layer. The direct observation of such noncollinear magnetic structure in ultrathin films has not been achieved, however, due to the lack of a depth-resolved technique. We have investigated the magnetic depth profile of Ni films in FeMn/Ni/Cu(100) by means of the depth-resolved XMCD [9,10] and PNR techniques, and have revealed the twisted magnetic structure in the ultrathin Ni layer.

Figure 1(a) shows Ni L-edge depth-resolved XMCD data for perpendicularly magnetized FeMn/Ni(16 ML)/Cu(100) films, taken in the normal incidence condition. Only the perpendicular component of Ni magnetization is observed at normal incidence. The depth-resolved XMCD data were recorded *in situ* at the undulator beamline BL-

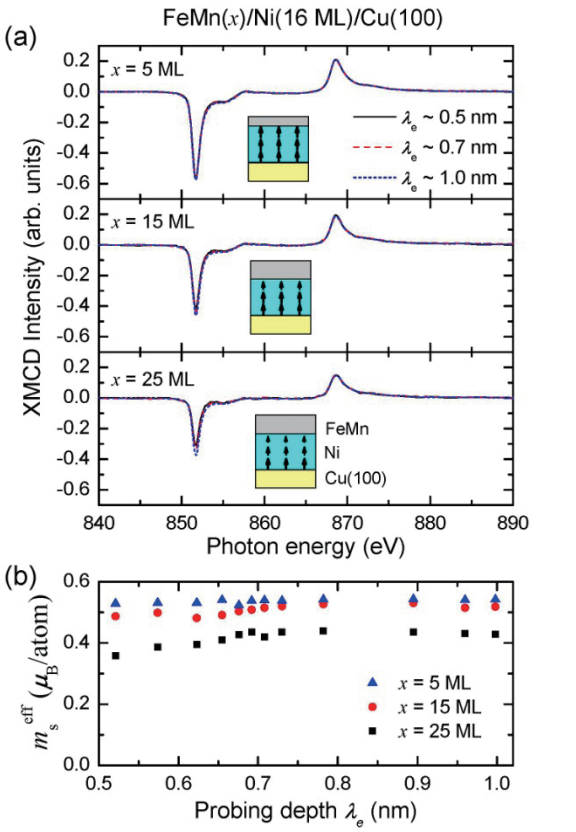

Fig. 1: Probing depth (λ_e) dependence of (a) normal-incidence Ni L-edge XMCD spectrum for FeMn(x)/Ni(16 ML)/Cu(100) and (b) averaged effective spin magnetic moment m_s^{eff} estimated by applying the sum rules to each spectrum.

16A at the Photon Factory, just after the growth of FeMn/Ni films on a Cu(100) single crystal in the same ultrahigh vacuum chamber. A pulsed magnetic field of 500–2000 Oe was applied before the measurement, and the magnet was retracted during the measurement, so that the remanent magnetization was detected. When the FeMn layer is in the paramagnetic state at 5 ML, the XMCD intensity is independent of the probing depth, λ_e, which directly indicates that the Ni film has a uniform magnetization. On the other hand, the XMCD intensity slightly decreases as λ_e decreases, i.e., the surface sensitivity increases, when the FeMn film starts to exhibit the AFM state at ~15 ML. It is suggested that the perpendicular magnetization component in Ni decreases around the interface with FeMn. This trend becomes more prominent at 25 ML FeMn, where the FeMn layer should be in a strong AFM state. Then, the perpendicular magnetization component is quantitatively estimated by applying the sum rules [11,12] to each XMCD spectrum, and is plotted in Fig. 1(b) as a function of λ_e. Note that the probing depth λ_e corresponds to the effective escape depth of the emitted electrons. Since the XMCD technique yields an averaged magnetic moment per atom, the estimated magnetic moment is an average over the whole Ni layer weighted by a factor of $\exp(-z/\lambda_e)$, where z represents the depth from the top of the Ni layer. It is thus revealed that the perpendicular magnetization component in perpendicularly magnetized Ni films decreases around the interface with the FeMn layer when FeMn exhibits the AFM state.

Next, we discuss the in-plane magnetized film. Figure 2(a) shows Ni L-edge depth-resolved XMCD data for an in-plane magnetized FeMn(15 ML)/Ni(6.5 ML)/Cu(100) film, taken in the grazing incidence condition. The in-plane magnetization component is mainly observed at grazing incidence. In contrast to the perpendicularly magnetized films, the XMCD intensity is almost independent of λ_e, though the FeMn layer should be in the AFM state at 15 ML. Such discrepancy between the in-plane and perpendicular magnetized films suggests that the magnetic moment in Ni films tends to be oriented to the in-plane direction around the interface with FeMn by a magnetic anisotropy interaction with AFM FeMn. That is, the magnetic moment in perpendicularly magnetized Ni films might be canted to the in-plane direction around

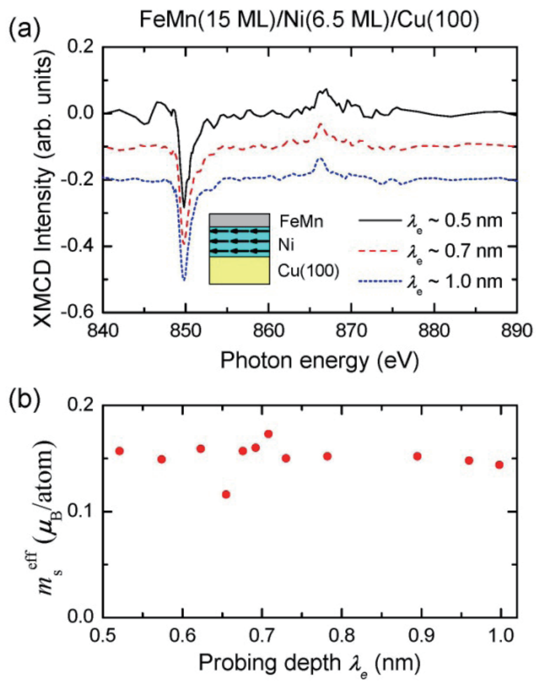

Fig. 2: Probing depth (λ_e) dependence of (a) grazing-incidence Ni L-edge XMCD spectrum for FeMn(15 ML)/Ni(16 ML)/Cu(100) and (b) averaged effective spin magnetic moment m_s^{eff} estimated by applying the sum rules to each spectrum.

the interface with AFM FeMn, whereas that in in-plane magnetized Ni films does not have to rotate because it is already in the in-plane direction.

It is thus suggested that the spin moment in Ni is twisted from the perpendicular to the in-plane directions towards the interface with AFM FeMn. To confirm the twisted magnetic structure, it is necessary to detect the in-plane magnetization component in Ni around the interface with FeMn for the perpendicularly magnetized film. However, it is likely that the canted spin moments around the interface form some domain structure in the remanent state, resulting in no net in-plane moment. Therefore, it is difficult to directly observe the canted spin moments by the depth-resolved XMCD technique because the technique can be applied only to the remanent magnetization state.

To investigate the magnetic depth profile under a magnetic field, the PNR technique was applied to a perpendicularly magnetized FeMn/Ni/Cu(100) film. For the PNR measurement, homogeneous films were prepared on a Cu single crystal, and the films were capped with a 50–100 monolayer (ML) Cu overlayer to protect them from the air. The PNR experiment was carried out at BL-17 (SHARAKU) in the Materials and Life Science Experimental Facility, Japan Proton Accelerator Research Complex, using a pulsed neutron source (Fig. 3). The magnetic field and the neutron polarization are in the in-plane direction of the film, whereas the remanent magnetization of Ni is in the perpendicular direction. Therefore, only the in-plane magnetization component induced by the magnetic field is detected. Figure 4 shows the PNR curve at a magnetic field of 1 kOe, which is much smaller than the saturation field, ~5 kOe. The obtained data was

Fig. 3: First PNR data for Cu/FeMn/Ni/Cu(100) taken at BL-17 (SHARAKU).

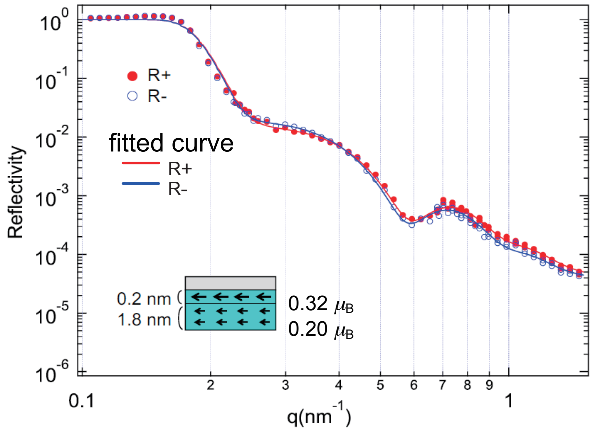

Fig. 4: Observed and fitted PNR curves, R+ and R–, for Cu(74 ML)/FeMn(21 ML)/Ni(11 ML)/Cu(100) film measured at a magnetic field of 1 kOe. The magnetic field is parallel to the quantization axis of the neutron and is perpendicular to the surface normal direction. The magnetic structure model used in the fitting procedure is shown in the inset.

fitted by using a two-region model in which the Ni film is separated into two parts, because the interface part is expected to have a different in-plane magnetization component from that in the inner part under a weak magnetic field. The magnetic structure of AFM FeMn was not considered in the fitting procedure. The magnetic moments of the interface and inner parts were optimized, as well as the thickness of each part. The estimated Ni magnetic moments in the top and bottom parts are 0.32 and 0.20 μB/atom, respectively. By combining this result with the depth-resolved data, it is suggested that the magnetic moment in Ni rotates from the perpendicular to the in-plane direction as approaching the top interface, as depicted in Fig. 5. This model is consistently supported by the assumption that the step-induced magnetic frustration between AFM FeMn and FM Ni enhances in-plane magnetization in the Ni layer [8].

Thus we have investigated the anomalous magnetic structure around the interface between FM Ni and AFM FeMn layers by a combination of the depth-resolved XMCD and the PNR techniques. The depth-resolved XMCD data has shown that the perpendicular component of the Ni film decreases around the interface with FeMn compared to the inner part of the film, when the film exhibits perpendicular magnetization. On the other hand, the in-plane component is kept constant through the whole film in the case of in-plane magnetization. Moreover, the PNR data under a weak in-plane magnetic field for a perpendicularly magnetized film reveal that the in-plane magnetic

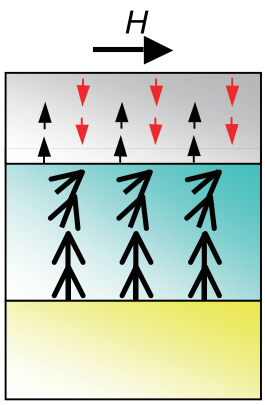

Fig. 5: Schematic illustration of magnetic structure of FeMn/Ni/Cu(100) under a weak in-plane magnetic field, *H*. The Ni magnetic moment gradually rotates from the perpendicular to in-plane directions as approaching the interface to AFM FeMn.

component in Ni is larger at the interface with FeMn than that in the inner part of the Ni layer. These results consistently suggest that the Ni spin moment is canted to the in-plane direction around the interface with FeMn.

5-3 Voltage-induced change in magnetization of ferromagnetic films grown on ferroelectric substrates

One of the most usual ways to control the direction of the spin moment in a magnetic material is to apply a magnetic field generated by an electromagnet. It requires a relatively large current to generate a magnetic field. On the other hand, it has been shown that the spin direction can be switched by a spin polarized current, in which a much smaller current is required compared with the switching by a magnetic field especially in the case of nanometer-scale magnets such as those in a magnetic recording device. Moreover, electric field-induced magnetization switching was recently demonstrated [13], which has a great potential to reduce the energy consumed in the magnetization switching process.

The idea of controlling magnetization by an electric field has been proposed as the "multiferroic" concept [14]. For instance, $BiFeO_3$ has been extensively investigated as one of the most promising multiferroic materials. Unfortunately, the temperature at which such materials show the multiferroic property is usually far below room temperature in the case of single compound materials. On the other hand, it was reported that the coercive field of a ferromagnetic Fe thin film grown on a ferroelectric $BaTiO_3$ substrate changes when a voltage is applied between the Fe film and the bottom of the substrate even at room temperature [15]. Since the mechanism of the effects of the voltage on the magnetic properties is not well understood, we have started XMCD, EXAFS, and PNR studies on the effects of voltage on ferromagnetic thin films grown on ferroelectric substrates.

The thin films are grown on a ferroelectric substrate, which is mounted on a sample holder as shown in Fig. 6, in a high vacuum chamber. The substrate temperature during the film growth can be controlled to optimize the growth condition. A capping layer, e.g. Au, is deposited on the film to protect the film from the air, and the sample is removed from the vacuum chamber to make an electric contact with the film. The XMCD measurement is performed by using the same holder, by which up to ±1 kV is applied during the measurement between the film and the bottom of the substrate.

Figures 7 and 8 show preliminary results for a ~4 nm FeCo thin film grown on a $BaTiO_3$ substrate. The sign of the applied voltage is defined with respect to the bottom of the substrate. The magnetization curve shows a large dependence on the applied voltage as indicated in Fig. 7. The film shows a square loop at negative voltages, while an

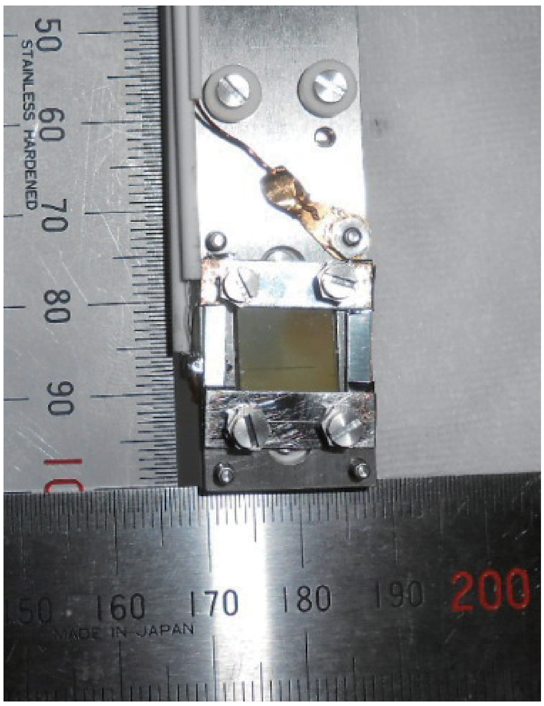

Fig. 6: $BaTiO_3$ substrate mounted on a sample holder for the XMCD measurement, by which up to ±1 kV is applied between the surface and the bottom of the substrate.

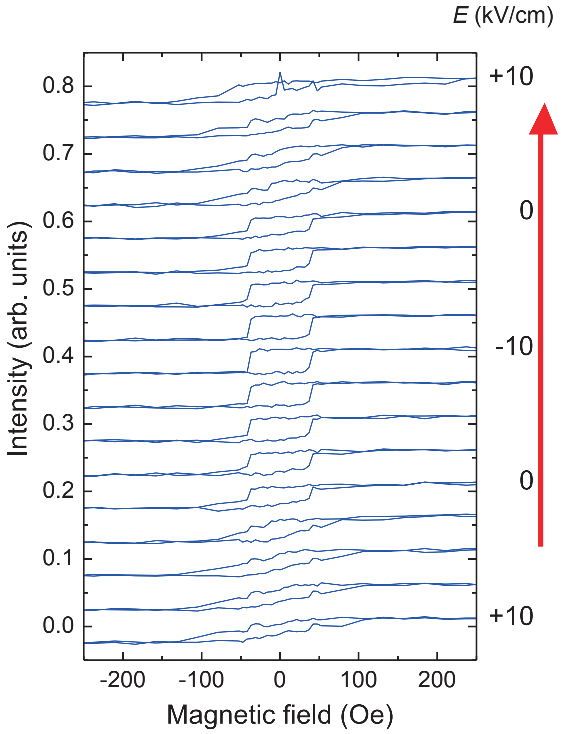

Fig. 7: Applied voltage dependence of the magnetization curve for FeCo/$BaTiO_3$(001) taken by using Fe L-edge XMCD at the grazing incidence configuration at room temperature.

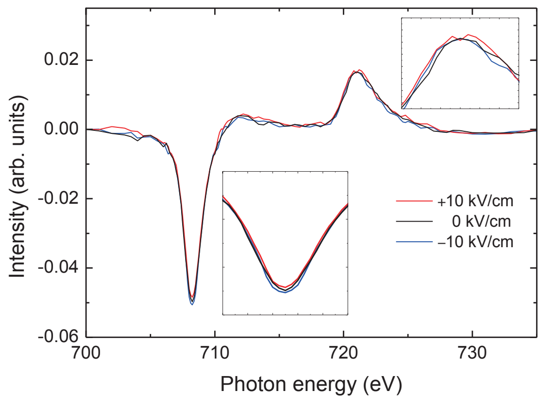

Fig. 8: Fe L-edge XMCD spectra taken by applying different voltages to FeCo/$BaTiO_3$(001).

inclined loop is seen at positive voltages. Such a difference in the magnetization curves can be interpreted as a change in magnetic anisotropy of the film from in plane at positive voltages to perpendicular at negative voltages.

Figure 8 shows Fe L-edge XMCD spectra taken at different applied voltages. The overall shape of the spectra seem similar to each other, but it is found that the XMCD intensity at the L_3 edge (~708 eV) is smaller at negative voltage, whereas an opposite trend is seen at the L_2 edge (~721 eV), though the data quality is too poor to deduce a firm conclusion. This suggests that the in-plane orbital magnetic moment in the FeCo film is larger at negative voltages, which may be related to the larger in-plane magnetic anisotropy. Further investigations including EXAFS and PNR experiments are now under way.

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