Interface-Driven Noncollinear Magnetic Structure and Phase Transition of Fe Thin Films

We investigated the magnetic structure and its temperature dependence of Fe thin films grown on the MgO(001) surface by means of nuclear resonant X-ray scattering (NRS). By fabricating Fe films δ-doped with ^{57}Fe, depth-resolved analysis of the magnetic structure was performed. The Fe film was found to have a noncollinear magnetic structure, which was also confirmed with first-principles calculations. Moreover, the magnetic phase transition is suggested to start at the interface at a lower temperature than the entire film.

The magnetic structure of magnetic thin films is of fundamental interest as well as of practical importance for magnetic storage devices. One particular feature of a thin magnetic film is that the symmetry breaking at the interface causes the interface magnetocrystalline anisotropy through the spin-orbit interaction, which might compete with the bulk magnetocrystalline anisotropy and shape anisotropy. When competing interactions are present, the magnetization is not necessarily uniform in the entire film, and a noncollinear magnetic structure might appear [1, 2]. The phase transition behavior at the interface, furthermore, could be different from that of the bulk due to the modification of the exchange interaction at the interface [3].

In our previous study, we investigated the magnetization of Fe films with NRS of X-rays in a glancing incidence condition, which revealed magnetization canting at the surface of the film [4]. In the present report, we demonstrate direct depth-resolved analysis of the magnetic structure of Fe films grown on MgO(001). NRS-sensitive-^{57}Fe was δ-doped either at the interface or in the middle of the film, where the magnetization at the interface or middle of the film was selectively probed by NRS [5]. The experiment was performed at AR-NE1A.

The hyperfine structure of the ^{57}Fe nucleus can be measured as quantum beats in the time spectrum of NRS. The direction of the internal magnetic field B_{hf} at the ^{57}Fe nucleus site is analyzed on the basis of the selection rule for the magnetic-dipole transition depending on the incident direction k_{SR} and the polarization direction of magnetic field H_{SR} of the incident X-ray [6]. For α-Fe with B_{hf} of 33 T, the main quantum beat frequencies in the cases of ($B_{hf} \perp H_{SR}$ and $B_{hf} \perp k_{SR}$), ($B_{hf} \parallel H_{SR}$ and $B_{hf} \perp k_{SR}$) and ($B_{hf} \perp H_{SR}$ and $B_{hf} \parallel k_{SR}$) are (124 and 72), (72) and (72 MHz), respectively.

Figure 1 shows the NRS time spectra and the results of frequency analysis of the sample ^{56}Fe (20 nm)/^{57}Fe (1 nm)/MgO(001). In **Fig. 1 e-h**, one quantum beat frequency of 72 MHz is dominant at all azimuthal angles, which indicates the existence of the perpendicular magnetization component. The experiments for the sample ^{56}Fe (10 nm)/^{57}Fe (1 nm)/^{56}Fe (10 nm)/MgO(001), on the other hand, clearly showed the existence of the parallel magnetization component. From these results, the angles of the magnetization direction at the interface and middle of the film with respect to the surface parallel direction were estimated at 58±8° and 6±5°. This suggests that the Fe film on MgO(001) has a noncollinear magnetic structure with the magnetization canted near the interface. We also conducted similar experiments for the Fe films grown on Al_2O_3(0001). In contrast to the results for the film on MgO(001), the magnetization direction was found to be mainly parallel to the interface both at the interface and middle of the film.

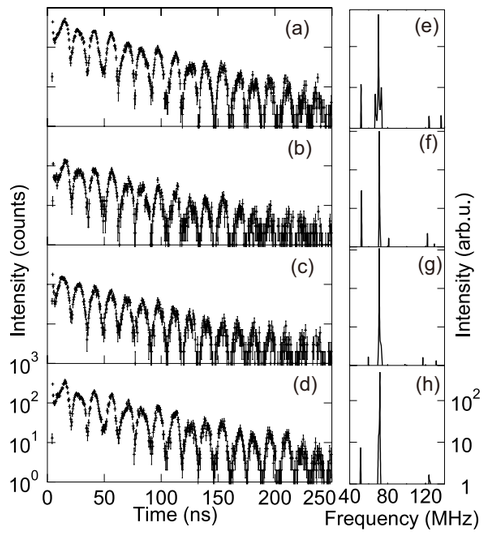

Figure 1 Time spectra of the nuclear resonant scattering for ^{56}Fe/^{57}Fe/MgO(001) at a glancing angle of 5.2 mrad and azimuths of **(a)** MgO[1$\bar{1}$0], **(b)** MgO[010], **(c)** MgO[110] and **(d)** MgO[100]. The frequency spectra obtained for **(a)-(d)** are shown in **(e)-(h)**, respectively.

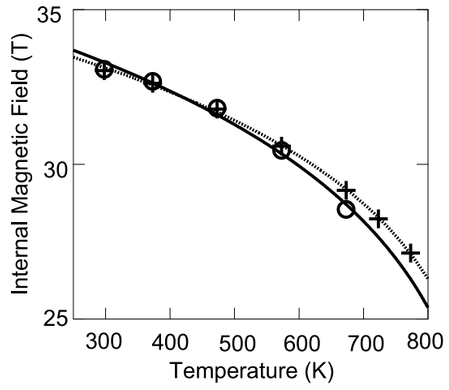

Figure 2 Temperature dependence of B_{hf} taken for ^{56}Fe/^{57}Fe/MgO(001) (○) and ^{56}Fe/^{57}Fe/^{56}Fe/MgO(001) (+). Solid and dotted curves are fits with a common T_c value to the data of the former and latter samples, respectively.

The temperature dependence of the B_{hf} value for ^{56}Fe/^{57}Fe/MgO(001) and ^{56}Fe/^{57}Fe/^{56}Fe/MgO(001) was evaluated by frequency analysis, which is plotted as a function of the sample temperature in **Fig. 2**. The experimental data suggest that B_{hf} at the interface becomes smaller than that of the middle of the film with increasing temperature, suggesting that the magnetic phase transition starts at the interface at a lower temperature than the entire film.

The detailed noncollinear magnetic structure of Fe/MgO(001) was examined by first-principles calculations [5]. By taking account of a perpendicular magnetocrystalline anisotropy energy and the interlayer exchange energy, it is found that the local magnetization direction $\theta(z)$ changes gradually from $\theta_{\rm int}$ = 59.3° to $\theta_{\rm mid}$ = 55° for the Fe (25 ML)/MgO(001) model (1 monolayer (ML) = 0.142 nm), as shown in **Fig. 3a**. Furthermore, we evaluated the stability of a noncollinear magnetic structure for the film with various thicknesses by taking account of the interface anisotropic energy, exchange energy and the shape anisotropic energy. As a result, it is found that the noncollinear magnetic structure with $\theta_{\rm int} \leq 40°$ and $\theta_{\rm mid}$ = 0° is energetically stable for the Fe film thickness of 26 nm as shown in **Fig. 3b**, which is roughly consistent with the present experiment result.

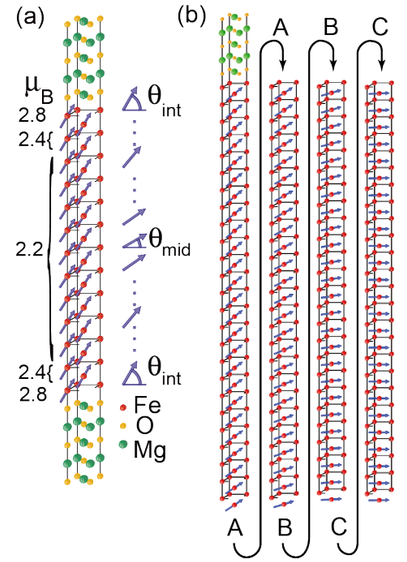

Figure 3 (a) Schematic figure of the slab model for Fe/MgO(001) used in the first-principles calculations. θ_{int} and θ_{mid} are the angles of the Fe spin moments at the interface and middle of the film. The values are the magnitude of the local spin moments in μ_B. **(b)** Schematic figure of MgO/Fe(001) used in the model calculations. The MgO/Fe(001) interface contains 5 ML of MgO and 200 ML of Fe, where the characters A, B, and C at the bottom of the Fe layers denote the layers that are connected to those indicated by the same characters at the top. The spatial change of the local magnetization direction $\theta(z)$ of Fe atoms is shown for W = 26 nm and θ_{int} = 40°.

REFERENCES

[1] D. L. Mills, *Phys. Rev. B* **39**, 12306 (1989).
[2] R. C. O'Handley and J. P. Woods, *Phys. Rev. B* **42**, 6568 (1990).
[3] U. Gradmann, *J. Magn. Magn. Mater.* **100**, 481 (1991).
[4] T. Kawauchi, K. Fukutani, M. Matsumoto, K. Oda, T. Okano, X. W. Zhang, S. Kishimoto and Y. Yoda, *Phys. Rev. B* **84**, 020415 (2011).
[5] T. Kawauchi, Y. Miura, X. Zhang and K. Fukutani, *Phys. Rev. B* **95**, 014432 (2017).
[6] G. V. Smirnov, *Hyp. Int.* **123**, 31 (1999).

BEAMLINE

AR-NE1A

T. Kawauchi[1], Y. Miura[2, 3], X. -W. Zhang[4] and K. Fukutani[1]
([1]The Univ. of Tokyo, [2]Kyoto Inst. of Tech., [3]Osaka Univ., [4]KEK-IMSS-PF)