Novel Properties of Magnetic Ultrathin Films Studied by *In Situ* Spectroscopic Methods

Department of Materials Molecular Science
Division of Electronic Structure

YOKOYAMA, Toshihiko	Professor
NAKAGAWA, Takeshi	Assistant Professor
TAKAGI, Yasumasa	Assistant Professor
EGUCHI, Keitaro	Graduate Student
NAKANO, Hirohito	Graduate Student*
FUNAKI, Yumiko	Secretary
TOYAMA, Yu	Secretary (Nanotechnology Platform project)
YASUI, Yuco	Secretary (Graduate school affairs)

Novel properties of magnetic metal ultrathin films have been attractive both from fundamental interest and from technological requirements. We are especially interested in drastic modification of metal thin films through the film–substrate interaction and/or a surface chemical treatment. The magnetic properties are characterized by means of several kinds of *in situ* spectroscopic methods like MOKE (Magneto-Optical Kerr Effect) using UV-visible lasers and XMCD (X-ray Magnetic Circular Dichroism) using synchrotron radiation soft X-rays, and UV magnetic circular dichroism photoelectron emission microscopy (UV MCD PEEM) using such ultrashort pulsed UV lasers.

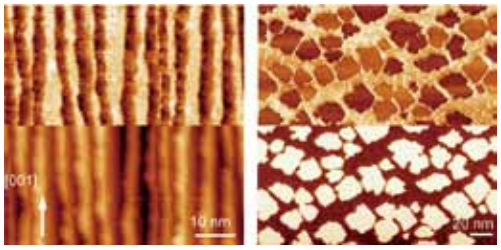

Figure 1. STM images of nanowires for Fe 0.3 ML (~6 atom width) (left) and nanodots for Fe 0.4 ML. The top and bottom panels correspond to the spin-polarized *dI*/*dV* and topographic images, respectively. The right-top panel clearly shows magnetically single domain dots (arrows denote the magnetization directions along [1-10]).

1. Giant Magnetic Anisotropy and Coercivity in Fe Island and Atomic Wire on W(110)[1)]

Hard magnets is an important industrial material both for data storage and power applications like electromotive actuators and power generators. Although the magnetic energy product (BH_{max}) that characterizes hard magnets has improved in the last century, rare earth (RE) magnets like $Nd_2Fe_{14}B$ have marked a saturation of the maximum available value of BH_{max} = 500 kJ/m^3 since 1990. Moreover, the replacement of the RE has been demanded due to the scarcity of RE, and novel approaches to improve hard magnets have attracted interests. Magnetic nanostructures with single atomic layers have a possibility to manifest prominent magnetic properties such as magnetocrystalline anisotropy energy (MAE) and coercivity (H_c) that remarkably differ from the properties of bulk materials. The Fe monolayer on W(110) has been extensively studied as an ideal two-dimensional ferromagnetic system with a pseudomorphic growth mode and an extremely large strain due to a large difference in the lattice constants between Fe and W. Owing to its extreme hard magnet character, the anisotropy energy or the coercivity has not been directly determined from the hard axis magnetization curve measurements. In this work, we directly measure the giant MAE as well as large coercivity in Fe nanodots and nanowires grown on W(110) by using our ultrahigh vacuum superconducting magnet XMCD system installed in UVSOR-II. A remarkable difference between the dots and the stripes is found in the coercivity, although the MAE is similar for both systems.

Figure 1 shows the spin-polarized and topographic STM (scanning tunnel microscopy) images of Fe nanowires and nanodots. The measurements were performed in Johannes Gutenberg Mainz University through the collaboration with Prof. H. J. Elmers group. In the spin-polarized STM image of the nanodots, a clear contrast (darker and brighter islands) can be seen, implying that the nanodots are magnetically ordered as single domains with the magnetization easy axis of the [1-10] crystal axis.

Figure 2 shows the anisotropic magnetization curves of Fe 0.25 ML nanodots and nanowires at 5 K taken by measuring the Fe L_3-edge circularly polarized X-ray absorption intensities using Beamline 4B at UVSOR-II. The G.I.[1-10] and G.I.[001] directions imply that the magnetization direction is tilted by 35° toward the surface normal with respect to the given crystal axis direction. The apparent coercivities are found to be larger than ~4 T for both the nanodots and nanowires. The anisotropic magnetic field, which is obtained by the extrapolation of the hard-axis magnetization curve (green lines in Figure 2) up to the saturated magnetization, can be esti-

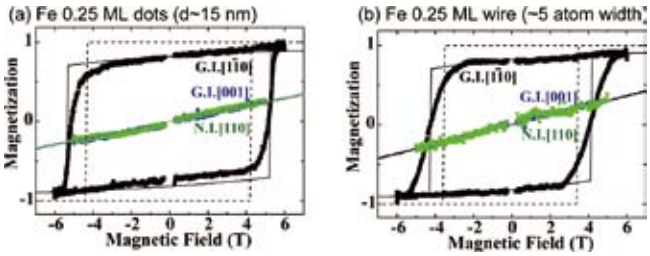

Figure 2. Anisotropic magnetization curves of 0.25 ML Fe nanodots (a) and nanowires (b) at 5 K. The data were recorded by measuring the Fe L_3-edge circularly polarized X-ray absorption intensities. Huge coercivities along the [1-10] easy axis (black lines) and anisotropic magnetic fields along the hard axes (green) were observed for both nanodots and nanowires.

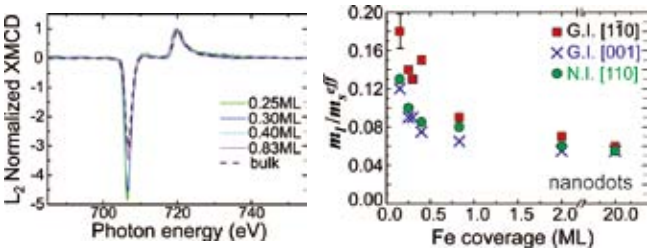

Figure 3. (left) Fe L-edge XMCD spectra of Fe nanodots on W(110) at 5 K along the G.I.[1-10] axis. (right) Ratio of the orbital magnetic moment (m_l) relative to the effective spin magnetic moment (m_s^{eff}). Smaller nanodots (lower Fe coverage) show larger orbital magnetic moments.

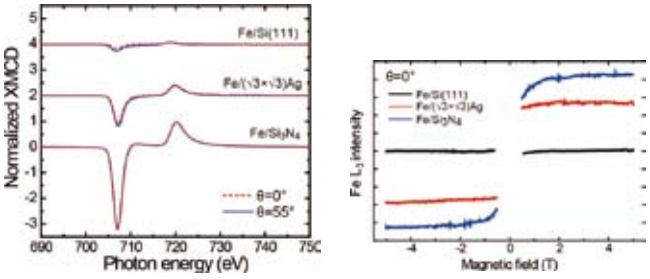

Figure 4. (left) Fe L-edge XMCD of Fe on clean Si(111) (1.6 ML), Ag/Si(111) (2.0 ML) and Si_3N_4 (1.6 ML) at H = ±5 T and T = 5 K. The X-ray incidence angles θ are 0° (normal incidence) and 55° (grazing). (right) Magnetization curves of Fe on clean Si(111) (1.6 ML, black), Ag/Si(111) (2.0 ML, red) and Si_3N_4 (1.6 ML, blue), taken at T = 5 K and θ = 0°.

mated to be >15 T. Huge magnetic anisotropy can be concluded both for the nanodots and nanowires. By fitting the experimental data with a simple anisotropic magnetic energy model, the magnetic anisotropic constants were determined to be ~1 meV for both the in-plane and out-of-plane directions, which is a few hundred times larger than that of bulk *bcc* Fe.

Figure 3 shows the Fe L-edge XMCD spectra of the nanodots at 5 K. The spectra were normalized with the L_2-edge peak tops. As the Fe coverage (nanodot size) decreases, the L_3-edge negative peak intensity increases drastically, implying a larger orbital magnetic moment as shown in the right panel of Figure 3. The orbital magnetic moment of the edge Fe atoms may be enhanced.

2. Magnetic Properties of Fe Nanostructures on Si_3N_4/Si(111)-(8×8) and Ag/Si(111)-(√3×√3)R30°[2)]

The magnetic properties of ferromagnetic transition metals on Si substrates have been widely investigated for the exploitation of new magnetic devices. Since clean Si surfaces react with transition metals very easily to form usually non-magnetic transition-metal silicides, it is essential to insert some inert film between transition metals and Si substrate. No reports have been however published for epitaxially ordered substrates on Si(111). In the present study, we investigated growth processes and magnetic properties of Fe deposited on well-defined Si_3N_4/Si(111)-(8×8) and Ag/Si(111)-(√3×√3) R30° by using STM and XMCD.

Figure 4 show the Fe L-edge XMCD spectra and the corresponding magnetization curves of Fe deposited on Si_3N_4/Si(111)-(8×8), Ag/Si(111)-(√3×√3)R30° and clean Si(111)-(7×7). It is clearly found that the Fe XMCD signals is enhanced in the sequence of Si(111) < Ag/Si(111) < Si_3N_4/Si(111). The XMCD sum-rule analysis yields the spin magnetic moments as m_s = 0.17 μ_B (clean), 1.25 μ_B (Ag), and 2.62 μ_B (Si_3N_4). On clean Si(111)-(7×7), the spin magnetic moment is almost quenched. On the contrary, Fe/Si_3N_4 has a much larger spin magnetic moment, which is even larger than that of *bcc* bulk Fe (2.2 μ_B). Fe/Ag/Si(111) shows a intermediate property. Such a drastic difference among the three substrates is caused by the fact that the Si_3N_4 substrate most effectively suppresses the silicide formation. Moreover, it is also found that the magnetic moment of Fe on Ag/Si(111) is lost quite easily by annealing at 500 K (m_s = 0.32 μ_B), while that of Fe on Si_3N_4 is sufficiently kept even when the sample is annealed at 500 K (m_s = 2.19 μ_B).

References

1) T. Nakagawa, Y. Takagi, T. Yokoyama, T. Methfessel, S. Diehl and H.-J. Elmers, *Phys. Rev. B* **86**, 144418 (2012).
2) K. Eguchi, Y. Takagi, T. Nakagawa and T. Yokoyama, *Phys. Rev. B* **85**, 174415 (2012).

* carrying out graduate research on Cooperative Education Program of IMS with Kyoto University