# Characterization of Magnetic Ultrathin Films by Novel Spectroscopic Methods

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Novel properties of magnetic metal ultrathin films have been attractive both from fundamental interest and from technological requirements. We are interested in drastic modification of metal thin films by surface chemical treatment such as adsorption-induced spin transitions and morphological changes. The magnetic properties are characterized by means of several kinds of spectroscopic methods like MOKE (Magneto-Optical Kerr Effect) using lasers and XMCD (X-ray Magnetic Circular Dichroism) using synchrotron radiation. Moreover, we are exploiting a new technique of ultraviolet (UV) magnetic circular dichroism (MCD) photoelectron emission microscopy (PEEM) in order to perform spatiotemporal magnetic imaging.

### 1. Enhanced Photoemission Magnetic Circular Dichroism Using Free Electron Laser at UVSOR-II

In 2006, we discovered surprising enhancement of the visible/ultraviolet photoemission MCD when the photon energy was tuned to the work function threshold.<sup>1)</sup> In the previous experiments, we employed Cs-coated films in order to reduce and control the work function of the magnetic thin film samples. It is essentially important to verify the enhancement of the photoemission MCD of the pure metal thin films without Cs deposition for the exclusion of some possible Cs-induced changes of the electronic structure. In this work, we have measured a magnetization curve of clean Ni films on Cu(001) by using a free electron laser (FEL) at UVSOR-II that is tunable and extremely intense.<sup>2)</sup>

Figure 1(a) shows the magnetization curve of 8 monolayer (ML) Ni on Cu(001) obtained by the total photoemission MCD measurement in the applied external magnetic field using the FEL,<sup>3)</sup> whose photon energy was tuned around the work function of Ni. The MCD asymmetry, defined as  $(I_{left}-I_{right})/(I_{left}+I_{right})$  where  $I_{left}$  and  $I_{right}$  are the photoemission intensities using left- and right-circularly polarized lights, respectively, is found to be ±5%. The value is around two orders of magnitude more intense than the conventional

MOKE measurement, as in our previous observations.<sup>1)</sup> Moreover, we have measured a photoemission MCD magnetization curve of Gd-coated Ni on Cu(001) by using a HeCd laser. The work function is again close to the photon energy. The result is shown in Figure 1(b). A similar enhancement of the photoemission MCD was observed.

We have consequently confirmed that the enhancement of photoemission MCD around the work function threshold is a universal behavior, applicable to photoelectron emission microscopy. This work was carried out in collaboration with the UVSOR machine group of Prof. M. Kato and Dr. M. Hosaka (present affiliation: Nagoya University).



**Figure 1.** Photoemission MCD magnetization curves of Ni on Cu(001). (a) Clean 8 ML Ni using FEL at UVSOR-II and (b) 2 ML Gd coated 10 ML Ni using a HeCd laser. Both the films show the work functions close to the photon energies.

### 2. Novel Magnetic Microscope: Ultraviolet Magnetic Circular Dichroism Photoelectron Emission Microscopy

The enhancement of the photoemission magnetic circular dichorism in the UV region paves a new way to develop UV MCD PEEM for the investigation of nanostructured magnetism. At present, MCD PEEM is performed as XMCD PEEM using third-generation synchrotron radiation X-ray sources. UV MCD PEEM allows us to do in-laboratory experiments when tunable deep UV lasers are available. Moreover, when ultrashort pulsed lasers are employed, pump-and-probe UV MCD PEEM measurements provide us a time resolving power of ~100 fs rather easily, which is two to three orders of magnitude faster than the present standard experiments using the third-generation synchrotron radiation sources. In the present experiment, we have constructed an UV laser PEEM apparatus and have successfully observed the first UV MCD PEEM images of the ultrathin film.<sup>2)</sup>

Figure 2 (left) is our UV MCD PEEM apparatus. In this experiment, as we had no deep UV lasers available, the Ni films was coated with Cs to match the work function to the photon energies of the employed UV lasers. A right panel of Figure 2 shows the magnetic image of the Cs-coated 12 ML Ni film on Cu(001) using a Ti:sapphire laser (second harmonics 400 nm). The image was given by the subtraction between the two images obtained by using the left- and right-circularly polarized lights. Beautiful magnetic domains can be seen; the light and dark areas are ascribed to the downward and upward magnetizations, respectively. This is the first observation of the UV MCD PEEM of the ultrathin film; although the UV MLD PEEM images (MLD: Magnetic Linear Dichroism) have been already reported, the results was given for a thick Fe film of 100 nm, and the contrast is only 0.19%.<sup>4)</sup>

We have succeeded in the observation of two-photon MCD PEEM of the same sample using a Ti:sapphire fundamental light (800 nm). Preliminary pump-and-probe data were also obtained with the time resolution of ~100 fs, which show the magnetization recovery with the time evolution. The experiments are in progress in collaboration with Prof. Y. Matsumoto (present affiliation: Kyoto University) and Dr. K. Watanabe.



**Figure 2.** (left) Photo of our UV MCD PEEM apparatus (Elmitec, PEEMSpector) and (right) UV MCD PEEM image of Cs-coated 12 ML Ni on Cu(001), taken using the second-order harmonics of a Ti: sapphire laser (400 nm). In inset, the magnetization curve of a similar film is also shown.

# 3. Magnetism of Self-Assembled Co Nanorods Grown on Cu(110)-(2x3)N

Magnetic properties of low dimensional magnets has recently attracted much interest due to their importance for further dense magnetic recording media. From the view point of fundamental physics, these materials are interesting for their magnetic anisotropy, Curie/blocking temperature. In this work, we have investigated structural and magnetic properties of self-assembled Co nanorods<sup>5</sup> with ~1 nm thick grown on Cu(110)-(2×3)N. This work was performed in collaboration with Prof. F. M. Leibsle (University of Missouri, Kansas, U. S.



**Figure 3.** (a) STM image (100 nm  $\times$  100 nm) of Co (1.5 ML equivalence) nanorods on Cu(110)-(2 $\times$ 3)N. (b) Angular dependent magnetization curves of 2.0 ML Co by longitudinal and polar MOKE. (c) Angular dependence of the Co *L*-edge XMCD spectra normalized to the *L*<sub>II</sub>-edge peak at ~793 eV. (d) Thickness dependence of the relative orbital magnetic moment at 90 K.

A.) and Prof. M. Przybylski (Max-Planck Institut, Halle, Germany).

Figure 3(a) shows the STM images. The Co rods grows with the  $(1\times 6)$  periodicity. The superstructure is consistent with the LEED pattern. From the Auger electron spectra (no figure) it is found that the N atom is located always at the top of the surface in spite of Co deposition on the  $Cu(110)-(2\times3)N$ surface. Magnetic properties have been characterized by MOKE and XMCD. Figure 3(b) shows the angular dependence of MOKE. A hysteresis loop was seen only along the [001] axis that is perpendicular to the rod axis within the surface plane. The magnetic easy axis is thus [001], which is interesting since macroscopic magnetic rods are likely to make their magnetization along the rod axis. In order to reveal the origin of the easy axis perpendicular to the rod axis, the Co Ledge XMCD spectra were recorded. Figure 3(c) shows the spectra. By comparing the relative intensity of the  $L_{\rm III}$ -edge peak (~778 eV), one can find the sequence of the orbital magnetic moments  $m_{\text{orb}}$  as  $[001] > [1\overline{1}0] > [110]$ . This implies that the spin-orbit interaction determines the magnetic easy axis. It is also found that below the Co thickness of 2 ML, the orbital magnetic moment is enhanced drastically. This supports the [001] magnetic easy axis in the present nanorods.

#### References

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#### Award

NAKAGAWA, Takeshi; SSSJ Young-Researcher Lecture Award, The Surface Science Society of Japan (2006).