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. In 2007, we have installed an *in situ* XMCD system with a superconducting magnet (up to 7 T) and a liq. He cryostat (down to 5 K). Some results obtained by the XMCD system are described below.

We are also exploiting new techniques of ultrafast time resolved ultraviolet (UV) magnetic circular dichroism (MCD) photoelectron emission microscopy (PEEM) in order to perform spatiotemporal magnetic imaging. Moreover, we are developing a two-photon photoemission MCD technique including PEEM.

1. First Observation of Ultrafast UV Magnetic Circular Dichroism Photoemission Electron Microscope Images

In 2006, we discovered surprising enhancement of the visible/ultraviolet photoemission MCD from ultrathin Ni films on Cu(001) when the photon energy was tuned to the work function threshold.¹⁾ Based on this discovery, we succeeded in the first observation of UV MCD PEEM images of ultrathin magnetic films.²⁾ This method allows us to do in-laboratory MCD PEEM measurements instead of the usage of synchrotron radiation XMCD PEEM. 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 that of XMCD PEEM.

Since 2007 we have been reconstructing the measurement

system by installing an ultrashort-pulsed deep UV laser and new UHV chambers to combine the MOKE, PEEM and photoemission experiments. Although the system is still under construction, we have tentatively observed time-resolved UV MCD PEEM images by using the previous setup. This work is a collaboration with Profs. Kazuya Watanabe and Yoshiyasu Matsumoto in Kyoto University.

Ultrafast UV MCD PEEM images were recorded by means of the pump-and-probe technique, in which the pump and probe lights were respectively the linearly-polarized fundamental (800 nm, ~70 fs) and the circularly-polarized secondorder (400 nm, ~200 fs) harmonics of a Ti:sapphire laser. The sample is Cs-coated Ni (12 monolayer) grown epitaxially on Cu(001).

Figure 1 shows the femtosecond time resolved UV MCD PEEM image and the time evolution of the local magnetization observed for the first time. Although the present result is rather tentative, we will measure more beautiful images in near future by using the new apparatus.

Moreover, we have succeeded in the first observation of the two-photon MCD PEEM. By employing a newly installed tunable deep UV laser, the maximum MCD asymmetry was found to be quite large. Especially, at 45° incidence the MCD is maximized, and this is quite useful for two-photon UV MCD PEEM.



Figure 1. Time resolved UV MCD PEEM image (left, $10\mu m$ area) and the time dependence of the local magnetization (right) of Cs-coated Ni(12ML)/Cu(001). Time resolution is ~200 f.

2. X-Ray Magnetic Circular Dichroism under High Magnetic Field and Extremely Low Temperature³⁾

XMCD is a powerful tool for the investigation of magnetism since it provides valuable information on element specific orbital and spin magnetic moments. A XMCD measurement system with a superconducting magnet and a liq. He cryostat is, however, not so popular in the world especially for public usage, although XMCD measurements under high magnetic fields are crucial for the investigation of magnetic anisotropy, because the saturated magnetization along the hard axis can be achieved around several tesla. We have thus constructed an in situ UHV soft X-ray XMCD system.

Figure 2 shows the schematic view and the photo of the present UHV XMCD system, which is usually installed at Beamline 4B of UVSOR-II. The magnetic field applied is \pm 7 T (typically \pm 5 T) and the sample temperature is ~5 K using liq. He. Samples are prepared in the preparation chamber and are transferred to the measurement chamber under UHV condition. The polar angle of the sample can be rotated by 360°, allowing us to examine angle dependence of XMCD.

In order to demonstrate the usefulness of the system, we have investigated angle dependent Co *L*-edge XMCD of Co(0.4 ML) on Cu(001). Since the sample shows strong inplane magnetic anisotropy, the saturation along the perpendicular magnetization direction (hard axis) requires a magnetic field of >3.4 T (5 T was applied). Through the analysis of the angle-dependent XMCD spectra, which can be done with saturated magnetization data even for the hard axis. As a result, the enhancements of the magnetic moments compared to the corresponding bulk values were clearly elucidated: ~15% for the spin magnetic moment, and ~96% and ~53% for the orbital magnetic moments along the surface parallel and normal directions, respectively. The surface magnetization is found to be essentially different from the bulk one.



Figure 2. Schematic view and photo of the XMCD system with a superconducting magnet and a liq. He cryostat.

3. Magnetism of Self-Assembled Co Nanorods Grown on Cu(110)-(2x3)N⁴⁾

Magnetic properties of low dimensional magnets has recently attracted much interest due to their importance for further dense magnetic recording media. In this work, we have investigated magnetic properties of self-assembled Co nanorods grown on Cu(110)- $(2\times3)N^{5}$) using the superconducting magnet XMCD system described above. This work was performed in collaboration with Prof. F. M. Leibsle (University of Missouri, Kansas) and Dr. X. -D. Ma and Prof. M. Przybylski (Max-Planck Institut, Halle).

Magnetic properties have been characterized by MOKE and XMCD. Figure 3(a) shows the angular dependence of Co L-edge in situ XMCD obtained by using the superconducting magnet at 4.9 K. Angle dependent magnetization curves of the Co nanorods recorded by MOKE (not shown) and XMCD [Figure 3(b)] show that the magnetic easy axis is perpendicular to the rod within the substrate plane, irrespective of the Co thickness down to 0.8 ML. From Figure 3(b), one can recognize that a high magnetic field is necessary to magnetize the sample along the magnetic field direction. The analysis of the magnetization curves clarify that the magnetic anisotropy is not dominated by the shape anisotropy but by the magnetocrystalline anisotropy. The XMCD analysis reveals significant enhancement of the orbital magnetic moment along the easy axis compared to the hard axes $([001] > [1\overline{10}] > [110])$. The magnetocrystalline anisotropy is found to be directly related to the anisotropy of the orbital magnetic moment.



Figure 3. (a) Angle dependent Co *L*-edge XMCD spectra of Co/ Cu(110)-(2×3)N (Co 0.8 ML) at T = 4.9 K and $H = \pm 5.0$ or ± 3.0 T. The lower and upper spectra correspond to those normalized with the edge jumps (×2 magnified) and the L_2 peak top intensity (+5 shifted), respectively. (b) Magnetization curves at T = 4.9 K recorded with the L_3 peak top. The simulated magnetization curves (light blue dotted lines) using a simple magnetic anisotropy model are also shown.

References

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