II-F Surface Magnetism of Ultrathin Films: Search of New Phenomena and Exploitation of New Techniques

Noble properties of magnetic thin films such as perpendicular magnetic anisotropy (PMA) and giant magneto-resistance (GMR) have extremely attracted scientific and technological interests. The origin of perpendicular magnetic anisotropy of ultrathin metal films is not fully understood and is an important subject in fundamental physics but is useful for high-density recording media. The GMR property is already utilized for read-heads of hard disk drives, although quantitative understanding of the GMR is still to be improved.

Our research subjects are twofold. The first one is to find out new important phenomena concerning surface magnetism. We have been investigating drastic changes of magnetic properties of ultrathin metal films by using surface chemical modification such as atoms/molecules adsorption on the surface. This is studied by several sophisticated techniques such as the synchrotron radiation x-ray magnetic circular dichroism (XMCD), the visible-light magneto-optical Kerr effect (MOKE) and the magnetization induced second harmonic generation (MSHG) techniques. A goal of these works is spin engineering by which the magnetization of ultrathin metal films and nanowires can be controlled artificially.

The second one is to exploit new techniques for the investigations of surface magnetism. Last year we discovered surprising enhancements of the magnetic circular dichroism (MCD) in the threshold photoemission, which provide the possibility of the visible and ultraviolet (UV) MCD photoemission electron microscopy (PEEM).

II-F-1 Effect of Adsorbate Carbon on Spin Reorientation Transitions in Cu-Capped Ultrathin Ni Films on Cu(001)

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We have reinvestigated the Cu capping effect for Ni films on clean Cu(001) by means of polar and longitudinal MOKE measurements in order to account for previous discrepancies[1,2] among the results regarding whether the Cu capping stabilizes perpendicular or in-plane magnetization. Figure 1(a) shows the Ni thickness dependence of the polar and longitudinal MOKE results of Ni/Cu(001) before and after Cu capping. The perpendicular magnetization from the polar MOKE appears at smaller thickness in Cu-capped Ni/Cu(001). We can immediately conclude that Cu capping stabilizes perpendicular magnetization.

We also found that the previous erroneous observation of in-plane stabilization could be accounted for by the presence of a small amount of C contamination, which was revealed to stabilize perpendicular magnetization surprisingly. Figure 1(b) shows the Ni thickness dependence of the polar MOKE results of Ni/C/Cu(001) before and after Cu capping. In contrast to the Ni films on clean Cu(001), the perpendicular magnetization is apparently unstabilized after Cu capping. The C atoms were found to act as surfactants and were always located at the top surface. As regards perpendicularly magnetized films on clean Cu(001), the enhancement of the coercivity with Cu capping was observed. This finding indicates that Cu does not act as a simple magnetism killer but effectively suppresses the surface anisotropy that favors in-plane magnetic anisotropy.

References

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2) H. W. Zhao, Y. Z. Wu, C. Won, F. Toyoma and Z. Q. Qiu, *Phys. Rev. B* **66**, 104402 (2002).

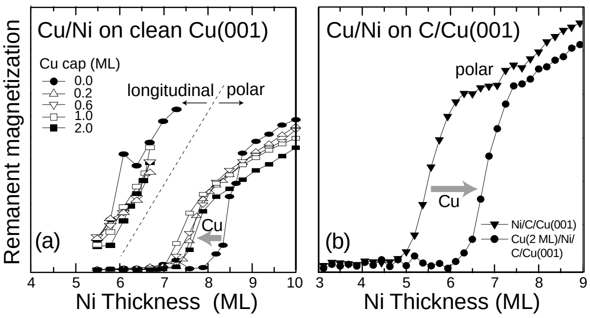

Figure 1. (a) Remanent longitudinal and polar MOKE intensities of Cu-capped Ni films grown on clean Cu(001) as a function of Ni film thickness. The critical thickness of the spin reorientation transition is shifted from ~8.5 ML to a thinner side (~7.5 ML) with increasing the amount of capped Cu. (b) Remanent polar MOKE intensities for Ni films on C-contaminated Cu(001) as a function of Ni film thickness. In contrast to (a), the critical thickness of the spin reorientation transition is shifted from ~5.5 ML to a thicker side (~7.0 M) with Cu capping.

II-F-2 Effect of Surface Chemisorption on the Spin Reorientation Transition in Magnetic Ultrathin Fe Film on Ag(001)

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We have investigated the effect of surface chemisorption on the spin reorientation transitions in magnetic ultrathin Fe films on Ag(001) by means of the polar and longitudinal MOKE and XMCD measurements. Remanent perpendicular magnetization and the coercive fields of the Fe films on Ag(001) before and after gas (O_2, NO and H_2) adsorption at 100 K are shown in Figure 1. It is found by the MOKE that adsorption of O_2 and NO induces the shift of the critical thickness for the transitions to a thinner side, together with the suppression of the remanent magnetization and the coercive field of the Fe film. This implies destabilization of the perpendicular magnetic anisotropy. On the other hand, H_2 adsorption is found not to change the magnetic anisotropy, though the enhancement of the coercive field is observed. The XMCD reveals that although both the spin and orbital magnetic moments along the surface normal are noticeably reduced upon O_2 and NO adsorption, the reduction of the orbital magnetic moments are more significant. This indicates that the destabilization of the perpendicular magnetic anisotropy upon chemisorption of O_2 and NO originates from the change of the spin-orbit interaction at the surface.

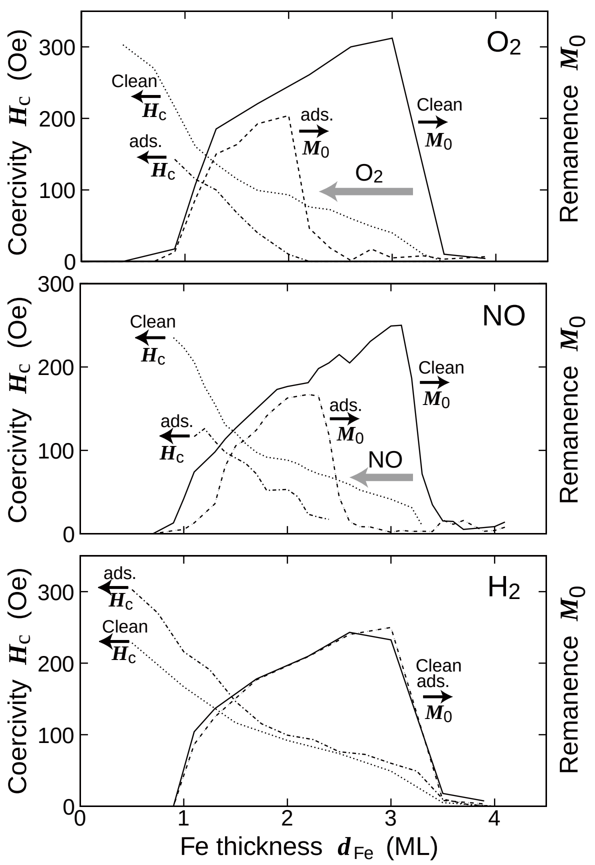

Figure 1. Remanent magnetization and the coercive field of the Fe films on Ag(001) before and after gas (O_2, NO and H_2) adsorption recorded at 100 K by the polar MOKE measurements. O_2 and NO adsorption destabilizes perpendicular magnetic anisotropy, while H_2 does not change the critical thickness.

II-F-3 Magnetic Circular Dichroism near the Fermi Level: Possibility of UV MCD PEEM

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It has long been believed that the MCD in the visible and ultraviolet regions is in general too weak to apply for a noble nano-scale magnetic imaging technique of PEEM. In this study, we discovered surprising enhancement of the MCD intensity near the Fermi level using visible and ultraviolet lasers. Figure 1(a) shows the MCD asymmetry of a perpendicularly magnetized Cs-coated 12 ML Ni film on Cu(001) as a function of $h\nu-\Phi$ ($h\nu$ the photon energy and Φ the work function). The work function was changed with the aid of Cs adsorption. More than 10% MCD asymmetry is achieved near the photoemission threshold. The MCD asymmetry is found to be enhanced only near the threshold and to drop down to 0.1% at the photon energy larger than the work function by 0.6 eV. A theoretical calculation also shows enhanced MCD near the photoemission threshold, qualitatively in agreement with the experimental results. Other ultrathin films of 6 ML Ni, 15 ML Co, and 3 and 15 ML Fe on Cu(001) were also investigated. It is found that the perpendicularly magnetized films show much larger MCD asymmetries than the in-plane magnetized films as in the Kerr effect.

Moreover, we have performed the measurements of magnetization curves on clean Ni/Cu(001) using the free electron laser (FEL) from UVSOR-II in order to eliminate the possibilty of the Cs effect. This part is a collaboration with a UVOSR machine group (Prof. M. Kato, Dr. M. Hosaka *et al.*). Figure 1(c) shows the hysteresis loop taken with the photon energy of 5.37 eV. Although the photon energy was not optimized, the MCD asymmetry is found to be as much as 5–6%. This supports the idea that the threshold photoemission provides huge enhanced MCD especially in the perpendicularly magnetized films.

This discovery enables us to exploit a new technique of UV MCD PEEM. At present, x-ray MCD PEEM is widely available for the investigations of nano-scale magnetic imaging of magnetic thin films. This technique however requires third generation synchrotron radiation light sources and cannot be used in laboratories. It is also difficult to obtain information on subpicosecond ultrafast spin dynamics due to the pulse width of the synchrotron radiation. If the UV MCD PEEM techniques can be used in near future, these difficulties can be overcome. We are now developing the new tchnique of UV MCD PEEM.

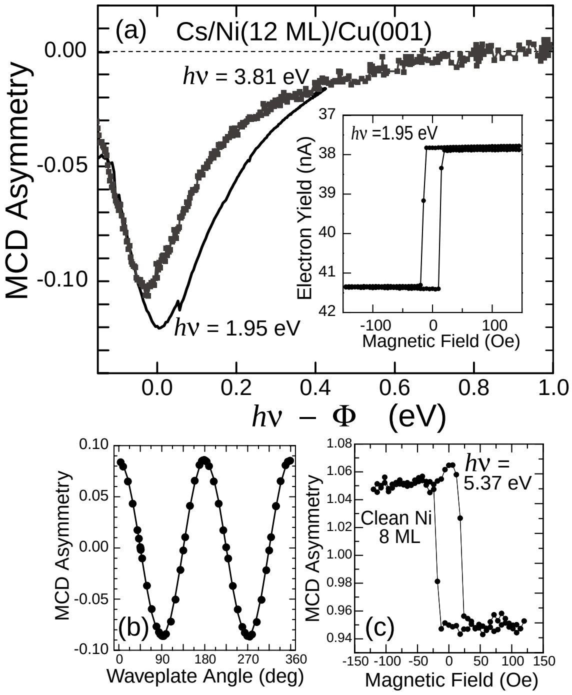

Figure 1. (a) MCD asymmetry from Cs/Ni(12ML)/Cu(001) at normal incidence as a function of $h\nu-\Phi$ ($h\nu$ = 1.95 or 3.81 eV). The work function varied by changing the Cs coverage. The largest MCD asymmetries are as much as 10% near the photoemission threshold. The inset shows the typical magnetization curve. (b) Azimuthal angle dependence of the quarter-wave plate. The angles 0°, 45° and 90° correspond to the left-circularly, linearly, right-circularly polarized lights, respectively. The cosine curve clearly shows the successful bservation of MCD. (c) The magnetization curve of clean Cs-free Ni/Cu(001) taken by using the FEL from UVSOR-II. Intense MCD was observed, eliminating the possibility of the Cs effect for the MCD enhancement.