

Resolving Subsurface Magnetism at Atomic Scale by "Diffraction Spectroscopy"

Future data storage densities will soon need to exceed one terabyte (10^{12} bytes) per square inch, requiring bits just 10 nm or less across. However, this is the scale at which surface magnetism appears. Thus, it is critical to understand unusual magnetic effects from a *nanoscopic* point of view. The magnetism of outermost atoms can be detected individually by magnetic STM, whereas much difficulty lies in the case of subsurface atoms. So far, several methods have been developed for probing the magnetic structure of buried interfaces and multilayers. Recently, Amemiya *et al.* have succeeded in evaluating the magnetic moments of surface and subsurface using the emission angle dependence of Auger electron probing depth [1]. However, no atomic-layer resolved characterization technique for subsurface magnetic structures has been developed until now.

A newly developed technique, *diffraction spectroscopy*, enables direct access to the subsurface region, which connects surface and bulk worlds [2]. We combined two existing techniques: X-ray absorption spectroscopy and Auger electron diffraction (AED) measurement. X-ray absorption near edge structure (XANES) and X-ray magnetic circular dichroism (XMCD) measurements by Auger electron yield detection are powerful analysis tools for the electronic and magnetic structures of surfaces. However, all the information from atoms within the electron mean-free-path range is averaged into the obtained spectra. Forward-focusing peaks (FFPs) in AED patterns indicate the directions of atoms surrounding the excited atom. Taking advantage of the FFP as an excellent element- and site-selective probe, we disentangled spectra from different atomic layers.

The spin reorientation transitions (SRTs) of a magnetic epitaxial film from in-plane to perpendicular direction are intriguing phenomena. A Ni ultrathin film on a Cu(001) surface exhibits both SRT and reversal SRT, which are regarded as a basic starting point of nanoscale magnetism. We used *diffraction spectroscopy* to visualize both the magnetic and electronic properties of subsurface layers on the atomic scale in a nondestructive way.

We used the display-type

analyzer (DIANA) [3,4] installed at the circularly polarized soft X-ray beamline **BL25SU** for measuring AEDs. As illustrated in Fig. 1(a), electrons emitted from the sample were energy-analyzed and their angular distributions were projected onto a fluorescence screen. The Ni LMM AED pattern excited with normal incident soft X-rays (852.8 eV) are shown in Fig. 1(b). The fcc cluster indicates the relation of crystal orientation and FFPs. FFPs clearly appear along the directions of atoms surrounding excited atoms in the AED pattern. The intensity at an extremely low emission angle especially in the [100] direction is mainly due to the emission from the outermost surface atoms. The FFPs in the [101] and [001] directions indicate the existence of Ni atoms at the second and third layers, respectively.

We obtained a XANES spectrum by monitoring Auger electron intensity while scanning excitation photon energy. The intensity of different FFPs corresponds to the signal of different atomic layers. Figure 2(a) shows the atomic-layer resolved Ni L_3 -XANES spectra of a 15 ML Ni film. The first- to fourth-layer spectra were extracted from [100], [101], [001] and [103] FFP intensities, respectively. Note that the intensity of the so-called 6-eV satellite is diminished in the surface layer spectrum. This is due to the atomic and electronic structures peculiar to the outmost surface layer where atoms bond to a relatively few neighboring atoms. The L_3 peaks in the spectra of the surface layer shift to higher photon energy compared to those in the spectra of interior layers by about 0.1 eV.

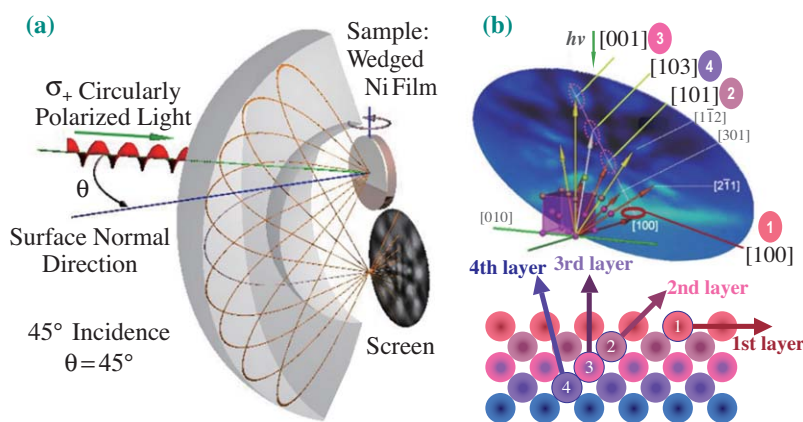


Fig. 1. (a) Schematic diagram of display-type analyzer. An Auger electron diffraction (AED) snapshot is taken in a short time, typically 1 sec. (b) AED pattern of Ni thin film (kinetic energy: 841.0 eV) together with fcc cluster. Electron-emitting atoms are specified for forward focusing peaks.

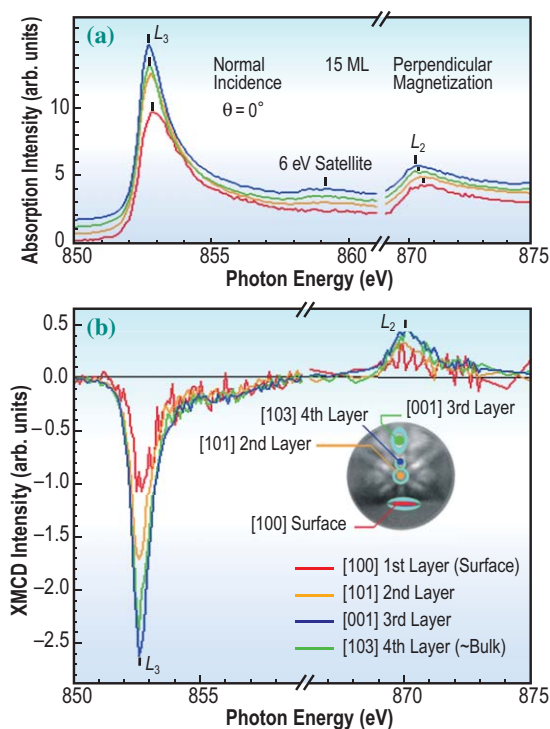


Fig. 2. (a) Atomic-layer-resolved Ni L-XANES and (b) X-ray magnetic circular dichroism (XMCD) spectra of perpendicularly (15 ML) magnetized Ni films.

The L_3 shifts correspond to the surface core level shifts. The XMCD spectrum is the difference between the Ni L-XANES spectra excited with positive (σ_+)- and negative (σ_-)-helicity light. Figure 2(b) shows the atomic-layer-resolved XMCD spectra for the 15 ML perpendicularly magnetized films. We analyzed Ni magnetic structures of various film thicknesses on the atomic scale and deduced the spin ($\mu_{\text{spin}}^{\text{eff}}$) and orbital (μ_{orb}) magnetic moments for each atomic layer.

The orbital magnetic moments are more enhanced at the surface layers compared with the spin magnetic moments in the case of 8 ML (in-plane); the variations in the moments are rather small in the case of 15 ML (perpendicular). The orbital is sensitive to the local bonding configuration, while the spin reflects the long-range magnetic ordering. At a thickness beyond SRT, the magnetic moment orientation in a perpendicular direction is caused by the spin magnetic moment in the interior layers, whereas in the region below SRT, where the spin magnetic moment is small, the orbital magnetic moment of the surface layers determines the orientation to be in-plane. SRT at 10 ML is based on such a subtle balance of magnetic moments. Knowing exactly how these magnetic moments change throughout the structure will be useful for making perpendicular magnetic recording devices. Perpendicular magnetic domains are stably packed closer together for greater density than in-plane

magnetic domains, which leads to more data per square inch.

In conclusion, we have developed a direct method for atomic-layer resolved analysis of electronic and magnetic structures for surfaces and thin films. This technique opens a door for the study of the correlations of the atomic, electronic, and magnetic structures of nanostructures at the surfaces.

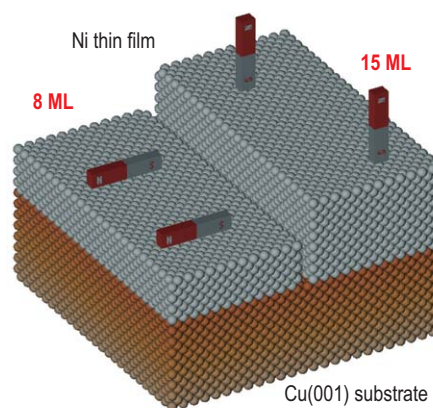
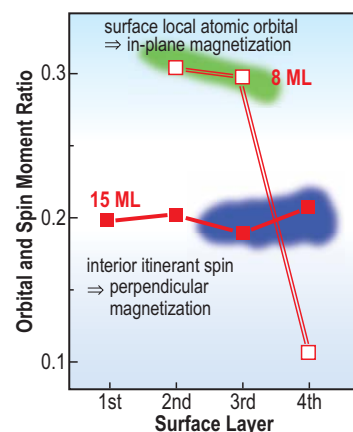


Fig. 3. Ratio of orbital to spin components ($\mu_{\text{orb}}/\mu_{\text{spin}}^{\text{eff}}$) of each atomic layer in the Ni thin film on a Cu(001) surface.

Fumihiko Matsui^{a,*}, Tomohiro Matsushita^b and Hiroshi Daimon^a

^a Graduate School of Materials Science, Nara Institute of Science and Technology
^b SPring-8/JASRI

* E-mail: matui@ms.naist.jp

References

- [1] K. Amemiya *et al.*: Phys. Rev. B **71** (2005) 214420; *ibid.* **72** (2005) 201404(R).
- [2] F. Matsui, T. Matsushita, Y. Kato, F.Z. Guo, M. Hashimoto, K. Inaji and H. Daimon: Phys. Rev. Lett. **100** (2008) 207201.
- [3] H. Daimon: Phys. Rev. Lett. **86** (2001) 2034.
- [4] T. Matsushita *et al.*: Phys. Rev. B **75** (2007) 085419.