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 especially 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 UV-visible lasers and XMCD (X-ray Magnetic Circular Dichroism) using synchrotron radiation soft X-rays.

Moreover, we have been exploiting new techniques based on UV photoemission magnetic circular dichroism (MCD) such as ultrafast time resolved UV MCD photoelectron emission microscopy (PEEM) for spatiotemporal magnetic imaging.

1. Two Opposite Spin Reorientation Transitions in Fe-Covered Ni/Pd(111)[1)]

Magnetic anisotropy has widely been investigated in last decades, and especially perpendicular magnetic anisotropy has attracted much interest because of practical applications to high density recording media. We discovered two opposite spin reorientation transitions (SRTs) in Fe-covered Ni/Pd(111) magnetic films and investigated the origins of the two SRTs.

Pure Ni/Pd(111) films always show in-plane magnetization, irrespective of Ni thickness. Deposition of Fe 1 monolayer (ML) on Ni(6 ML)/Pd(111) causes a transition to perpendicular magnetization, and further Fe 3 ML deposition leads to a return to in-plane magnetization. The first SRT from in-plane to perpendicular magnetization is attributed to a large perpendicular orbital magnetic moment of a single Fe layer, which gives a significant impact on the Fe–Ni interface that stabilizes perpendicular magnetic anisotropy. The second SRT coincides with the structural change in the Fe film from the *fcc* to *bcc* phase, where the reduction in the orbital magnetic moment along the perpendicular direction suppresses the perpendicular magnetic anisotropy stability. It can be proposed that the origin of the second SRT to in-plane magnetization is attributed to cooperative contributions of the structural transformation of the Fe film and the enhanced demagnetizing field.

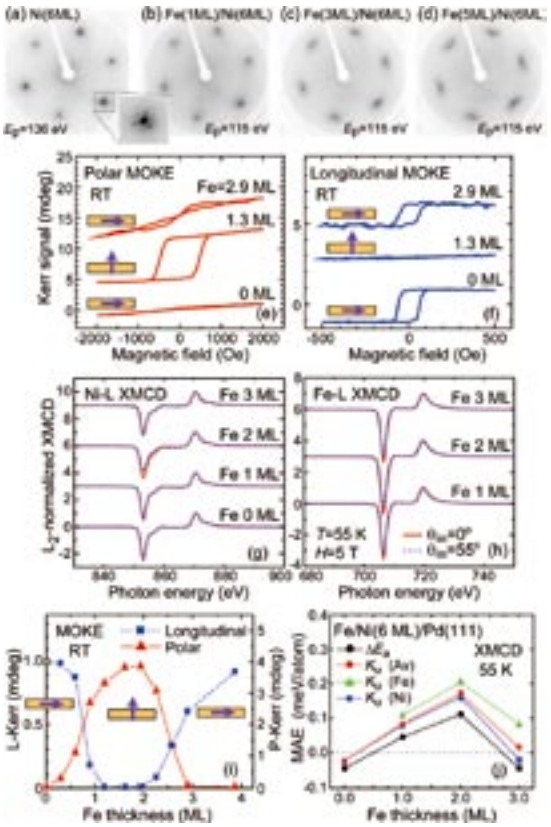

Figure 1. (a-d) Variation of LEED patterns of Ni(6ML)/Pd(111) during Fe deposition up to 5 ML. The structure of Fe changes to *bcc* around 3 ML. (e,f) Typical perpendicular (e) and in-plane (f) magnetization curves of Fe/Ni(6ML)/Pd(111) recorded by polar and longitudinal MOKE, respectively. (g,h) Ni (g) and Fe (h) L-edge XMCD spectra of Fe/Ni(6ML)/Pd(111). (i) Polar and longitudinal MOKE intensities of Fe/Ni(6ML)/Pd(111) as a function of Fe thickness. Two opposite SRTs occur at Fe 1 and 3 ML. (j) Magnetic anisotropy energies estimated by XMCD. The perpendicular magnetic anisotropy is maximized at Fe 2 ML.

Figures 1(a-d) show low energy electron diffraction (LEED) patterns of Ni(6ML)/Pd(111) during Fe deposition. In Ni(6ML)/Pd(111), Moiré spots are clearly visible, implying that Ni is found to grow epitaxially but not pseudo- morphically on Pd(111) with maintaining the inherent Ni–Ni distance. For more than 3 ML Fe deposition, the fundamental spots are elongated, indicating the formation of *bcc* Fe. The structural transformation is concluded to take place around 3 ML Fe.

Figure 1(i) plots the polar and longitudinal MOKE (magneto-optical Kerr effect) intensities at room temperature. It is clearly found that two opposite SRTs occur at Fe 1 and 3 ML, between which the perpendicular magnetization is stabilized. Figures 1(g) and 1(h) exhibit Ni and Fe L-edge XMCD (X-ray magnetic circular dichroism) spectra, which provide spin and orbital magnetic moments and also the microscopic magnetic anisotropy constants as Figure 1(j). The perpendicular magnetic anisotropy is maximized at Fe 2 ML and is subsequently reduced at higher Fe coverage. The second SRT around 3 ML Fe can thus be ascribed to the structural transition to the *bcc* Fe phase, cooperated with the increase in the demagnetization field (shape anisotropy).

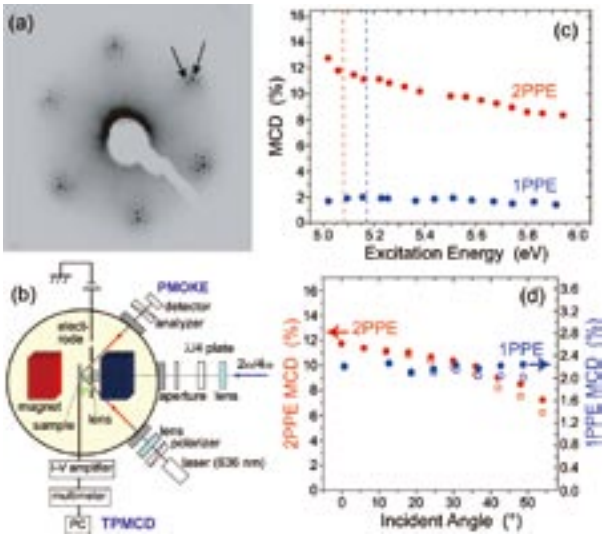

Figure 2. (a) LEED patterns of Co(4.5ML)/Pt(111). (b) 1PPE and 2PPE MCD measurement setup. (c,d) 1PPE and 2PPE MCD asymmetries of Co(4.5ML)/Pt(111) as functions of (c) excitation energy ($h\nu$ and $2h\nu$ for 1PPE and 2$h\nu$, respectively) and (d) the laser incidence angle. The blue and red vertical lines are the work function thresholds, which are slightly different from each other, depending on the sample preparation.

2. Observation of One- and Two Photon Photoemission Magnetic Circular Dichroism of Perpendicularly Magnetized Co/Pt(111)

In 2006, we discovered surprising enhancement of the UV-visible photoemission magnetic circular dichroism (MCD) from ultrathin Ni films on Cu(001) when the photon energy was tuned to the work function threshold.[2] Based on this discovery, we succeeded in the first observation of UV MCD PEEM images of ultrathin magnetic films.[3] This method allows us to perform in-laboratory MCD PEEM measurements instead of the usage of third-generation synchrotron radiation XMCD PEEM. Moreover, ultrafast UV MCD PEEM images were successfully obtained by using ultrashort pulsed lasers and a pump-and-probe technique.[4] In 2009, we observed two-photon photoemission (2PPE) MCD and also 2PPE MCD PEEM images from Ni/Cu(001).[5] The 2PPE technique is essentially important for the excitation of deeper valence levels such as semiconductors. In this work, we have exemplified noticeable superiority in 2PPE MCD compared to 1PPE MCD in another example of perpendicularly magnetized Co/Pt(111). This work has been performed under IMS international collaboration program with K. Hild, G. Schönhense and H. J. Elmers (Mainz Johannes Gutenberg-Universität, Germany), and with K. Tarafder and P. M. Oppeneer (Uppsala University, Sweden).

Figure 2(a) shows the LEED pattern of 4.5 ML Co grown on Pt(111). Additional spots can clearly be seen around the fundamental Pt(111) (1×1) spots, which are ascribed to the Moiré patterns from the Co lattice. The Co film is thus found to grow epitaxially but not pseudomorphically on Pt(111). Figure 1(b) depicts the experimental setup. We employed a mode-locked tunable Ti:Sapphire laser (80 MHz, 2.5 W, 680–1020 nm, 100 fs) with the third and fourth-order harmonic generators. The photoemission from the sample was collected by the anode and the drain sample current was recorded. The sample Co(4.5ML)/Pd(111) shows perpendicular magnetic anisotropy with a coercive field of ~580 Oe.

Figure 1(c) shows the 1PPE and 2PPE MCD asymmetries as a function of the excitation energy. At the work function threshold we obtain maximum values of 1.90% for 1PPE and 11.7% in the case of 2PPE, the latter being as much as 6.2 times larger, elucidating the efficiency of the 2PPE MCD. Figure 1(d) exhibits laser incidence angle dependence of the 1PPE and 2PPE MCD asymmetries. The 2PPE MCD asymmetry is found to be gradually reduced with the increase in the incidence angle, while for 1PPE there is almost no angle dependence observable. These behaviors are significantly different from those of Ni/Cu(001).[5] The measured MCD asymmetries are discussed in two excitation models as well as on the basis of spin-polarized energy-band calculations. The ab initio calculated and measured 1PPE MCD responses are in good agreement. An explanation of the large 2PPE MCD signal is provided in terms of specific inter-band excitations.

References

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