



Magnetic circular dichroism study of ultrathin Ni films by threshold photoemission and angle resolved photoemission spectroscopy

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ABSTRACT

Threshold photoemission magnetic circular dichroism (MCD) for 12 monolayer (ML) Ni thin films grown on Cu(001) was measured with a total electron yield method. The obtained MCD asymmetry using the total electron yield method reaches as much as 10% near the photoemission threshold. On the other hand, the MCD asymmetry in angle resolved photoemission spectra with the photon energy of 5.28 eV was also investigated for Cs/Ni(12 ML)/Cu(001) and was found to give a similar MCD asymmetry. The high MCD asymmetry in the total electron yield can be interpreted by the argument that the angle and energy selection is spontaneously achieved in the total electron yield near the photoemission threshold for low photon energy excitation.

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1. Introduction

Magnetic circular dichroism (MCD) is derived from both spin–orbit coupling and exchange interaction in magnetic materials. MCD is now an indispensable technique for the investigation of thin film magnetism. The MCD effect in X-ray region is widely recognized as XMCD, which has a strong merit to measure element specific magnetization with high sensitivity [1] due to the strong spin–orbit coupling in core levels. MCD effects in valence states are usually measured in transmission or reflection. Among them, magneto-optical Kerr effect (MOKE) is of importance for the investigation of magnetism of thin films in laboratories, because feasibly available light sources, like diode lasers or discharge lamps, make the MOKE measurement possible and its experimental setup is quite simple [2]. However, the spatial resolution of MOKE is limited by the diffraction limit of light. With the photoelectron emission, the valence band MCD combined with electron microscope may give better spatial resolution. However, its sensitivity to the magnetization is usually so low due to the weak spin–orbit coupling in the valence band that it is difficult to investigate the microscopic magnetic structure of thin films. Although it is known that angle and energy resolved photoemission substantially improves the sensitivity, this technique is not suitable for photoemission electron microscope (PEEM). High MCD asymmetry in

the total electron yield is thus desirable. One can find only very few examples for the PEEM measurements of magnetic domains using ultraviolet magnetic dichroism [3]. Recently, it has been reported that the valence band MCD integrated in angle and energy gives high magnetic asymmetry near the photoelectric threshold [4–7].

The total electron yield measurement for the valence band gives rough information about the electronic structure since it measures the energy and angle integrated photoelectrons. Valence band studies with angle resolved photoemission spectroscopy (ARPES) are demanded to explain the detailed origin of the MCD asymmetry observed in the total electron yield. MCD in ARPES gives fruitful information about the effect of the spin–orbit coupling in electronic bands. It has been shown that angle- and energy-resolved photoemission often provides strong MCD and/or magnetic linear dichroism (MLD) asymmetry [8–10]. The high asymmetry comes from a state selective measurement of the band structure by ARPES. However, to our knowledge, the ARPES experiment with low photon energies less than 10 eV, close to the energy for the photoemission threshold for transition metals, is not available.

In this article, we present MCD measurements on Ni(12 ML)/Cu(001) both by the total electron yield and ARPES using low energy photons ($h\nu < 6$ eV). The total electron yield can produce large MCD asymmetry comparable to that obtained by ARPES in spite that the total electron yield does not have to require an energy or angle analyzer. We address that the threshold photoemission and the photoelectron diffraction effect at the crystal surface for the excitation with low energy photons are substantially important for such large MCD asymmetry.

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2. Experimental

Experiments were done in separate chambers, one for the total electron yield measurement in magnetic field, the other for ARPES. The threshold photoemission (TP) MCD experiment was done in UHV with an UHV-compatible electromagnet (max. 3000 Oe) [5,11]. The base pressure was less than 2×10^{-10} Torr during the experiment. The sample drain current was measured as the total photoelectric yield. To avoid distortion of the electric current measurements due to an external magnetic field, an anode with an applied voltage of +1 kV was placed in front of the sample. The MCD asymmetry, A , is defined as $A = (I^{++} - I^{+-}) / (I^{++} + I^{+-})$, where I^{++} (I^{+-}) is the sample current for the parallel (antiparallel) orientation between photon spin and majority electron spin.

ARPES was performed in a different chamber with a hemispherical electron analyzer (SPECS, Phoibos 100). The overall energy resolution together with the photon resolution was ~ 40 meV. The light incidence angle was 0° , and the photoelectron emission angle was 0° with the angle resolution of $\pm 1^\circ$. A pulse coil was placed ~ 5 mm behind the sample and a magnetic field normal to the sample surface was generated. To change the sample work function, Cs was deposited from commercial dispensers at a rate of 0.01 monolayer (ML)/min. When ~ 0.15 ML Cs adsorbs on the sample, the work function decreases to ~ 1.5 eV.

A Cu(001) crystal was used as a substrate in all the measurements shown in this article, which was cleaned by repeated cycles of Ar^+ sputtering and annealing at 825 K. Ni was evaporated from a nickel rod by electron bombardment with the sample temperature kept at 300 K, and the Ni thickness was calibrated by the observation of reflective high energy electron diffraction (RHEED) oscillation during the growth. With an ultrashort pulse and broad band laser (< 100 fs, 80 MHz, 1.2–1.8 eV), photoelectrons were excited by its fourth-order harmonics (4.9–6.0 eV) generated by BBO crystals. The incident light was circularly polarized by a quarter-wave plate together with a polarizer.

3. Results and discussion

Fig. 1 shows photon energy dependence of MCD asymmetry for Ni(12 ML)/Cu(001) surface by the total electron yield. Ni films grown on Cu(001) show perpendicular magnetization easy axis from 10 ML up to ~ 50 ML [12]. The present 12 ML Ni film on Cu(001) was perpendicularly magnetized, which was found to

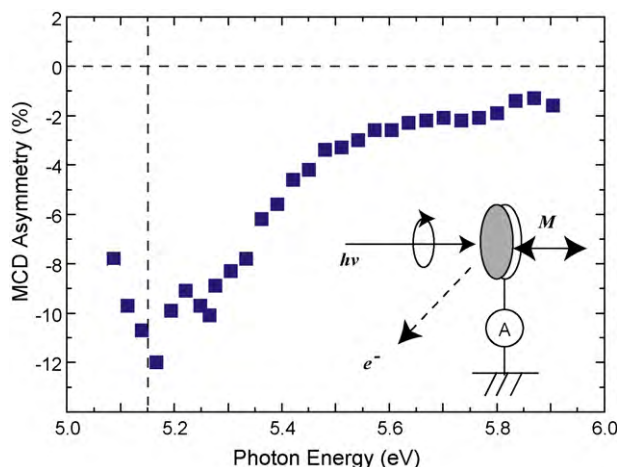


Fig. 1. MCD asymmetry from a perpendicularly magnetized Ni(12 ML)/Cu(001) as a function of the photon energy. The MCD asymmetry is measured by the total electron yield as shown in the inset. The light incidence angle is 0° . The work function estimated from the sample current against the photon energy is 5.15 eV as indicated by the vertical dashed line.

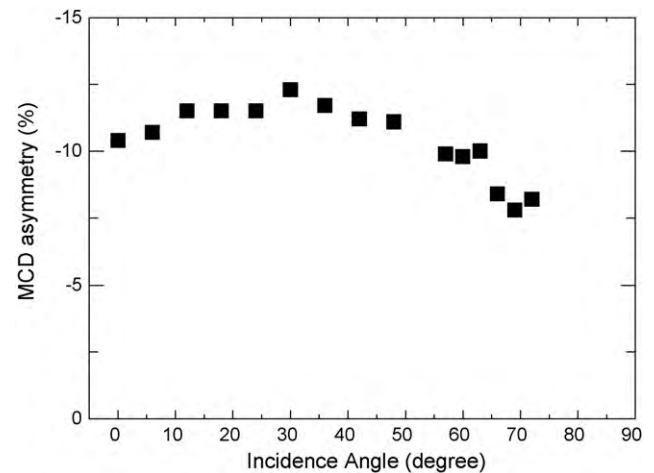


Fig. 2. Light incidence angle dependence of MCD asymmetry from a perpendicularly magnetized Ni(12 ML)/Cu(001). The zero degree corresponds to the normal incidence. The photon energy used is 5.3 eV, while the sample work function is 5.15 eV.

exhibit a rectangular magnetization curve with almost 100% remanence. The work function of the sample was estimated to be 5.15 eV from the photon energy dependence of the total electron yield. The light incidence angle was 0° and the MCD asymmetry was measured in a magnetic field of 200 Oe. As in Fig. 1, Ni(12 ML)/Cu(001) shows $\sim -10\%$ MCD asymmetry near the photoemission threshold. Away from the threshold, the absolute value of the asymmetry decreases with increasing the photon energy. At the photon energy of 5.9 eV, 0.75 eV higher than the photoelectric threshold, the MCD asymmetry is $\sim -1\%$. This energy dependent feature is also obtained for the experiment with varying the work function by Cs deposition at a fixed photon energy [4]. When the photon energy increases, the total electron yield measures the integrated photoelectron intensity over a wider energy range towards a higher binding energy, which could cancel the MCD effect due to the integration of different spin bands and opposite spin–orbit splitting bands [4,9].

Fig. 2 shows light incidence angle dependence of MCD asymmetry on the same sample as in Fig. 1. The photon energy used was 5.3 eV. The normal incidence is defined as 0° . The MCD asymmetry is nearly constant from 0° to 45° and then it gradually decreases for larger incidence angles. Since the MCD asymmetry is measured by the low photon energy, light reflection at the crystal surface plays an important role. In the case of X-ray MCD, the asymmetry varies as $\cos \theta$, where θ is the angle between the magnetization and the direction of the incident beam. This simple relationship is due to the fact that the X-ray is not reflected except grazing angles close to 90° . On the other hand, the ultraviolet light is reflected at the surface and the reflected light, which has the inverted sign of the angular momentum, also contributes to the MCD effect. The angular dependence of MCD asymmetry does not follow the cosine law, showing complicated behavior like the magneto-optical Kerr effect [13].

Fig. 3 shows a set of angle resolved photoemission spectra for Ni(12 ML)/Cu(001) with opposite magnetization directions, while the helicity of the circular polarization is fixed. These spectra were measured in the normal emission and in remanence magnetization states after pulse magnetization at $H \sim 200$ Oe normal to the surface. This measures the electronic structure along the $\Gamma - X$ direction in fcc structure if we neglect the distortion of the Ni lattice on Cu(001). The photon energy used was 5.28 eV and the light incidence angle was 0° . These experimental configurations for APRES are similar to those for the total electron yield measurement. In order to lower the sample work function, we deposited 0.15 ML Cs onto the sample, which resulted in the work function of 1.5 eV. This

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