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LETTER TO THE EDITOR

A direct test of x-ray magnetic circular dichroism sum rules for strained Ni films using polarized neutron reflection

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Abstract. We have directly compared the values of the magnetic moment obtained from x-ray magnetic circular dichroism (XMCD) measurements with those obtained from polarized neutron reflection (PNR) measurements on strained Ni films grown on Cu(001)/Si(001). The PNR measurements show that the absolute magnetic moments differ from that of bulk Ni. We find agreement within experimental errors between the two magnetometry measurements, confirming that the XMCD sum rules are applicable to this strained low-symmetric system.

Despite the fact that the pioneering theoretical study of Erskine and Stern [1] suggested that it is possible to obtain the spin polarization of the unoccupied d electronic states by using circularly polarized light, it is only recently with the advent of powerful synchrotron light sources that extensive experimental and theoretical studies using x-ray magnetic circular dichroism (XMCD) have begun. A key advantage of this technique is its high element selectivity [2–4]. Further interest in XMCD has been added by the development of magneto-optical sum rules [5, 6] derived for XMCD, which connect XMCD measurements to the ground-state values of the orbital (m_{orb}) and spin (m_{spin}) moments. Because of their important implications for magnetic materials and the fact that they were derived from a single-ion model which neglects several aspects of the band structure arising in real systems, there have been many attempts to verify the validity of XMCD sum rules in determining element-specific orbital and spin moments [7–12]. Chen *et al* [7] confirmed the applicability of the sum rules to bulk-like Co and Fe films. But the applicability of the x-ray sum rules for low-symmetry systems remains unclear. Several difficulties may be expected in such an attempt, especially given that the magnetic moment in low-symmetry systems is likely to be different from that of the bulk.

In order to overcome the expected difficulties in testing their validity, comparing the result of XMCD sum rules with the results of another magnetometric measurement on the same sample provides the simplest approach. In this letter we report the measurements of the total magnetic moment on the same strained Ni/Cu buffer/Si(001) samples with two different techniques, XMCD and polarized neutron reflection (PNR). Ni thin films grown on Cu buffer/Si(001) structures were chosen for the comparison for two reasons. First, a

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decreased moment has been reported in thin Ni films incorporated into Cu/Ni/Cu/Si(001) structures [13]. Our detailed study of the relation between the moment and structural parameters will be presented elsewhere [14]. Second, because only a single magnetic layer with a relatively large (51 Å) Ni thickness is involved, accurate measurements of the atomic moments and thickness can be made from the PNR measurements. Therefore this system offers an ideal opportunity for testing the validity of the XMCD sum rules in determining the magnetic moments in non-bulk-like magnetic systems.

Si(001) substrates were degreased and etched in diluted HF solution for 12 minutes prior to loading into the growth chamber. The base pressure was in the 1×10^{-9} mbar range and during deposition increased to 5×10^{-9} mbar. Cu buffer layers were grown at 10 Å min^{-1} using an electron-beam-heated Mo crucible and the epitaxial Ni films at 1.5–2 Å min^{-1} by electron beam evaporation. A two-step Cu buffer layer was prepared with different thicknesses upon which a 51 ± 4 Å Ni film was deposited (sample A: 51 Å Ni/490 ± 4 Å Cu and sample B: 51 Å Ni/771 ± 7 Å Cu). All thicknesses quoted are based on fits to the PNR data. The cleanliness of the films was checked by Auger electron spectroscopy (AES). Reflection high-energy electron diffraction (RHEED) indicated that sample B shows sharper RHEED images than that of sample A. A Cu capping layer was further deposited for *ex situ* measurements.

It is well known that strained Ni/Cu(001) shows perpendicular magnetic anisotropy at this thickness due to a magnetoelastic contribution [15]. Our polar magneto-optical Kerr effect (MOKE) measurements confirmed the full perpendicular remanence of our samples.

In order to measure the absolute atomic magnetic moment, PNR experiments were carried out on these samples. PNR has recently been developed as a direct method of determining atomic moments with high accuracy [16, 17]. The main advantage of PNR over conventional magnetometry techniques is that it is a self-calibrating magnetometric technique since the difference between the spin-up and spin-down reflectivities yields the total magnetic moment of the layer while the period and structure of the reflectivity oscillations independently give information on the thickness in the nm range and structure of the constituent layers.

Table 1. The structural and magnetic parameters determined by PNR and XMCD sum rules. Each film thickness has been confirmed separately by x-ray measurements. In particular the Ni thickness has been confirmed to be 50 ± 2 Å by x-ray measurements.

	Sample A	Sample B	
Si/Cu interface/Å	50	50	PNR
Cu thickness/Å	497 ± 4	771 ± 7	
Ni thickness/Å	51 ± 4	51 ± 4	
Cu capping/Å	18	18	
Cu/air interface/Å	20	20	
Magnetic moment/μ_B	0.50 ± 0.02	0.53 ± 0.02	
Magnetic moment/μ_B	0.55 ± 0.03†	0.58 ± 0.03†	Sum rules

† The errors in magnetic moments determined by applying XMCD sum rules come from the repetitive measurements.

The PNR measurements were carried out on the G2-2 reflectometer ($\lambda = 4.14$ Å) at the Orphée reactor, Saclay (France), using sample rotation (0.001° resolution) [18] to vary the perpendicular wavevector. Polarized neutrons with spins parallel (spin-up) and antiparallel (spin-down) to the in-plane sample magnetization (perpendicular to the scattering plane) are

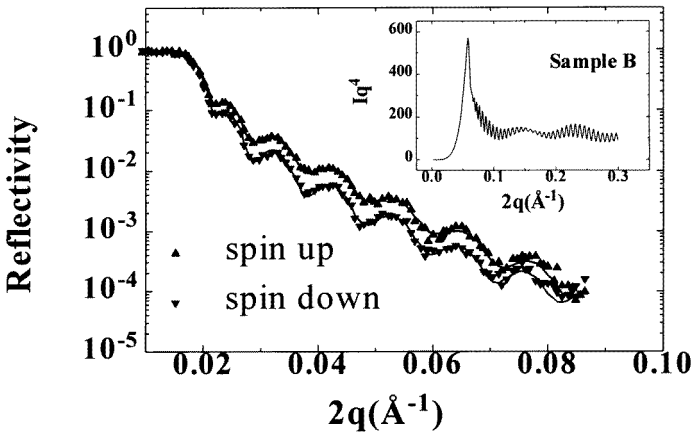

Figure 1. PNR data (triangles) and fits (continuous lines) for sample A. The inset shows the x-ray reflectometry measurement of sample B to confirm the values of thicknesses determined by PNR. The I on the y-scale (in Iq^4) for the x-ray data is the reflected intensity.

reflected off the surface of the sample at grazing incidence. The beam polarization has been found to be better than 98%. All data given are corrected correspondingly. The sample was held at 300 K and during the measurement one side of the sample was covered by a Cd mask. A 6.5 kOe external field was applied in the plane to saturate the sample magnetically and it was confirmed separately by MOKE that this field strength was sufficient to saturate the film in the plane [19]. Figure 1 shows the measured spin-dependent reflectivities of sample A and fits as a function of the neutron scattering vector $(2q)$, where q is the component of the neutron wavevector perpendicular to the plane of the film. Six well defined oscillations are seen in both the spin-up and spin-down reflectivities, and a clear splitting in intensities is observed as expected for a film of this thickness. In fitting the data, the thickness and magnetization were adjusted, and the bulk scattering densities were assumed. The results are summarized in table 1. From the fit to data we can exclude the possibility of a significant magnetic moment depth profile. However, as the neutron q-range is limited, we cannot determine the layer thicknesses with very high accuracy. Therefore, we have performed x-ray reflectometry measurements on both parts of the sample. The results of x-ray measurements for sample B are shown in the inset of figure 1. The Ni thickness is found to be 50 ± 2 Å in agreement with PNR results.

The XMCD experiments were performed at beam line 1.1 of the synchrotron radiation source at Daresbury (UK) with ∼80% circularly polarized x-rays. The $L_{2,3}$ absorption spectra were obtained at room temperature in total-electron-yield mode where the sample current is recorded as a function of photon energy. During the measurement the sample was magnetically saturated in a 1 T field perpendicular to the sample surface. The magnetic circular dichroism signal, $\sigma_M = \sigma^+ - \sigma^-$, is the difference between the x-ray absorption spectra (XAS) measured with the circular polarization of the beam fixed and the sample magnetization parallel (σ^+) and antiparallel (σ^-) to the propagation vector of the light. The sample magnetization was switched by reversing the direction of the field externally with a superconducting magnet.

An important condition that needs to be checked before directly comparing the results of the two techniques is that the two techniques probe the same thickness range of the

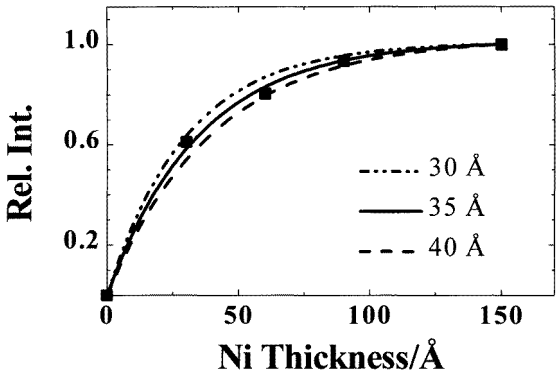

Figure 2. The relative intensity (Rel. Int.) of the Ni L_3 edge jump in the XAS spectra of 30 Å Cu/Ni step wedge (30, 60, 90 and 150 Å)/600 Å Cu/Si(001), normalized by the incident photon flux and, then, divided by the intensity of a 150 Å thick Ni film, is fitted with different effective probing depths (or escape depths) by assuming exponential decay of the intensity with travel distance.

film. PNR investigates the whole Ni film while our detection method (total electron yield) of XMCD has a limited effective probing depth (or escape depth). In order to determine the escape depth we have separately used the absorption spectra of a 30 Å Cu/Ni step wedge/600 Å Cu/Si(001) structure, which was normalized by the incident flux. We determined a probing depth of ∼35 Å by fitting a Ni L_3 edge jump by assuming that the signal from each Ni layer is decaying exponentially with travel distance (figure 2). Therefore, as long as the sample does not have a significant magnetic moment depth profile which has already been confirmed by PNR, the two techniques can be compared.

Figure 3(a) shows the normalized XAS for sample A with two opposite magnetizations. A constant background was subtracted from the absorption spectra, and then the spectra were normalized by the average signal over the higher-energy range where no more dichroic signal is observed. Figure 3(b) shows the corresponding XMCD spectrum which is not corrected for the incomplete polarization of the incident light, and figure 3(c) shows the average XAS spectrum $(\sigma^+ + \sigma^-)$.

The XMCD sum rule states that for 3d transition metals $m_{\mathrm{orb}}, m_{\mathrm{spin}}$ and the magnetic dipole term (m_T) [5, 6] can be determined from the XAS and XMCD spectra using the following equations:

$$\frac{m_{\mathrm{orb}}}{n_h} = -\frac{4q}{3rp_{\mathrm{pol}}} \qquad \frac{m_{\mathrm{spin}} - 7m_T}{n_h} = -\frac{6p - 4q}{rp_{\mathrm{pol}}}. \tag{1}$$

Here, q and r are defined by

$$p = \int_{\mathrm{L_3}} \sigma_M \, \mathrm{d}\omega \qquad q = \int_{\mathrm{L_3+L_2}} \sigma_M \, \mathrm{d}\omega \qquad r = \int_{\mathrm{L_3+L_2}} (\sigma^+ + \sigma^-) \, \mathrm{d}\omega \tag{2}$$

and n_h is the number of holes in the 3d band, and p_{pol} is the degree of the polarization of the light. Figures 3(b) and (c) also show the integrated curves of the XMCD and XAS spectra. The integration of the linearly polarized XAS is obtained after subtracting a step-like background from the XAS spectrum with the steps placed at the peak positions, as shown in figure 3(c) [7, 11, 20]. We assumed the relative step heights above the L_3 and L_2 edges to have the statistical weights of 2/3 and 1/3 respectively. We have tried to estimate the size of m_T by applying the magnetic field perpendicular to the photon incidence direction

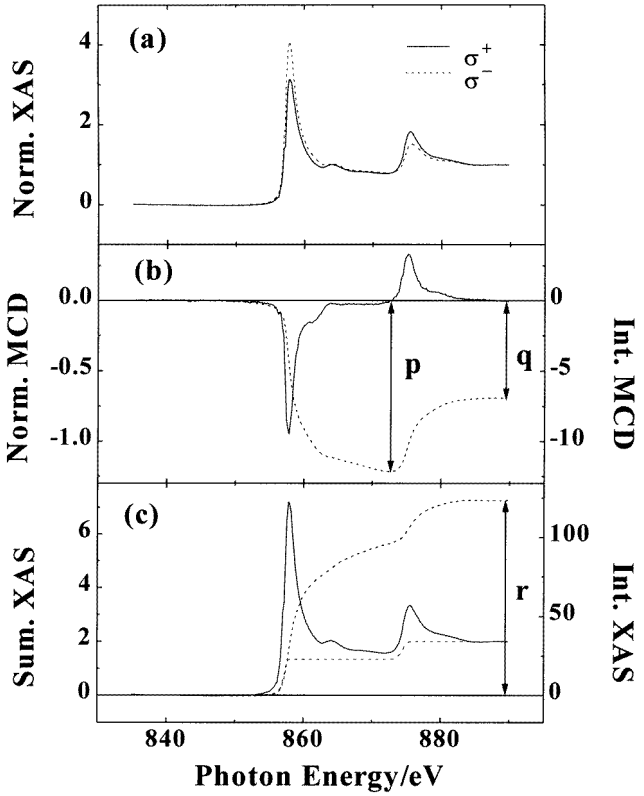

Figure 3. (a) Normalized XAS. (b) Normalized (solid line) and integrated (dotted line) XMCD spectra. These are not corrected for incomplete polarization of light. We choose p as the minimum of the integration curve. (c) Summed XAS, $\sigma^+ + \sigma^-$ (solid line) and the corresponding integrated spectrum (dotted line). The integration curve was obtained by subtracting the two step functions shown from the summed XAS.

in the XMCD measurements, in which the spin magnetic moment disappears [21, 22], but within the noise no contribution was seen. Therefore the total magnetic moment is given by $m_{\mathrm{orb}} + m_{\mathrm{spin}}$.

In order to determine the absolute atomic magnetic moment and, therefore, to compare it with that of the PNR, it is necessary to know p_{pol} and n_h. However, because p_{pol} depends crucially on the exact beam position, the mirror setting, and the aperture, the real polarization can be different from the estimated one [12]. Moreover, no experimental values for n_h are available, while the published theoretical values for n_h vary widely [23]. Using the same model calculation, the number of holes is found to change within a few per cent for different metallic environments [24]. Therefore, we have used the ratio of n_h/p_{pol} determined by an XMCD measurement on a thick polycrystalline film which has a bulk moment. In accordance with this small calculated change, the measured XAS area, which can be assumed to depend on n_h, shows a minimal change for both systems [25] in our experiment.

Table 1 summarizes the results of this work. By comparing the total magnetic moment we can still test the validity of XMCD sum rules in this strained low-symmetric system instead of comparing m_{orb} and m_{spin} separately. We find agreement within experimental

errors between the results of the two different measurements. The agreement of the sizes of the relative changes for the two samples is especially remarkable. XMCD measures the moment due to the 3d band. In reality there are contributions from the 4s and 4p bands [7–9]. For bulk Ni, the contribution of the diffuse moment due to the 4s electrons was calculated to be −7% of total moment [11, 26]. But in our case this contribution is already taken into account in calculating n_h/p_{pol} from the polycrystalline sample and we do not expect much change of this 4s contribution to the total moment compared with that of the bulk because of the broad width of the 4s band.

In conclusion, we directly compared the total magnetic moment determined by XMCD sum rules with the results of PNR measurements on the same strained Ni film prepared on Cu(001) buffer layers. We found an agreement within experimental errors between the two techniques, so directly confirming the validity of the XMCD sum rules in determining the total magnetic moments in these low-symmetry thin-film structures, in which the magnetic moments are different from that of bulk. But we emphasize here that by using a standard reference sample (a thick polycrystalline Ni film) we were able to avoid several difficulties, associated with the determination of n_h and p_{pol}. In view of this problem, we conclude that, in general, the applicability of the XMCD sum rules is limited in practice to measurements of relative changes of magnetic moment.

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