ESRF	**Experiment title:** *The complete experiment of spin-resolved photoemission from the valence band of simple ferromagnets.*	**Experiment number**: HE873
Beamline: ID12B	**Date of experiment**: from: 6/12/2000 to: 12/12/2000	**Date of report**: 26/2/2001
Shifts: 18	**Local contact(s)**: N.B. Brookes	*Received at ESRF:*

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Original motivations and objectives of the experiment

The goal of the experiment was to test the validity and applicability of sum rules for valence band photoemission from 3d transition metals, by performing for the first time the complete spin-resolved photoemission measurement on ferromagnetic thin films. The theoretical framework established by Thole and van der Laan [1] ideally requires the combination of the three possible photon polarisations (right circular, linear and left circular) with the detection of the photoelectron spin polarisation in order to extract from the measured spectra the values of the spin and orbital magnetic moments and of other ground state properties. The quantitative derivation makes use of the intensities of the various spectra as integrated over the whole valence states. We proposed to apply those sum rules to two well known samples, such as Co and Ni films grown epitaxially on the (001) surface of a Cu single crystal. The growth mechanisms and the magnetic properties of a few atomic layers of Co/Cu(001) and Ni/Cu(001) were already described in the literature, so that a little effort was need for the characterisation of the samples. Extremely important was the fact that the samples have a nicely squared magnetic hysteresis loop: not only at room temperature one can reverse the magnetisation with relatively small magnetising fields (H < 100 G), but the remanent magnetisation is very close to the saturation magnetisation. This a very convenient feature for using photoemission, which is incompatible with any kind of magnetic field external to the sample.

Disregarding the angular distribution effects of the emission intensity [2] and assuming that the wide angular acceptance (±20 deg) of the used hemispherical analyser ensures a good integration over many Brillouin zones and over most of the photoelectron diffraction effects, we intended to apply in a direct way the sum rules given in reference [1]. At ID12B we were able to choose the magnetisation orientation and the sign of the photon helicity (left or right circular polarisation), and measuring the degree of spin polarisation of the photoelectrons, all referred to the same direction, i.e. the photo propagation vector. The 8 different spectra which can be measured by combining the values of magnetisation (M=[+], [-]), photon helicity (P=(+), (-)) and photoelectron spin (S=↑, ↓) are independent only 4 by 4. The redundancy is extremely useful for eliminating the instrumental asymmetries, by taking the geometrical average of the equivalent spectra. We define for convenience the following combinations:

Name	M=[+]	M=[-]
a	(+)↑	(-)↓
b	(+)↓	(-)↑
c	(-)↑	(+)↓
d	(-)↓	(+)↑

We then combine the 4 spectra in three ways:

$\tau = a+b+c+d$;

$\sigma = a-b+c-d$;

$\delta = a+b-c-d$.

By integrating the spectra over the whole of the valence band (thus including in our case the *sp*-like and the 3*d* states of cobalt and nickel, i.e. from zero to 12 or 17 eV binding energy) we can try to apply the following sum rules:

$2 < S_z > = (\Sigma / T) * <N_{3d\ electrons}>$;

$< L_z > = -2 (\Delta / T) * <N_{3d\ electrons}>$,

where the capital letters indicate the integrated intensities.

The spin and orbital moment derived in such a way can then be directly compared to those obtained by applying the usual sum rules for the absorption magnetic circular dichroism (MCD) at the $L_{2,3}$ edges of Co and Ni, as measured on the very same samples in the same environment and magnetisation conditions. From this comparison we expect to assess the degree of applicability of the photoemission sum-rules.

Experimental results

In the 18 shifts, in hybrid mode, we could prepare and measure 6 samples:

- 2 times Co/Cu, 7 to 8 ML thick films, magnetised along the sample surface;
- 2 times Ni/Cu, 8 ML thick films, magnetised along the normal to the sample surface;
- 2 times Ni/Co/Cu, 20 ML thick Ni films, magnetised along the sample surface.

In most of the cases the epitaxial growth was checked by LEED.

The contamination level was checked by photoemission spectroscopy and remained tolerable during the whole time of measurement for all of the samples, also in consideration of the moderate surface sensitivity of photoemission when detecting above 500 eV kinetic energy electrons.

We chose the 700 eV photon energy, as the best compromise between cross section evolution and monochromator second order rejection capabilities.

Despite the good performances of the beam line and of the mini-Mott polarimeter mounted on the hemispherical analyser, the experiment could not be made totally. Actually the instrumentation instabilities can be much better compensated in the data treatment for the σ spectra than for the δ spectra. This is because in the former case we make the differences (*a*-*b*) and (*c*-*d*), where the subtracted spectra are measured in parallel. In the latter case we make the difference (*a*+*b*)-(*c*+*d*), were the subtracted spectra are necessarily measured in different moments, and the x-ray beam instabilities can become more dangerous. Furthermore, the (Δ/T) ratio is expected to be at least 10 times smaller than (Σ/T).

This is why we can retain the data only for applying the sum rule on the spin magnetic moment $< S_z >$.

Each sample was measured in spin-resolved photoemission for at least 12 hours. The absorption MCD spectra were taken just before and just after the photoemission series of spectra. In the figures we show the results for one Co/Cu sample and one Ni/Co/Cu sample. The data show a very good reproducibility among equivalent samples, and even the comparison of the two types of Ni films look reasonable, by considering the known trend of the magnetic properties as a function of the thickness.

The quantitative results of $< S_z >$ show systematically that the photoemission sum rule leads to smaller values with respect to the absorption MCD. The discrepancy very much depends on the choice of the number of 3*d* holes (or of the number of 3*d* electrons $<N_{3d\ electrons}>$), and can go from 50% to almost zero, within the range of $<N_{3d\ electrons}>$ given by different authors in the recent literature. We think that understanding this discrepancy is the main issue of the data analysis. Moreover we believe that further experiments allowing to measure the orbital moment would be of the greatest usefulness within this perspective. Consequently, we have succeeded in our goal of having data to test the photoemission spin sum rule but need higher quality data to test the orbital sum rule. The higher stability ands flux on ID8 should make this possible.

[1] G. van der Laan and B.T. Thole, Phys. Rev. B **48**, 210 (1993).

[2] B.T. Thole and G. van der Laan, Phys. Rev. B **49**, 9613 (1994).

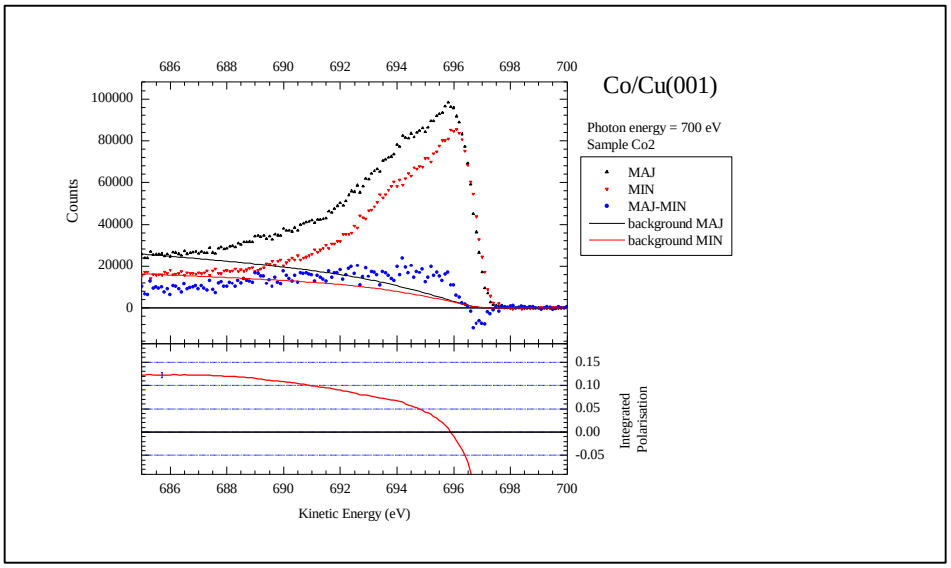

FIGURE 1
Co spin-resolved photoemission spectra, averaged over the two photon helicities

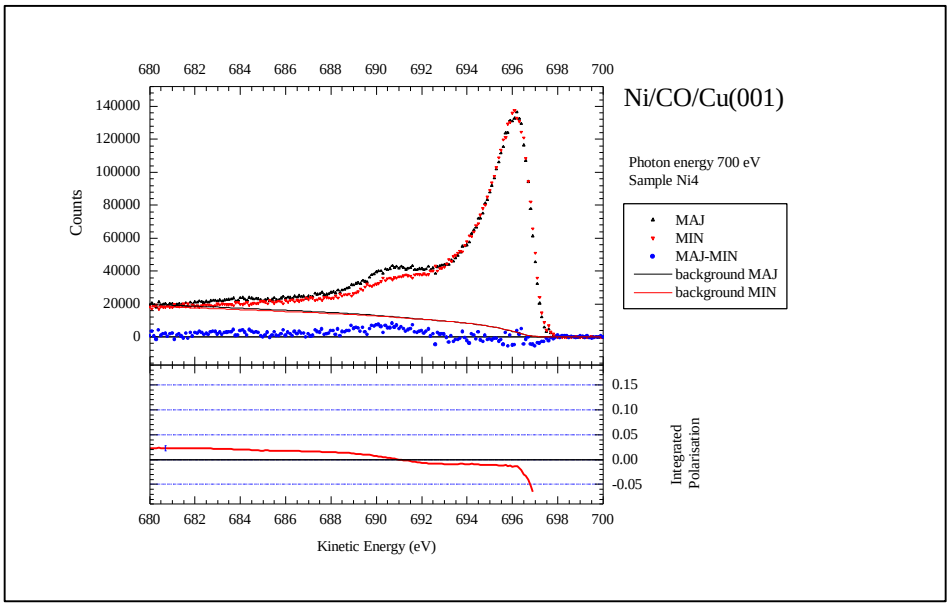

FIGURE 2
Ni spin-resolved photoemission spectra, averaged over the two photon helicities

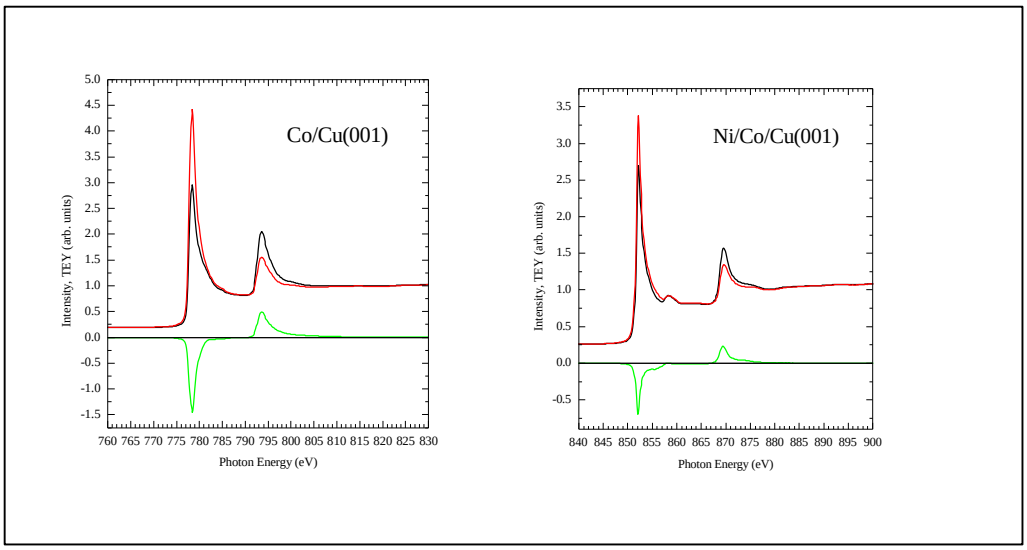

FIGURE 3
Co and Ni absorption MCD spectra