EUROPEAN **S**YNCHROTRON **R**ADIATION **F**ACILITY

INSTALLATION EUROPEENNE DE RAYONNEMENT SYNCHROTRON

ESRF

Experiment Report Form

The double page inside this form is to be filled in by all users or groups of users who have had access to beam time for measurements at the ESRF.

Once completed, the report should be submitted electronically to the User Office using the **Electronic Report Submission Application:**

http://193.49.43.2:8080/smis/servlet/UserUtils?start

Reports supporting requests for additional beam time

Reports can now be submitted independently of new proposals – it is necessary simply to indicate the number of the report(s) supporting a new proposal on the proposal form.

The Review Committees reserve the right to reject new proposals from groups who have not reported on the use of beam time allocated previously.

Reports on experiments relating to long term projects

Proposers awarded beam time for a long term project are required to submit an interim report at the end of each year, irrespective of the number of shifts of beam time they have used.

Published papers

All users must give proper credit to ESRF staff members and proper mention to ESRF facilities which were essential for the results described in any ensuing publication. Further, they are obliged to send to the Joint ESRF/ ILL library the complete reference and the abstract of all papers appearing in print, and resulting from the use of the ESRF.

Should you wish to make more general comments on the experiment, please note them on the User Evaluation Form, and send both the Report and the Evaluation Form to the User Office.

Deadlines for submission of Experimental Reports

- 1st March for experiments carried out up until June of the previous year;
- 1st September for experiments carried out up until January of the same year.

Instructions for preparing your Report

- fill in a separate form for each project or series of measurements.
- type your report, in English.
- include the reference number of the proposal to which the report refers.
- make sure that the text, tables and figures fit into the space available.
- if your work is published or is in press, you may prefer to paste in the abstract, and add full reference details. If the abstract is in a language other than English, please include an English translation.

ESRF	**Experiment title:** Determination of the spin-orbit constant of 3d transition metals using k-integrated spin-resolved photoemission	**Experiment number**: HE-1092
Beamline: ID08	**Date of experiment**: from: 29/08/2001 to: 04/09/2001	**Date of report**: 14/05/2002
Shifts: 18	**Local contact(s)**: Nicholas Brookes	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

Celine DE NADAI* ESRF
Alberto TAGLIAFERRI* Politecnico di Milano, Italy
Nicholas BROOKES* ESRF
Giacomo GHIRINGHELLI Politecnico di Milano, Italy
Jan MINAR* Institut Physikalische Chemie, Muenchen, Germany
H. Ebert Institut Physikalische Chemie, Muenchen, Germany

Original motivations and objectives of the experiment :

The goal of the experiment was to determine the spin-orbit (SO) coupling parameters ξ for a series of 3d transition metals in order to provide non-negligible references in the investigation of theoretical spin-orbit coupling dependent magnetic phenomena. Indeed the SO interaction, namely (ξ <**L**· **S**>**)**, has a strong influence on the electronic structure of materials and directly on the spin and orbital moments. In particular the SO interaction has been related to the magnetocrystalline anisotropy energy (MAE) of magnetic materials containing transition-metal elements, as for instance in van der Laan's model: $\delta E \sim -¼\ \xi$ **S.**[<$\mathbf{L}^{\uparrow}$-$\mathbf{L}^{\downarrow}$>]+$\xi^2/\Delta E_{ex}$[21/2 **S.**<**T**> + 2<$(L_\xi S_\xi)^2$>], showing its great influence on the determination of the magnetic anisotropy of thin magnetic films from XMCD and XMLD experiments.[1] The high importance of this parameter made us considered that the theoretical values of ξ usually used, should be compared to more accurate values extracted from experiments.

We proposed to measure systematically the spin-difference valence photoemission spectra of Co, Fe, Cr and Mn in order to relate it to ξ by comparison with a theoretical description of the Fano-effect in the valence band photoemission (VB-XPS) of the respective metals. This was previously tested by some of us on Cu and Ag, where these fully relativistic calculations, based on the one-step-model of photoemission and relativistic multiple scattering theory, were shown to agree quantitatively with spin- and angle-integrated experimental VB-XPS spectra of pure fcc-Cu.[2] It was shown as well that the shape and the amplitude of the spin-difference are linked to the strength and the diagonal part of the SO coupling.

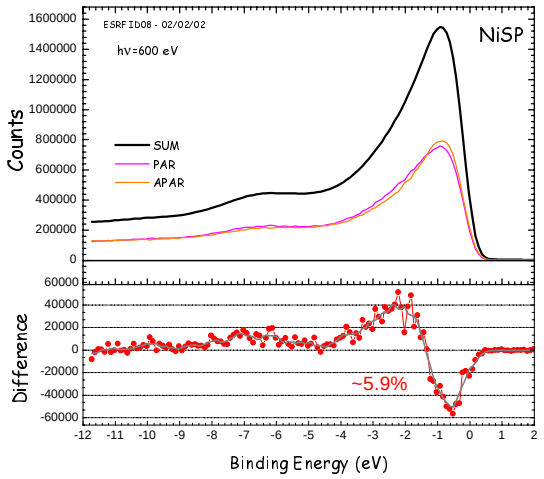
Experimental results :

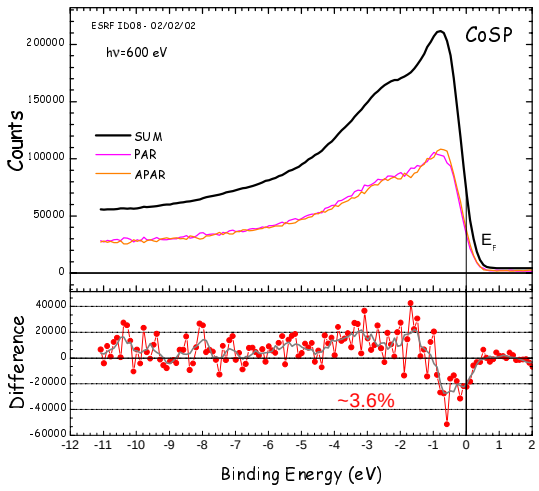
To extract the spin-orbit constant ξ, it was necessary to obtain the spin-orbit coupling, ξ<**L** · **S**>, averaged over all the magnetic effects and looking at the difference of the valence band spectra in terms of parallel and antiparallel alignment of the photoelectron spin and x-ray photon angular momentum. The spin-polarization of the valence band (i.e. the preferential spatial orientation of the photoelectron spin) has been measured with spin-resolved photoemission provided on ID08 using a 140 mm mean radius hemispherical analyser with a ± 20° angular acceptance, assuming a complete integration over the first Brillouin zone,

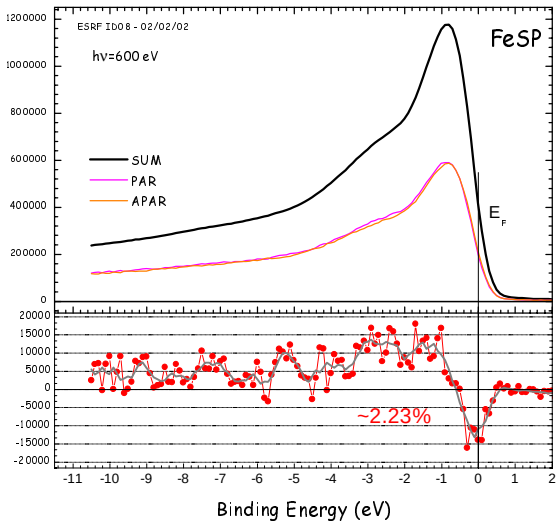
coupled to a mini-Mott 25kV spin polarimeter whom the high efficiency (Sherman function of 17%) and reliability were essential for these low intensity experiments.

In the 18 shifts we could prepare and measure three samples (Fe, Cr and Mn). Supplementary in-house beam time gave us the opportunity to extend the series to Co and Ni. Thick films of ~15-20ML were grown at room temperature in-situ in the XPS chamber in order to reach bulk properties, disregarding the orientation and to obtain a randomly magnetic orientation. This last point was important in order to supress the spin-polarisation of the photo-current due to the spin moment. The absence of contribution from the Cu(001) substrate were checked by XAS and the contamination level before and after measurement by XPS. Absorption MCD spectra were taken just before the spin-resolved photoemission measurements in order to verify the randomly magnetic orientation of the thick films. We chose the 600 eV photon energy as the best compromise between cross section evolution and monochromator second order rejection capabilities. Also in the spin-difference spectra, we have to take into account both $d \rightarrow p$ and $d \rightarrow f$ contributions that are of opposite sign resulting in a low spin-polarisation effect. Minar *et al* [2] showed that with increasing photon energy, the relative weight of the $d \rightarrow p$ contribution drops rapidly to contribute less than 10% above 50 eV.

Each sample was measured in spin-resolved photoemission for at least 12 hours. The measurements were corrected from the geometry and both +/- light helicities were considered in order to eliminate the systematic errors introduced by the experimental apparatus. The following figures show the reliable k-integrated spin-polarised photoemission spectra with good statistics for Ni, Co and Fe. The spin difference in the Mn and Cr cases was found too small to be measured with good statistics and reproducibility in the allocated beam time. Their combination with full relativistic calculations done in München to extract accurate spin-orbit constant are underway. The data were reproducible among equivalent samples.







The spin difference quantification doesn't follow the linear variation with the Z^2 (atomic number) predicted by Mackintosh *et al* [3]. For the interesting transition metals, the measured spin difference was expected to decrease by a factor of 2 in going to Mn, but already a factor of 4 less is observed between Cu and Fe : Cu=9±1%, Ni=6±1%, Co=4±2%, Fe= 2.3±0.8%. In the same way, only a factor of 2 has been determined between the SO constants of Cu and Fe in the case of *ab initio* calculations of the magnetic moments of 3d transition metal impurities in and on the (001) surface of bcc Fe [4]. These discrepancies may be linked to solid state effects.

This work should bring new insights on the SO constant values of 3d transition metal that have to be used in the determination of MAE in thin magnetic films for instance.

This is the first experimental approach of the spin-orbit constant ξ determination in a series of transition metals over the entire Brillouin zone. These measurements of such small entities were made possible in a reasonable time due to the higher flux and degree of polarisation (~100%) of soft x-rays that is provided now on ID08.

References :

[1] P. Bruno, Phys. Rev. B**39**, 865 (1989) ; G. Van der Laan, J. Phys. : Condens. Matter. **10**, 3229 (1998); G. Van der Laan, Phys. Rev. Lett. **82**, 640 (1999)

[2] J. Minar *et al*, Phys. Rev. B **63**, 144421 (2001) ; J. Minar *et al*, Applied Phys. A **73**, 1 (2001)

[3] A.R. Mackintosh, O.K. Andersen, Electrons at the Fermi Surface, ed. Springford (1980)

[4] V. Popescu *et al*, Phys. Rev. B **64**, 184407 (2001)