EUROPEAN SYNCHROTRON RADIATION FACILITY

INSTALLATION EUROPEENNE DE RAYONNEMENT SYNCHROTRON

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.

	Experiment title: XMCD measurements on epitaxial alpha-U thin ilms	Experiment number:
Beamline: ID12	**Date of experiment**: from: 14/11/07 to: 21/11/07	**Date of report**: 11/02/08
Shifts: 18	**Local contact(s)**: Fabrice Wilhelm	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

Dr. SPRINGELL Ross*, E.S.R.F. Grenoble, France

Dr. WILHELM Fabrice*, E.S.R.F. Grenoble, France

Prof. STIRLING William G.*, E.S.R.F. Grenoble, France

Prof. LANDER Gerard H.*, I.T.U. Karlsruhe, Germany

Report:

The actinides occupy a unique position at the end of the periodic table and display some of the most interesting properties of any of the elements. Uranium is the only element to exist in the open orthorhombic (alpha-U) structure at room temperature and is the first of the actinides to have a significant number of electrons in the *5f* shell (between 2 and 3). These electrons can display both itinerant and localised behaviour and are responsible for magnetic ordering in a large number of uranium compounds. Alpha-U itself is paramagnetic with a near temperature-independent susceptibility of 380×10^{-6} emu/mole, which would translate to a moment of 0.7 $m\mu_B/T$, which should be easily detectable on ID12 based on our experience of small induced U *5f* moments, which we have measured previously. Experiments by Maglic *et al.* on the induced form factor f(Q) with polarised neutrons were interpreted by Hjelm *et al.* to exhibit a breakdown in Hund's third rule, where the spin and orbital moments were parallel to one another for a less than half-filled *5f* shell. We report here, the first attempt to measure the paramagnetism of uranium, using the XMCD technique.

Sample	Composition	Measurement
SN205	5000 Å alpha-U film [110] on sapphire	U M_4 edge, 5K, 6T
		U M_5 edge, 5K, 6T
Alpha-U foil	70 μm foil	U M_5 edge, 5K, 6T
		U M_4 edge, 5K, 6T
		hysteresis loop at M_4 edge 6T to -6T, 5K
		U M_4 edge, 300K, 6T
		U M_5 edge, 300K, 6T
SN181	500 Å *hcp*-U film [001] on sapphire	U M_4 edge, 5K, 6T

Table 1- Summary of the sample compositions investigated in this experiment and the measurements carried out on each sample. N.B. All samples were mounted in normal incidence.

The samples investigated are summarised in Table 1. The thin film samples, SN205 and SN181 were prepared at the Clarendon Laboratory in Oxford, UK, using dc magnetron sputtering in UHV. The substrates were epi-polished, single crystal sapphire plates, oriented [11.0]. Buffer layers of *bcc* [110] niobium, grown as a single domain, were employed to seed the growth of the uranium. Nb capping layers were used to protect the samples from oxidation. For the case of SN181 a further seed layer of Gd was used to promote the growth of *hcp*-U. The alpha-U foil was prepared at the Los Alamos National Laboratory, NM, US.

A summary of all of the measurements carried out during this experiment can be found in Table 1. XANES (x-ray absorption near-edge spectrum) spectra were recorded at both the U M_4 and U M_5 edges in a backscattering geometry with the angle of the incident x-ray beam normal to the sample surface. The XMCD spectra were obtained by flipping the helicity of the incoming circularly polarised x rays in a field of 6T. To ensure that the XMCD spectra are free of any experimental artifacts, the XMCD spectra were collected for both directions of the applied magnetic field (parallel and antiparallel to the incoming x-ray beam). The applied magnetic field was produced by a superconducting cryomagnet. At the energies of the U M_5 (3.55keV) and UM_4 (3.73keV) edges the Bragg angle of the double Si (111) crystal monochromator is above the Brewster angle and the degree of circular polarisation of the monochromatic beam is reduced to 35% and 45%, respectively.

Our initial investigation was into the paramagnetism of U-metal in its bulk, orthorhombic phase. Sample SN205 was studied initially, since the 5000 Å film is a single crystal alpha-U (oriented (110)) sample with a mosaic of less than 0.5º and is capped with a niobium layer for oxidation prevention. There was little or no detectable dichroism at either the M_4 or M_5 edges, see figure 1 which shows only the M_4 edge as an example.

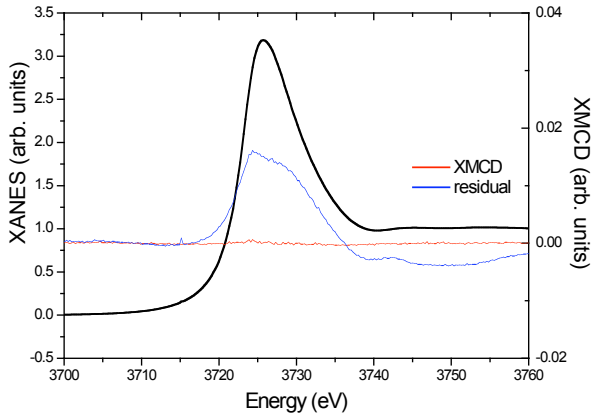

Figure 1 – XANES and XMCD signals measured at the U M_4 edge at 5K in 6T applied field for sample SN205.

Shown here are the XMCD and residual intensities taken as (xmcdp-xmcdm)/2 and (xmcdp+xmcdm)/2, respectively. In this case xmcdp is equivalent to the XMCD measured with the magnetic field applied parallel to the x-ray beam and xmcdm is for the field antiparallel. There is little detectable dichroism in this case and therefore no detectable paramagnetism in the U *5f* states for this sample. The large residual signal indicates that there may be a component of linear dichroism. Modifications to the bulk properties of metals by the growth of thin films is not uncommon and it is possible that the 5000 Å does not exhibit the same magnetic properties as that of bulk alpha-U. In order to further explore this point we carried out a similar set of measurements on an alpha-U single crystal foil (70 microns thick), which has been shown previously to exhibit bulk properties in close agreement to those acepted for alpha-U.

Figure 2 shows the XANES and XMCD spectra measured for the U foil at 5K and at 300K. A hysteresis loop measured at 5K is shown in figure 3. The XANES spectrum displays some near edge features, which resemble those of UO_2 and the XMCD varies significantly with temperature, which suggests that the origin of the dichroism, although magnetic may come from the paramagnetic state of UO_2 (which has an antiferromagnetic transition at 30.8K) and a much larger susceptibility than U metal, which is temperature dependent. The presence of an oxide layer on the U foil is not unexpected, but is of a large enough magnitude to render the deciphering of any signal from the paramagnetic alpha-U almost impossible. Future measurements are planned on a freshly prepared U foil with no oxide layer.

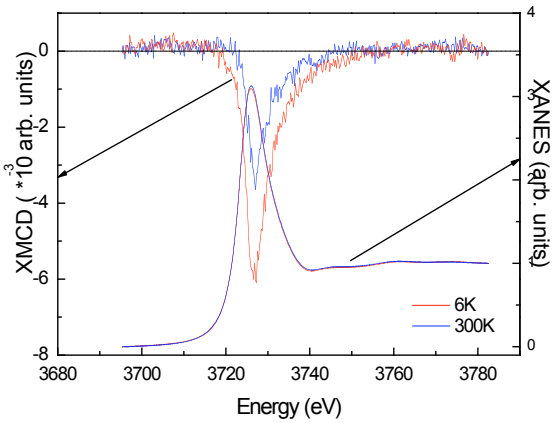

Figure 2 – XANES and XMCD spectra measured at 6K and 300K for the alpha-U foil in an applied field of 6T.

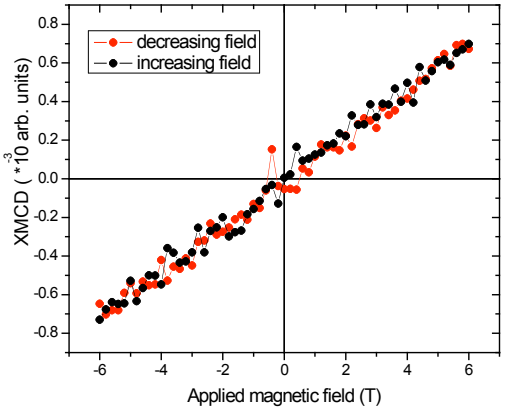

Figure 3 – XMCD signal as a function of field, measured at 6K .

We also report XANES and XMCD measuerements at the U M_4 edge on an *hcp*-U sample. Theoretical calculations have recently predicted magnetism in this structure, so we have probed the *5f* states for any evidence of dichroism. Figure 4 shows the measured XANES and XMCD spectra for sample SN181. The red and blue spectra represent the XMCD and the residual intensities, respectively. If the signal were magnetic in origin one would expect a reversal in sign as the field is reversed, but this was not the case and the XMCD, taken (xmcdp-xmcdm)/2, is negligible. Further investigations are planned to probe the magnetism of the *hcp*-U structure. The same conclusion is to present the xmcd and residual signal together, The residual signal may again arise from linear dichroism.

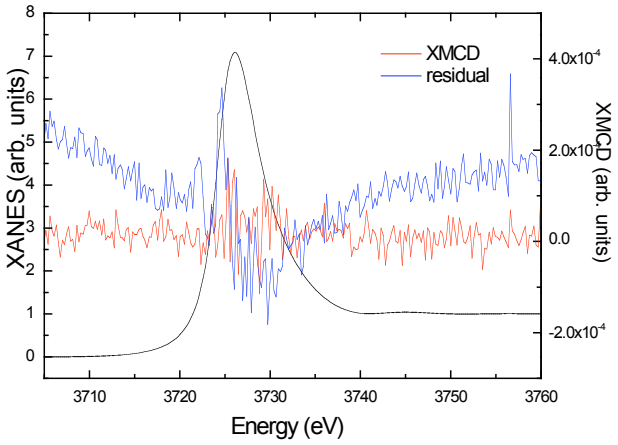

Figure 4 – XANES and XMCD spectra measured on sample SN181, at 6K, in an applied field of 6T.