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- 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:** XMCD measurements of UGe_2 under pressure	**Experiment number:** HE-3449
Beamline: ID12	**Date of experiment:** from: 2/12/10 to: 10/12/10	**Date of report:** 01/03/11
Shifts: 18	**Local contact(s):** Fabrice Wilhelm, Andrei Rogalev	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

Dr. SPRINGELL Ross*, LCN, UCL, London, UK

Dr. WALKER Helen*, E.S.R.F. Grenoble, France

Mr. BOSEGGIA Stefano*, LCN, UCL, London, UK

Report:

The discovery that ferromagnetism and superconductivity could coexist sparked a wave of scientific interest at the beginning of the millennium [1], yet there are still few materials that display this unique behaviour. The most commonly studied, is the compound UGe_2, which has pressure-temperature (see fig. 1) and magnetic field-temperature phase diagrams, rich with unusual physical phenomena [2]. Our proposed experiment attempted to measure the XMCD at the U $M_{4,5}$ edges (3728eV and 3552eV, respectively) at low temperatures (approx. 10K) as a function of pressure. These energies are at the soft end of the hard x-ray regime, as are many other absorption edges of experimentally interesting materials, not least other U-containing compounds of significant interest in the pressure domain, such as UIr, URhGe and UCoGe. It is this advance; in-situ pressure-dependent measurements of XMCD at these low energies in a magnetic field and at low temperatures, that will open up a whole new field of research at the ID12 beamline. The following report will outline a number of crucial steps and hurdles that have been overcome in order to achieve this goal.

The experiment was conducted in the second experimental hutch on ID12. We used the large, warm bore magnet, capable of temperatures down to 5K and magnetic fields of several Teslas. In order to reach the required pressures to study the phase region of interest it was necessary to use a diamond anvil cell (DAC). The warm bore magnet allowed access for cells up to 59 mm in diameter (easily achievable for the majority of available DACs). The standard set-up in an x-ray experiment under pressure involves illuminating the sample with access through the diamonds (see fig. 2). However, at the lower end of the hard x-ray regime, in the vicinity of the U $M_{4,5}$ edges the absorption due to

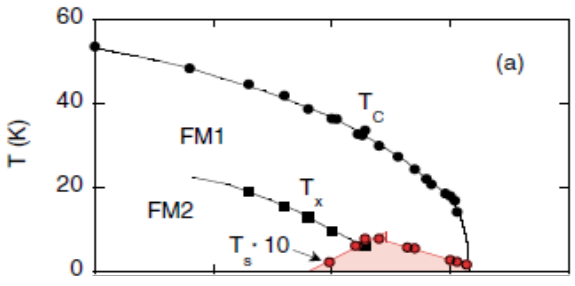

Fig. 1 The p versus T phase diagram of UGe_2. T_C is the Curie temperature and T_X is defined in the text. T_S is the superconducting temperature (onset). The lines through the data points are a guide to the eye, noting that T_S might change discontinuously at p_X and p_C.

the diamond anvils is very large, in fact, even for 100 microns thickness of diamond the transmission is only about 30%. Thus for thicknesses on the order of millimetres, the transmission falls well below 0.001%.

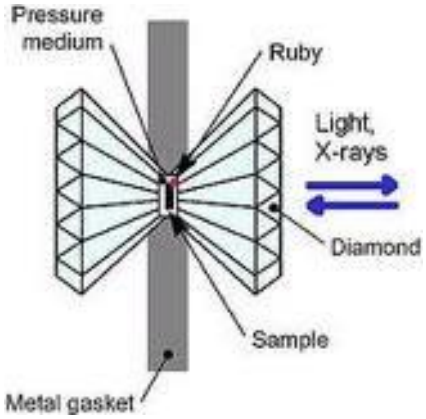

Fig. 2 - Standard geometry for a fluorescence measurement under pressure from a DAC, where the incident and detected x-rays both pass through the diamond anvils.

There are broadly two solutions to this problem:

1) To use perforated diamonds – in this case, one of the diamonds is cut so that the thickness of the anvil at the culette is only 100 microns thick.

2) To use a panoramic DAC – here, the access to the sample is through the gasket, hence the gasket material can no longer be a strong metal (e.g. rhenium), but must be a low Z substance of small wall thickness; usually Be or B (plus ¼ epoxy).

We attempted both of these solutions to some degree or other and a detailed description of their employment and experimental modifications will follow.

The panoramic cell available was made of a stainless steel, but was too sensitive to the strong magnetic fields inside the cryomagnet to be feasible in the planned XMCD measurements. However, the principle of a beryllium-copper (BeCu) panoramic cell, unaffected by magnetic field, is a very attractive one and would be an ideal solution. For the majority of this experiment we were restricted to the use of an amagnetic DAC (BeCu) with a perforated diamond (culette of ~200 microns). The thickness of the perforated diamond was nominally 100 micron, but in reality was closer to 200-300 microns. This difference is significant. The profile of the transmission through C, Be and B are summarised in figure 3 for 100 microns thickness.

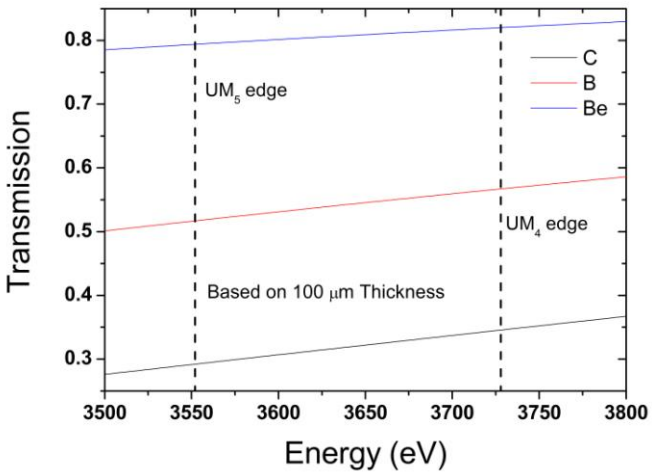

Fig. 3 - Transmission properties of x-rays of energies close to the uranium $M_{4,5}$ edges through diamond, boron and beryllium.

Figure 4 – The three principle components of an amagnetic DAC. From left to right; The base with attached solid diamond anvil, the top segment with perforated diamond, the helium gas membrane pressure drive.

The geometry of the experiment was carried out as in figure 2, where the diamond on the incident x-ray beam side was perforated. A rhenium gasket and He gas were used as the pressure medium, encased by the two diamond culettes. The amagnetic cell comprises of three principle sections, see figure 4. The diamonds were first carefully aligned and a rhenium gasket cut to size. The gasket was then indented to a thickness of just 80 microns, using a small amount of pressure. A hole of approx. 150 microns was then cut out of the centre of the indented region. The gasket was mounted on the base diamond and the sample inserted. A gold wire (25 micron diameter) was also attached to the sample in order to provide the means for total electron yield (TEY) measurements to accompany the standard fluorescence yield (FY).

A first test was made with the DAC mounted from the back port of the cryomagnet on a solid aluminium rod/plate as in figure 5. This first measurement was made at room temperature, in order to first test if the TEY and FY signals at both the Ge K edge and U M edges were detectable through the diamond culettes. The tests proved successful, although the detection of the sample was not easy, since the sample size was restricted to approx. 100 * 100 microns only.

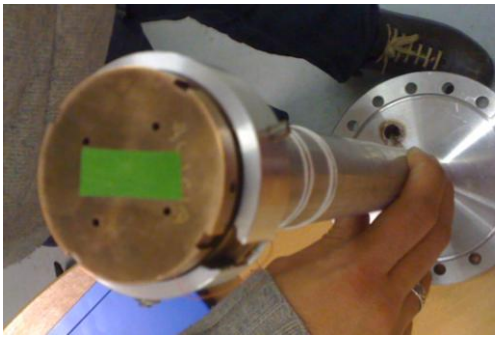

Figure 5 – mounting assembly from the back of the warm bore magnet.

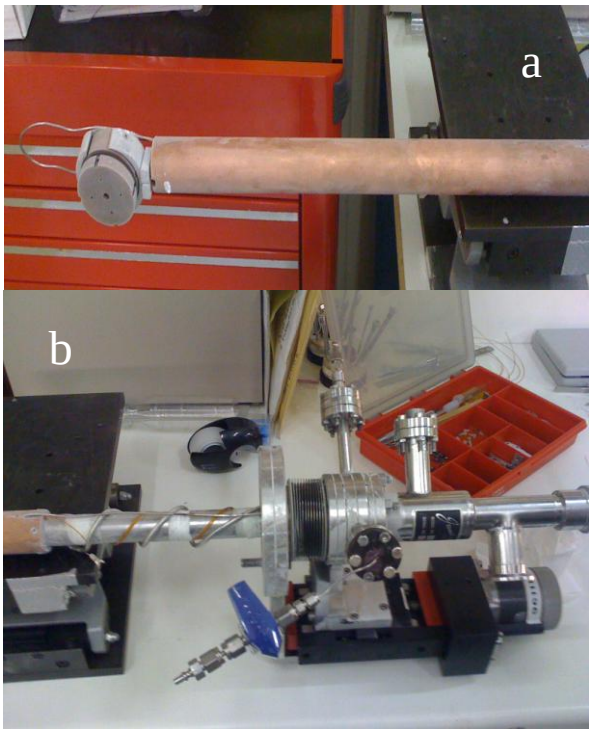

Figure 6a shows the rod with attached DAC, pressure line and copper shielding. 6b shows the collar of the cryostick that houses the connections to the TEY, heater, thermometer and pressure inlet. The blue valve at the bottom of the image is connected to the pressure line.

The next test was to attach the DAC to the cryostick, mounted from above. This orientation provides an extra degree of freedom for sample manipulation. The remaining experimental difficulties were to cool the DAC assembly and to apply pressure in-situ, maintaining the He gas line to the DAC in vacuum. The cryostick was adapted so that a pressure line followed the length of the rod and attached to the DAC. The stick was enveloped in a copper shielding in order to reach the lowest temperatures and a valve fitted to the external pressure outlet (see figures 6a and b). The valve allowed the pressure to be increased to the He gas membrane of the DAC and then closed, this was so that the gonfleur could be removed when the magnetic field was switched on; the stray fields are large enough from the warm bore magnet that the steel pot inside the gonfleur could be dislodged from its seat.

Once all of the obstacles had been overcome, there was only one night remaining to measure the XAS and XMCD signals. It was impossible to probe as much of the pressure-temperature phase diagram as was hoped, and it proved very difficult to detect the sample and there was not time enough remaining by the end of the experiment to get reasonable statistics on the U $M_{4,5}$ edges.

Provided more time is given for this experimental enquiry it would be better in the future to use a specially designed DAC for pressure measurements of XMCD signals at energies in the vicinity of the U $M_{4,5}$ edges. The ideal solution would be an amagnetic (BeCu construction for example) panoramic DAC. A design and images for such a cell are shown in figure 7a-d. The advantages over the perforated diamond set-up are numerous and could result in an improvement in statistics of more than two orders of magnitude:

1) Penetrating through Be or B gaskets, where the walls are only 75 microns thick instead of through the diamond anvils, reduces the absorption by a factor of ~10.
2) We can use solid diamonds (not perforated), which means that anvils with large culettes can still be used to reach reasonably high pressures (several GPa). The larger culettes mean that larger samples can be illuminated; an increase in size and therefore signal by a factor of ~4.
3) The panoramic cell dramatically increases the solid angle detected by the fluorescence photodiode (approx. 2cm from the DAC), compared to the standard set-up. The improvement in detected signal of approx an order of magnitude.

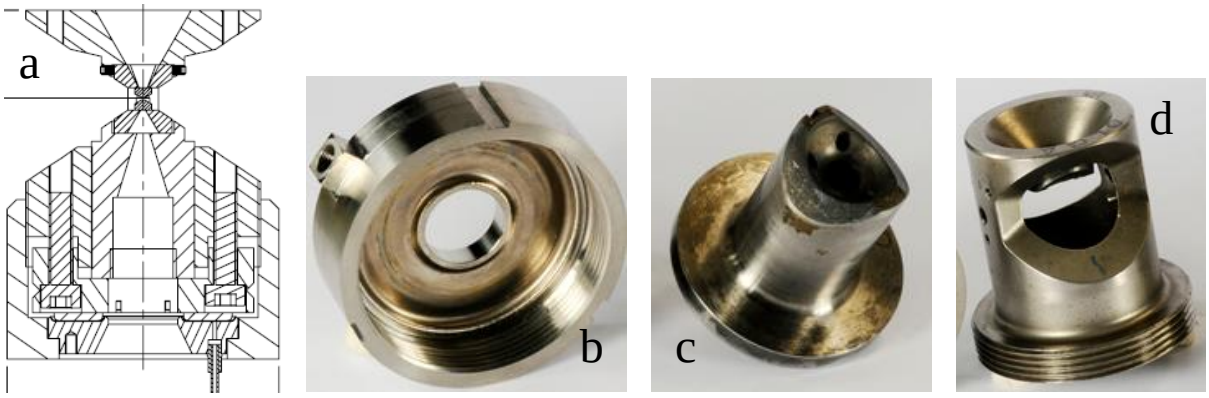

Figure 7 – a) is a schematic of a panoramic DAC of the sort that would be ideal in BeCu for XMCD measurements under pressure at the U M4,5 edges. 7b-d show images of the three components, equivalent to those of the standard DAC, the He gas membrane, the base pedestal and the top anvil mount, respectively.

The only drawback of this technique is the more complicated gasket fabrication, but modern laser cutting techniques can easily overcome this hurdle. In fact, at the beginning of this experiment we prepared several gaskets of this type.

Since this experiment we have sourced some funding for the development of a DAC, specifically for fluorescence in a magnetic field. We hope to continue this field of enquiry on the ID12 beamline, working together with the beamline staff to promote and improve the in-situ pressure sample environment. The potential for future investigation is vast and would provide a unique facility in the world at the present time for XMCD under pressures at low x-ray energies.

References

[1] S. S. Saxena *et al.* Nature **406**, 587 (2000)
[2] C. Pfleiderer and A. D. Huxley, Phys. Rev. Lett. **89**, 147005 (2002)