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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 via the User Portal:

https://wwws.esrf.fr/misapps/SMISWebClient/protected/welcome.do

Reports supporting requests for additional beam time

Reports can 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 valence state of Pu in $PuCoGa_5$**	**Experiment number:** **HC-1793**
Beamline: ID12	**Date of experiment:** from: 17-June-2015 to: 23-June-2015	**Date of report:**
Shifts: 18	**Local contact(s):** F. Wilhelm	*Received at ESRF:*

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Report:

$PuCoGa_5$ is a material that is a well-known superconductor at T_c = 18 K [1]. The mechanism of electron pairing in this material is unknown, but it certainly involves *d*-wave symmetry [2]. The first aspect of these experiments was to determine the valence of the Pu atoms in the structure. Already neutron experiments have shown that the Pu ion is not in a simple trivalent (Pu^{3+}) state with five 5*f* electrons [3]. DMFT calculations [4] have suggested that the Pu ground state is a mixture of $5f^5$ and $5f^6$ states, and predicted a value for the ratio $R = \mu_L/\mu_S$, where μ_L and μ_S are the orbital and spin moments, respectively. For a classic $5f^5$ trivalent Pu ion this value is R = –1.36, whereas the DMFT theory gives R = – 1.74. The second aspect was to examine the superconducting state (T < 18 K) and see whether the signal changed.

The data obtained at the lowest temperature are shown in Fig. 1. The spectral shape is unusual; the M_5 spectrum consists of two contributions, whereas the M_4 spectrum is very narrow (FWHM ~ 5 eV). Both these features are very similar to those found in a *localised* $5f^5$ system PuSb [5], and quite different from those found for the *itinerant* system $PuFe_2$ [6].

Treatment of the data shows that the branching ratio is close to that found for other Pu systems, and the total moment induced by the applied field of 17 T is 28(5) $m\mu_B$, which consists of μ_L = + 68(7) $m\mu_B$ and μ_S = – 40(4) $m\mu_B$, giving an R = – 1.70(25), in remarkable agreement with the DMFT [4] and with the neutron experiments [3] showing that the ground state is *not* purely $5f^5$.

Lowering the temperature below T_c showed that the signal slowly increased – as shown in Fig. 2. This increase is not understood. The situation for $T < T_c$ is complicated as for $H_{c1} < H < H_{c2}$ in a type-II superconductor (the situation here) there is a field penetration depth λ, which at T = 0 and H = 0 is 265 nm [7]. For H = 17 T this probably increases to almost 300 nm. The penetration depth for photons at the M_4 edge of Pu is also of this magnitude, making it unlikely that the x-rays penetrated into the superconducting material. It seems probable, therefore, that we are still probing the *normal* state even with the sample temperature below T_c. Furthermore, *if* we were probing the s/c state we should expect to see a reduction in the spin susceptibility, as observed in the NMR experiments [8], and therefore a change in the spectral form of the XMCD signal. This was not observed. On the other hand, in the normal state the susceptibility is essentially independent of temperature, so a Curie-Weiss-like rise at the lowest temperatures is unexpected.

Further considerations of the data are now in progress.

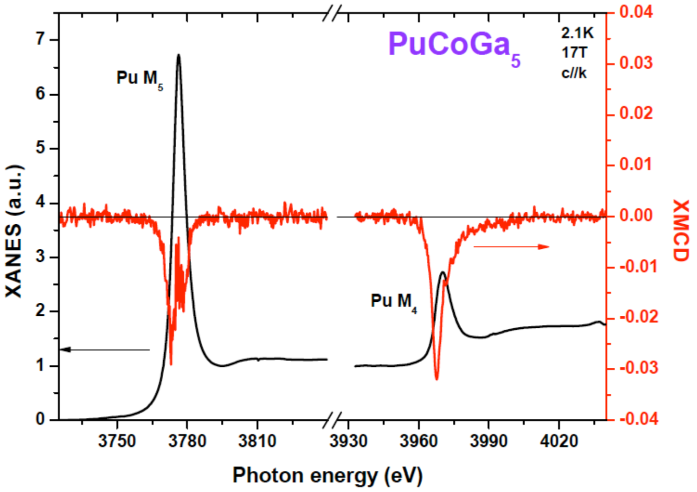

Fig. 1.
XANES and XMCD of $PuCoGa_5$ obtained at ID12 at 2.1 K with an pplied field of 17 T. Note that the spectral form of the XMCD signal does not change between the normal and superconducting states.

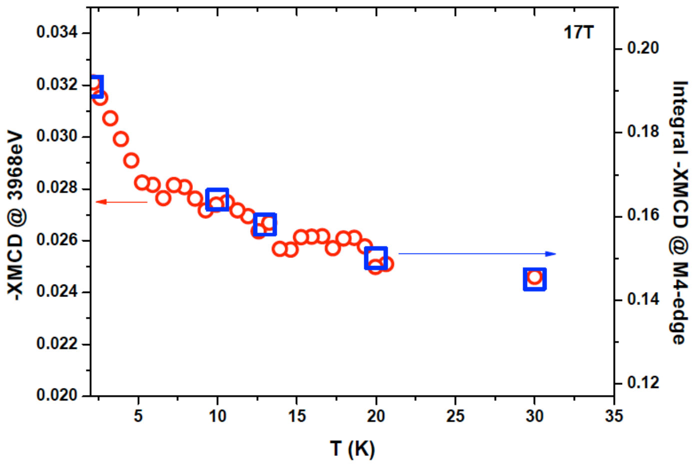

Fig. 2
Results for XMCD at low-*T* as measured at 17 T on ID12.

The penetration depth λ does not change much below ~ 8 K but we do not understand the increase at low temperature.

References
[1] J. Sarro *et al.,* Nature **420**, 297 (2002)
[2] D. Daghero *et al.,* Nature Comm., DOI: 10.1038/ncomms 1785 (2012)
[3] A. Hiess *et al.,* PRL **100**, 076403 (2008)
[4] M. E. Pezzoli *et al.,* PRL **106**, 016403 (2011)
[5] M. Janoschek *et al.,* PRB **91**, 035117 (2015)
[6] F. Wilhelm *et al.,* PRB **88**, 024424 (2013)
[7] R. Heffner *et al*., PRB **76** 064504 (2007)
[8] N. J. Curro *et al*., Nature **434**, 622 (2005)