EUROPEAN SYNCHROTRON RADIATION FACILITY

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 via the User Portal:
https://wwws.esrf.fr/misapps/SMISWebClient/protected/welcome.do

Deadlines for submission of Experimental Reports

Experimental reports must be submitted within the period of 3 months after the end of the experiment.

Experiment Report supporting a new proposal ("relevant report")

If you are submitting a proposal for a new project, or to continue a project for which you have previously been allocated beam time, you must submit a report on each of your previous measurement(s):

- even on those carried out close to the proposal submission deadline (it can be a *"preliminary report"),*
- even for experiments whose scientific area is different form the scientific area of the new proposal,
- carried out on CRG beamlines.

You must then register the report(s) as "relevant report(s)" in the new application form for beam time.

Deadlines for submitting a report supporting a new proposal

- 1st March Proposal Round - **5th March**
- 10th September Proposal Round - **13th September**

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.

Instructions for preparing your Report

- fill in a separate form for each project or series of measurements.
- type your report in English.
- include the experiment number 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:** High pressure magnetic properties of Mo4Ce4Al7C3, a new nano-lamellar phase combining ferromagnetism, Kondo lattice properties and mixed valence	**Experiment number**: HC-4131
Beamline: ID12	**Date of experiment**: from: 03 Feb 2021 to: 09 Feb 2021	**Date of report**: 01/09/2021
Shifts: 18	**Local contact(s)**: Fabrice Wilhelm	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

BARBIER Maxime* (ESRF + LMGP)

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Report:

The goal of the experiment was to carry out high pressure (HP) XAS measurements at the L_3-edges of Ce and Mo in the magnetic layered compound $Mo_4Ce_4Al_7C_3$. In particular, we wanted to assess the evolution of the Ce mixed valence up to 40 GPa, and to see if some of the electronic charge leaving the Ce 4*f* orbital as pressure (P) increase was transferred to the Mo 4*d* orbital. We had also planned on measuring the evolution of the XMCD signal at the Ce $L_{3,2}$-edge as a function of P, but a technical issue with the cryo-magnet kept us from performing these measurements.

A preliminary HP XANES experiment had been conducted on the Ce L_3-edge at ID12, with P going up to around 20 GPa. This showed a linear evolution of the 4*f* valence from ~0.75 (i.e 75% of the Ce atoms in the $4f^1$ configuration, the rest in $4f^0$) down to ~0.55 at 20 GPa. At this point, our hypothesis was that one of the two non-equivalent Ce lattice sites was fully $4f^1$ and carried the ferromagnetism of the compound, while the other one was $4f^0$ and in a mixed valence state. This implied that passed a certain applied pressure, this mixed valence site would be fully emptied of its 4*f* electronic charge, bringing the total valence of the compound to 0.5. This called for a new HP experiment going to at least 30 GPa, giving us a chance to witness this saturation at 0.5. This was done using a diamond anvil cell (DAC) with a partially perforated diamond, in order to increase the signal. The resulting XANES spectra as well as the evolution of the valence as a function of pressure are displayed on Figure 1 (a) and (b),

respectively. There is a stabilisation at a valence of ~0.5, which proves the assumption mentionned above.

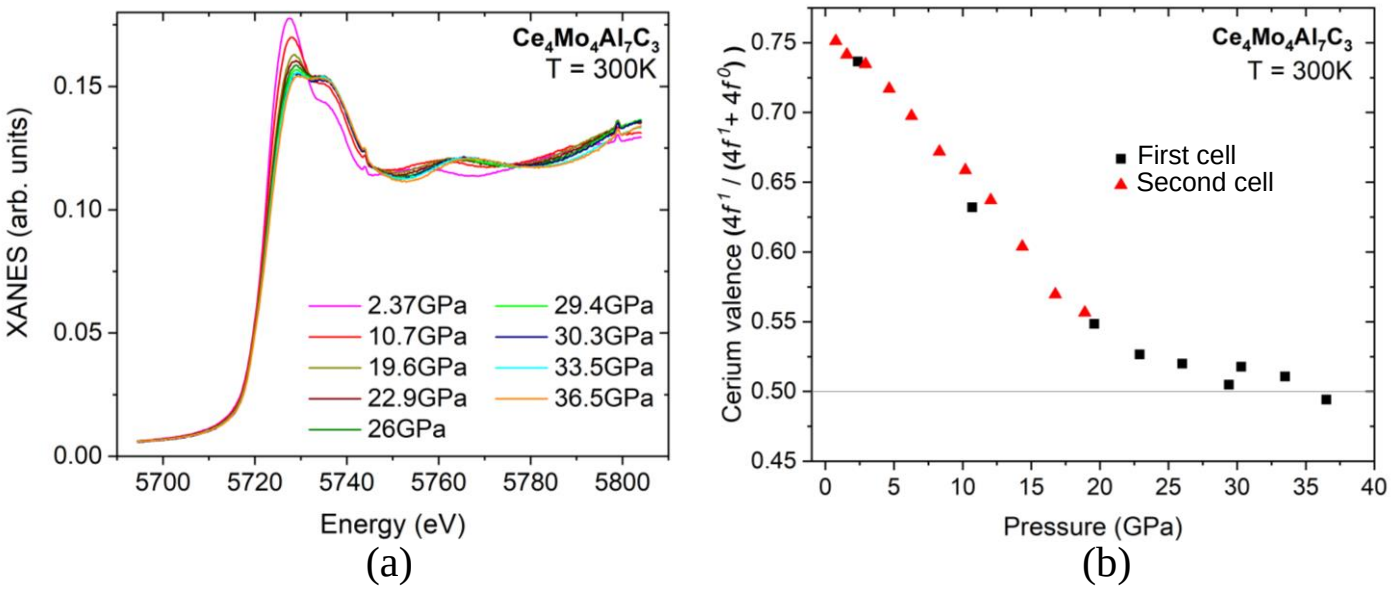

Figure 1: (a) Ce L_3-edge raw XANES spectra as a function of applied pressure. (b) Evolution of the mixed valence deduced from these spectra, with the data from the first HP experiment as well as this experiment.

The HP study at the L_3-edge of Mo was conducted using a fully perforated diamond, along with a thin diamond window. Indeed, given the considerable absorption in diamond at the energy of this edge, a partially perforated diamond would not allow for a good signal. This causes experimental constraints, and limits the attainable pressure at around 10 GPa. Nevertheless, as can be seen on figure 2, there is not a strong evolution of the shape or intensity of the white line as a funtion of pressure, which tells us that the Mo 4*d* valence remains quite stable up to 6.5 GPa.

Both these results will be published in a letter that will be submitted this autumn.

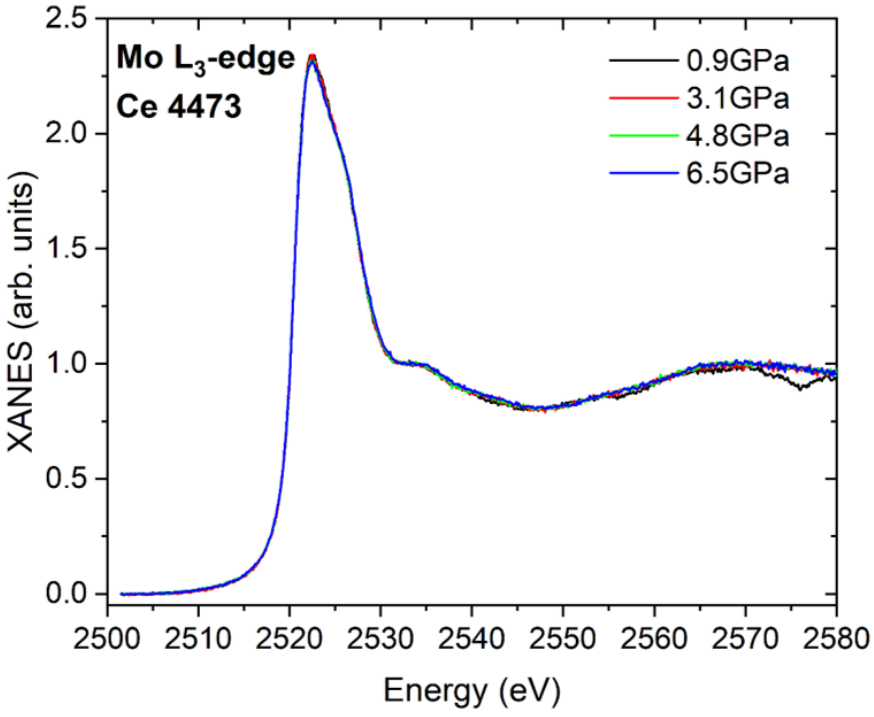

Figure 2: Evolution of Mo XANES with applied pressure.