EUROPEAN SYNCHROTRON RADIATION FACILITY

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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 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:** Inelastic X-ray Scattering Study of the Alpha-Uranium Phase of Cerium	**Experiment number**: HS-3578
Beamline: ID28	**Date of experiment**: 1-4/03/2008	**Date of report**: 31/08/08
Shifts: 9	**Local contact(s)**: A. Bossak	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

M.I. McMahon School of Physics and Centre for Science at Extreme Conditions
I. Loa* The University of Edinburgh
J. Jeppsson* Edinburgh, U.K
L.F. Lundegaard
G. Stinton.

Report: Uranium has the orthorhombic α-uranium (α-U) structure at ambient conditions. This phase has been the subject of a very large number of physical studies at low temperatures because of the formation of an incommensurately-modulated phase below 43K due to a charge-density wave (CDW). For a review of work prior to 1984, see [1].

Extensive inelastic neutron scattering studies of uranium at 300K using single-crystal samples revealed unusual phonon behaviour along the [100] direction, in particular a pronounced softening of the LO Σ_4 branch near q=(½,0,0) – see Figure 1 [2]. With decreasing temperature this branch showed a further softening, and low-temperature elastic neutron scattering studies revealed that the CDW below 43K corresponded to the condensation of this phonon. Prior to recent discoveries in a wealth of different elements at high pressure [3], uranium was the only element known to have an incommensurate phase.

The α-U structure also exists in the lanthanide elements as a high-pressure phase [4], and the very unusual lattice dynamics found in uranium raises the question as to whether similar behaviour would be found in other α-U phases. And, if similar phonon softening *is* found in other α-U phases, does this point to the formation of a CDW state at low temperatures in the same systems? We have reported a method by which single-crystals of the α-U phase of cerium can be grown above 5GPa [5], and in this inelastic x-ray scattering experiment we used such a sample to investigate the lattice dynamics of the α-U phase of cerium at 6GPa and 300K.

This experiment (HS-3578) was awarded 3 days of beamtime on beamline ID28. The crystal's orientation was determined previously at the SRS synchrotron, and the sample was quickly aligned on ID28. The high-quality crystal meant that high-quality spectra were obtained, as illustrated in Figure 2. The complexity of the expected phonon dispersion – obtained theoretically by B. Johansson and colleagues in Uppsala via electronic structure calculations – and the resulting complexity of the inelastic spectra meant that calculations of expected IXS peak intensities were essential in order to optimise data collection. In particular, it was essential to determine along which lines in reciprocal space to perform the IXS measurements in order to obtain optimal contract between the phonon branch of interest and neighbouring branches.

Selected experimentally-determined phonon branches are shown in Figure 3. The key results are the unexpectedly soft phonon branches around the Y point, both along (000)—(100) and (100)—(110), that are reminiscent of the softening observed near (½ 0 0) in uranium. It suggests the possibility that a CDW state forms in Ce at low temperatures with a modulation vector near (0.9,0,0).

However, the scattering geometries that would have provided the highest contrast between the

relevant phonon branch and neighbouring branches were not accessible due to the crystal orientation within the pressure cell (something that is not controllable). Further experiments on a sample with a different orientation relative to the DAC would allow us to obtain conclusive results. Further IXS beamtime would also be needed to collect additional data on selected optical branches in order to have sufficient coverage of the phonon spectrum for deriving a reliable model of the complete set of phonon dispersion curves.

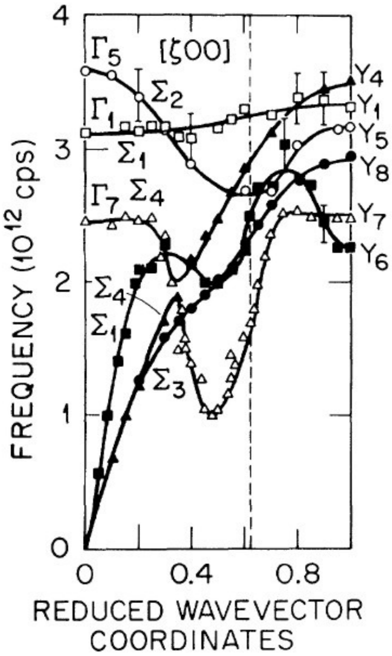

Figure 1: Dispersion curves along [100] in α-U uranium at 300K, showing the striking anomaly in the Σ_4 branch near (½,0,0) [from Ref. 2].

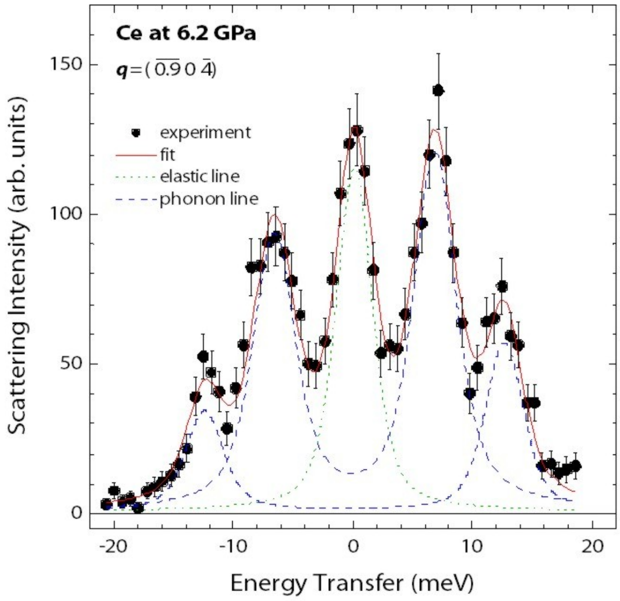

Figure 2: IXS spectrum of α-U Ce at 6.2GPa.

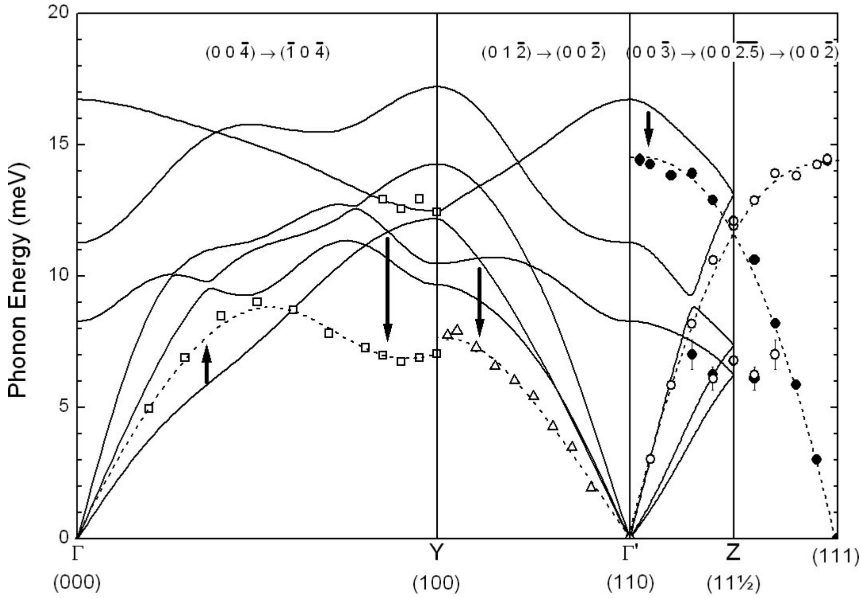

Figure 3: Selected phonon branches of Ce at 6.2 GPa measured by IXS (symbols) in comparison with results of preliminary first-principles phonon calculation (solid lines). Arrows highlight the unexpectedly soft phonon branches near the Y-point, and other deviations betwene theory and experiment.

References

[1] Smith & Lander, Phys. Rev. B **30**, 5407 (1984).
[2] Smith *et al*, Phys. Rev. Lett. **44**, 1612 (1980).
[3] McMahon & Nelmes, Chem. Soc. Rev. **35**, 943 (2006).
[4] Tonkov and Ponyatovsky, *Phase Transformations of Elements Under High Pressure* (CRC Press, Boca Raton, 2005).
[5] McMahon & Nelmes, Phys. Rev. Lett. **78**, 3884 (1997).