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:** **Phonon dispersion in damaged UO_2**	**Experiment number**: HC2508
Beamline: ID28	**Date of experiment**: from: 13/July to: 19/July/2016	**Date of report**:
Shifts: 18	**Local contact(s)**: L. Paolasini and A. Bosak	*Received at ESRF:*
Names and affiliations of applicants (* indicates experimentalists): ***S. Rennie (Univ. of Bristol, UK)** ***R. Springell (Univ. of Bristol, UK)** ***G. H. Lander (ITU, Karlsruhe)**		

Report:

Uranium dioxide has a key limitation as a reactor fuel; the thermal conductivity is compromised upon increased irradiation damage [1]. As the thermal conductivity falls, it is necessary to increase the reactor operating temperature in order to maintain a constant power output. This, combined with the degradation of structural integrity as a function of damage, are the key factors that limit the fuel lifetime. The predominant mechanism responsible for heat transfer in nuclear fuel is the phonons. It is therefore critical that we understand this mechanism, and how it is compromised as a function of irradiation damage. To investigate this, we proposed to measure the phonon dispersion curves and line widths of an irradiated and as-grown epitaxial thin film of UO_2, by grazing-incidence Inelastic X-ray Scattering (IXS) on ID28. Measurement of these line widths can be directly related to the anharmonicity, and thus provide information on the contribution of each phonon branch to the thermal conductivity.

The experimental data presented here are a result of a continuation experiment conducted on this subject. The initial measurements taken in July 2015 (**HC-1699**) were successful in obtaining phonon measurements in UO_2 thin films, however the measured phonons were weak, particularly at the zone boundaries. This was a result of a range of experimental limitations. With an incident angle of 0.2^{o} the majority of x-rays were being reflected from the surface, as the critical angle of UO_2 at 17.9 keV is ~ 0.3^{o} in two theta thus it is unsurprising that the phonons were exceedingly weak. This low incident angle also allowed for only 14 nm of the film depth to be probed and thus measurements are sensitive to surface defects. Additional experimental limitations included: non-uniformly damaged films and restricted access to phonon zones as a result of the sample mount, both of which also contributed to obtaining limited experimental data.

Despite this, the initial measurements were promising and we were granted further experimental time in July 2016 (HC-2508). In this experiment significant improvements were made. Firstly, uniform damage profiles were achieved through raster scanning the ion beam across the film surface. Secondly, the sample mount was developed such that a wider range of reciprocal space directions could be accessed. However perhaps the most significant improvement was in the development of the experimental technique used to measure phonons in thin films. While restricted to an incident angle of 0.2^{0}, an additional tilt of 1^{0} was added such that we moved away from the critical angle of UO_2. This allowed for distinct improvements in the phonon measurements, and resulted in a full exploration of the (220) and (400) positions along the transverse and longitudinal directions.

Experimental Results

Prior to the experiment, single crystal UO_2 [001], 0.5 μm thin films were grown on $SrTiO_3$ [001] at the University of Bristol via DC magnetron sputtering. Part of the film was kept to represent a pristine sample. To induce the damage, irradiation experiments were conducted at the Dalton Cumbia Facility, UK using 2.1 MeV He^{2+} ions. The ion beam was raster scanned across the sample to achieve a uniform damage profile with an average number of displacements per atom (dpa) of 0.15. In agreement with the previous IXS experiment, this level of damage was selected as it produced an observable shift in the UO_2 XRD Bragg peaks providing confidence that a change had occurred within the lattice.

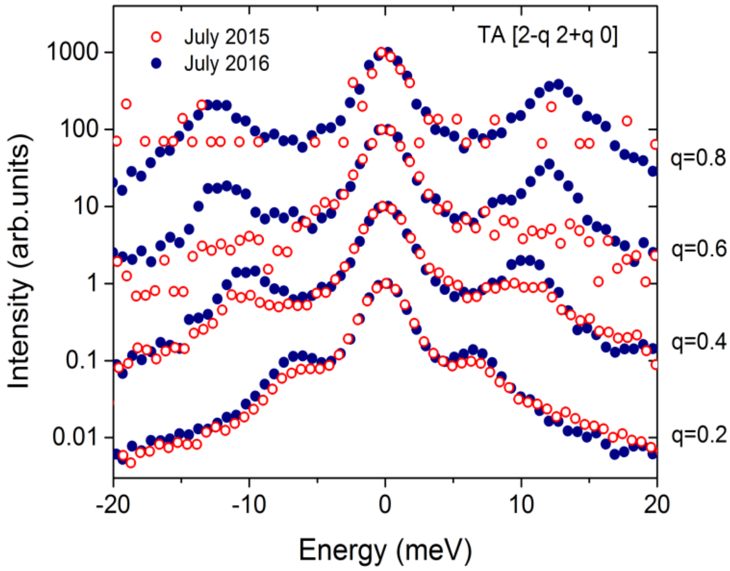

Figure 1: A comparisong of the transverse accoustic phonons measured from the (220) position for q values 0.2, 0.4, 0.6 and 0.8 from experimental runs conducted in July 2015 (red) and July 2016 (blue).

Figure 1, shows a comparison of the data acquired during the first and second experimental run. There is an increase in the signal/noise ratio between these two efforts. In addition the time taken to measure a phonon was also much shorter, so that the overall improvement between 2015 and 2016 was on average ten fold. However this improvement was increasingly more significant for the measurements observed at larger q values. As demonstrated in Fig. 2 for q = 0.8 it was not possible to observe a phonon in this position in July 2015, however a strong phonon signal was observed in 2016.

It is clear that the developments made in sample preparation and experimental technique have led to marked improvements in observing phonons in UO_2 thin films. However, unfortunately, the result of this improvement showed that the acoustic phonons are not measurably altered by 0.15 dpa of damage with 2.1 MeV He^{2+} ions (Fig. 2), despite the alteraction of the lattice parameter of the films, as observed with XRD.

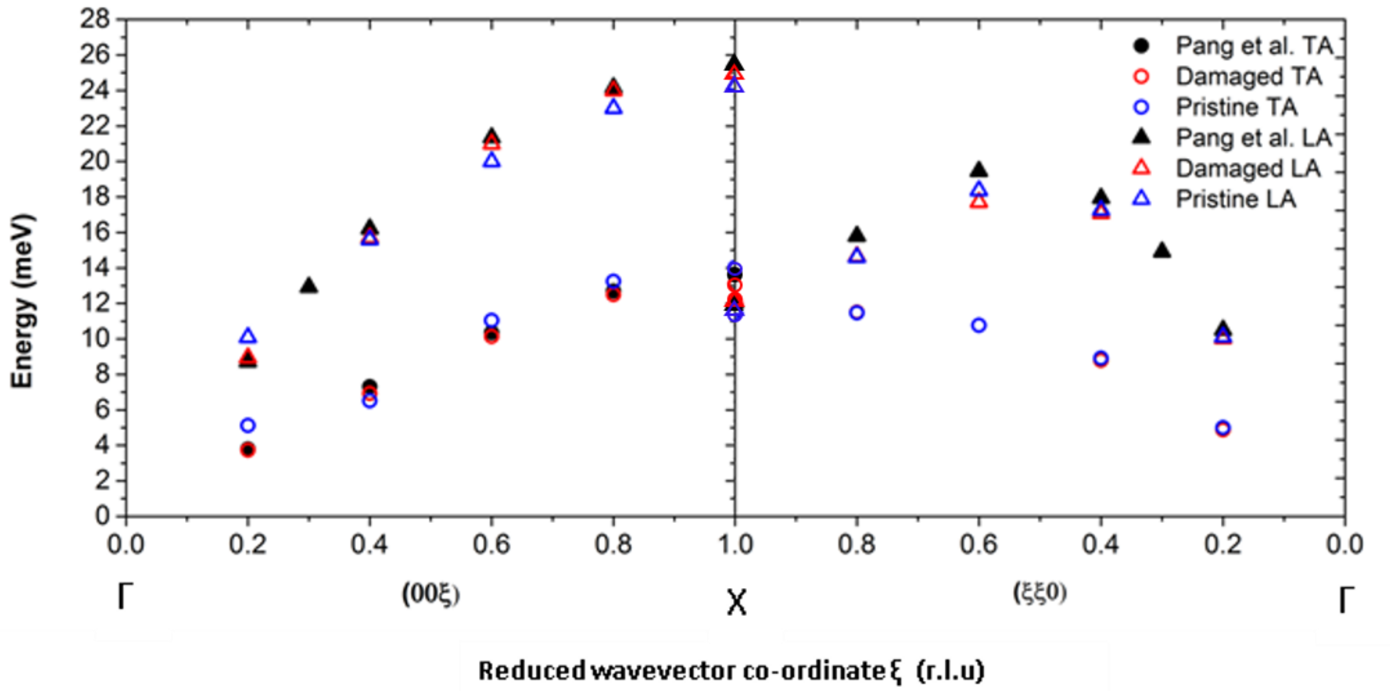

Figure 2: The transverse and longitudinal phonons of UO_2 along the (00ξ) and (ξξ0) directions as measured via IXS for pristine (blue) and damaged (red) thin films, in comparison with bulk data obtained by Pang et. al (black) via INS.

Figure 2 shows the experimental data obtained here, compared with that collected by Pang et. al. for bulk UO_2 measured via inelastic neutron scattering (INS) [2]. As shown, the observed phonon energies are in agreement with existing neutron data, however no clear trend can be depicted between the damaged and pristine measurements. We therefore believe that a greater level of damage is required to alter the phonon dispersion of UO_2.

Conclusions

The transverse and longitudinal accoustic phonons were measured for both a pristine and damaged UO_2 thin film along the (00ξ) and (ξξ0) directions via inelastic x-ray scattering at beamline ID28, ESRF. These measurements were a continuation of a previous experimental run (**HC-1699**), upon which significant advances have been made to develop a technique which allows for successful exploration of the phonon dispersion of thin film samples. Unfortunately the result of this improved experimental technique showed that the acoustic phonons are not measurably altered by 0.15 dpa of damage with 2.1 MeV He^{2+} ions, despite the alteraction of the lattice parameter of the films, as observed with XRD.

In spite of this, alterations in the phonon dspersion is predicted by point defect calculations currently being carried out by our colaborators B. Dorado *et al*. of the CEA [3], which are the first phonon calculations for damaged UO_2. Therefore while 0.15 dpa of damage with 2.1 MeV He^{2+} ions was not sufficient to induce such a change, we are hopeful that extended irradiation with a heavier ion such as Xe would stimulate a stronger damage response, measurable in the phonon dispersion.

References

[1] C. Ronchi *et al.,* J. Nucl. Matls. **327**, 58 (2004).
[2] J. W. L. Pang *et al.*, Phys. Rev. Lett. **110** 157401 (2013); *ibid* Phys. Rev. B **89**, 115132 (2014).
[3] B. Dorado, and P. Garcia, Phys. Rev. B **87**, 195139 (2013), B. Dorado *et al. ibid* Phys. Rev. B **86**, 035110 (2012).