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Deadlines for submission of Experimental Reports

- 1st March for experiments carried out up until June of the previous year;
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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.

	Experiment title: Phonon dispersion in damaged UO_2	Experiment number: HC-1699
Beamline:	Date of experiment: from: 15/07/2015 to: 21/07/2015	Date of report: 08/09/2015
Shifts: 18	Local contact(s): Luigi Paolasini and Alexsei Bosak	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

Sophie Rennie[1*], Ross Springell[1*], James Darnbrough[1*], Gerrard Lander[2*]

[1]Interface Analysis Centre, School of Physics, HH Wills Physics Laboratory, University of Bristol, Tyndall Avenue, Bristol, BS8 1TL, UK.
[2]Laboratory European Commission Joint Research Centre Institute for Transuranium Elements Postfach 2340 DE - 76125 KARLSRUHE

Report:

Outline

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. Here we present the initial results, which show the transverse acoustic phonon measured along the [ζζ0] direction to be both softer and broader with irradiation damage. While, first principal calculations of the phonon properties of UO_2 exist [3], there are no calculations for damaged material. We are collaborating with B. Dorado *et al*. of the CEA [4], where such calculations are in progress. Through comparison with our experimental results, this combined study has the potential to validate the applied modelling approach and thus support an improved understanding of the long term behaviour of nuclear reactor fuel upon irradiation.

Experimental

Prior to the experiment, single crystal UO_2 [001], 0.5 µm thin films were grown on $SrTiO_2$ [001] at the University of Bristol via DC magnetron sputtering. Damage was induced on one half of each sample via irradiation with 2.1 MeV He^{2+} ions at the Dalton Cumbia Facility, UK to achieve an average number of displacements per atom (dpa) of 0.15. This dpa 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 and as such would likely alter the phonon spectra.

In order to map out the phonon dispersion curve and measure phonon line widths, grazing incident inelastic x-ray scattering was performed on both the irradiated and pristine section of the UO_2 thin film in a horizontal scattering geometry at 17.794 keV. In this geometry the incident angle of the beam is 0.2^0, probing a sample depth of 140 Å such that no contribution from the substrate was observed. Transverse and longitudinal acoustic phonons were successfully measured at the (220) position along the [-ζζ0] and [ζζ0] directions. The observed phonon energies for the pristine sample were in agreement with existing neutron data [2], however the corresponding TA phonon measurements for the damaged region were consistently found to be at lower energy positions, interestingly no such changes were observed for the LA branch.

Due to the weak nature of the phonons, phonon line widths could only be obtained with significantly enhanced counting statistics. This was achieved at two q points, [2.3 2.3 0] and [1.8 2.2 0], along the LA and TA branches respectively. Figure 1 shows a comparison of these q points between the damaged and pristine region of the sample. These data were fitted using fit28 software, modelling the phonons with a damped harmonic oscillator and convoluting this with the measured analyser resolution function. The analyser resolution was measured to 3 meV, the values given for the phonon width in Figure 1 are the deconvoluted widths. As can be seen for the TA measurement taken in the damaged region (Fig. 1b), the phonon position and width are shifted from the pristine values (Fig. 1a). Interestingly, this shift is not observed for the LA measurement (Fig.1c & 1d), where the width and position of the phonon appear unaffected by the induced damage. These initial results therefore hint that the TA and LA branches respond differently to He^{2+} ion irradiation. While this is an interesting result, further measurements are required of additional off specular positions in order to support this conclusion.

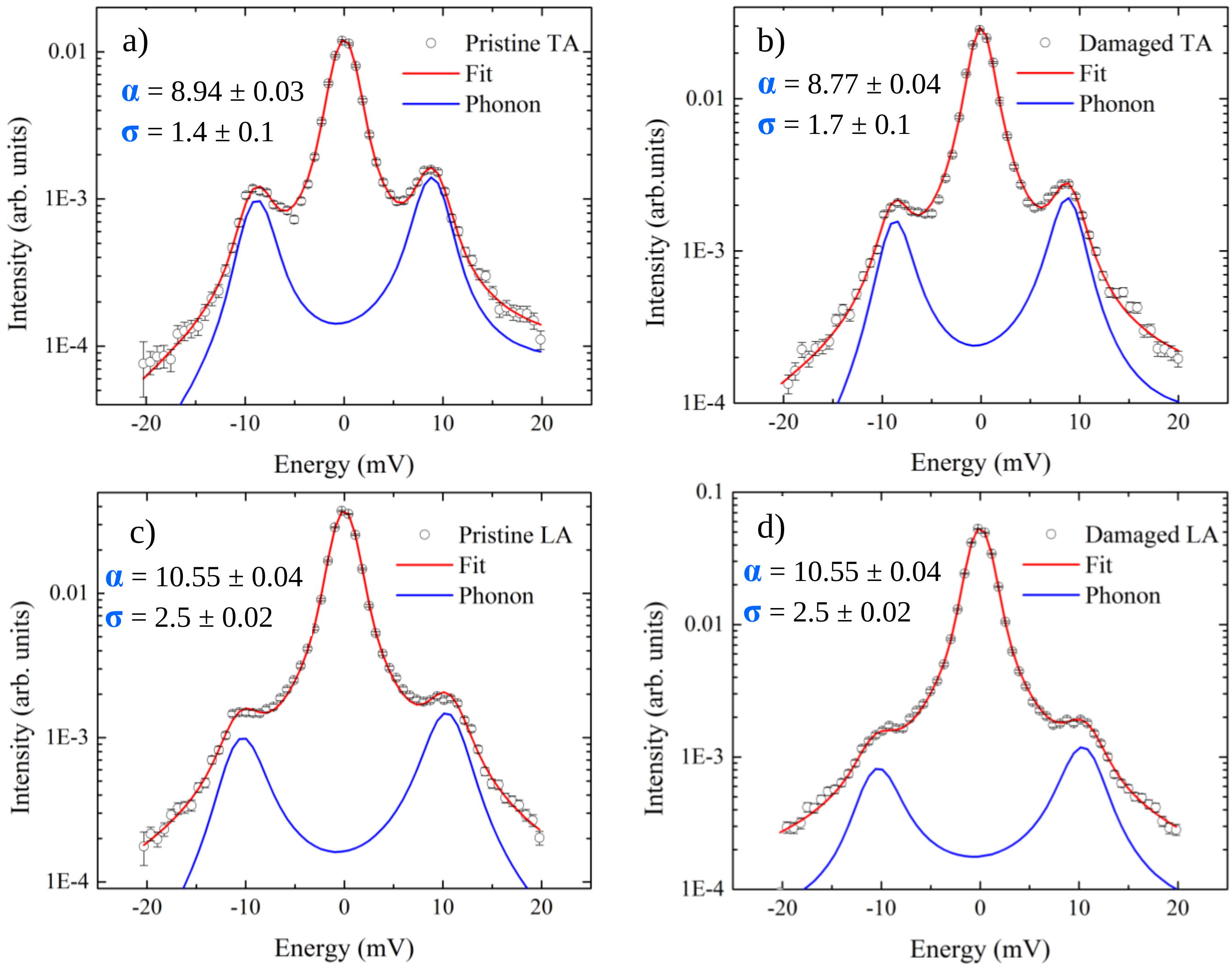

Figure 1: The transverse accoustic phonon at the (1.7 2.3 0) position (a,b) and longitudinal accoustic phonon at the (2.2 2.2 0) position (c,d) for a UO_2 thin film in the pristine and damaged regions respectively, where σ is the phonon width deconvoluted from the analyser resolution and α is the phonon energy position.

Conclusion

These initial results pave the way for a more complete study of the degradation of phonon transport upon fuel irradiation. Initial results indicate that there may be a difference between the stability of the longitudinal and transverse branch on exposure to He^{2+} irradiation. However, thus far, phonon measurements have only been performed at the (220) position along the [ζζ0] and [-ζζ0] directions, therefore in order to to give confidence in this experimental result it is necessary to compare the phonon positions and line widths for the longitudinal and transverse acoustic phonons for additional off specular directions. With further measurements, these results will be instrumental in validating modelling approaches and may pave the way to a more complete understanding of the microscopic origins of the degradation in the thermal conductivity of uranium oxide nuclear fuel with radiation damage.

References

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[3] M. Sanati, R. C. Albers, T. Lookman, and A. Saxena, Phys. Rev. B **84**, 014116 (2011)
[4] B. Dorado, and P. Garcia, Phys. Rev. B **87**, 195139 (2013), B. Dorado *et al. ibid* Phys. Rev. B **86**, 035110 (2012)