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Engineering and Physical Sciences Research Council

Characterisation of Irradiation Damage Using X-Ray Diffraction Line Profile Analysis

Royce Training: Ion Beam Irradiation and Characterisation - Best Practice | Mar 2023

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Motivation

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Environment → **Microstructure** → **Behaviour**

Environment:
- Fast neutron (charged particle) irradiation
- Elevated temperature

→ Microstructure:
- Irradiation damage <a> and <c> loops

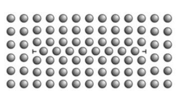
→ Behaviour:
- Radiation strengthening and ductility loss
- Plastic strain localisation (dislocation channelling)
- Dilatation – irradiation growth and creep

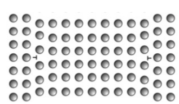
At doses ≲ 5 dpa:

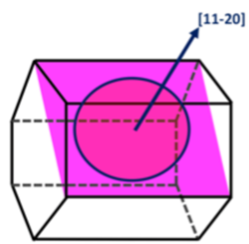
<a>-loops

1st and 2nd order prismatic planes

Interstitial and vacancy





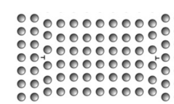


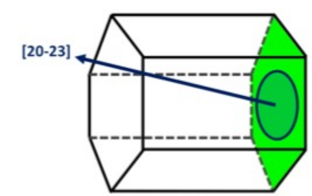
At higher doses:

<c>-loops

Basal planes

Vacancy





MIDAS Methodologies

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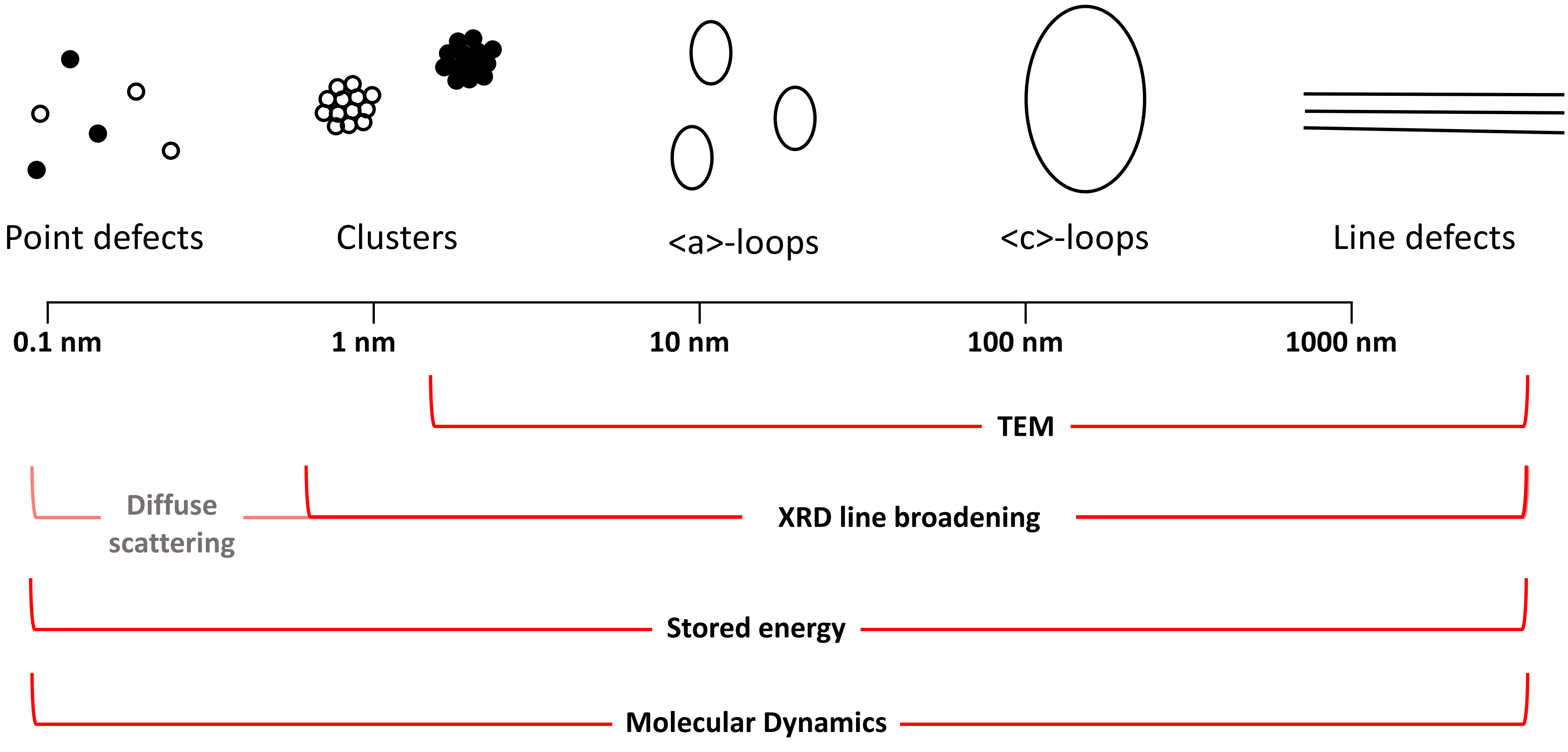


MIDAS Methodologies

TEM

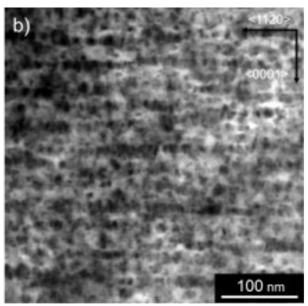
Small volume investigated
Loop size, shape and nature
Only see loops >2 nm
Messy at high doses\small loops
Sample preparation tricky

+

XRD

Large volume measured
Dislocation density, loop type
Captures effect of all loops
Accurate at any dose
Easier sample preparation

→ Complementary

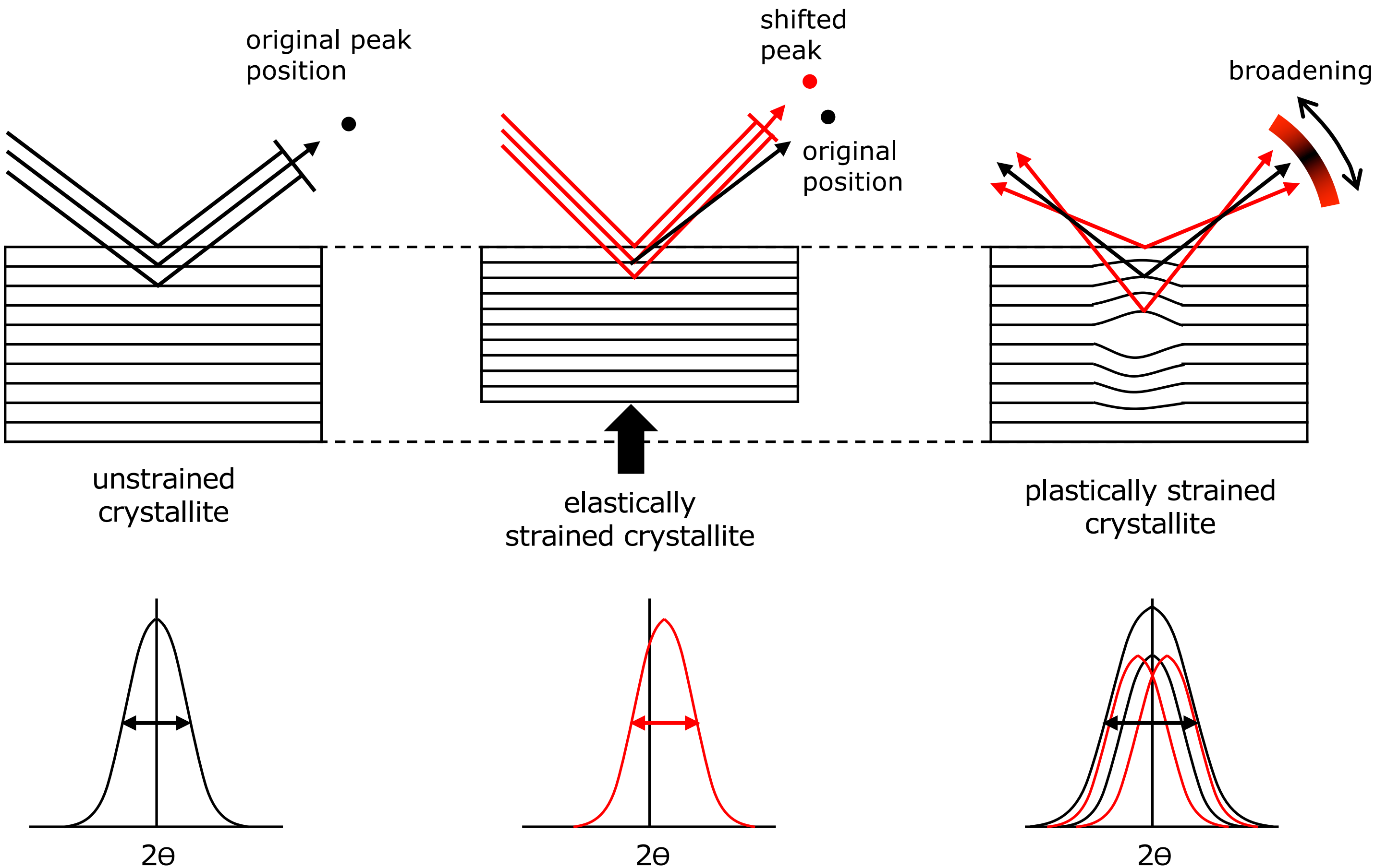


A. Harte et al., The effect of matrix chemistry on dislocation evolution in an irradiated Zr alloy, Acta Mat., 130 (2017) 69-82

	'Bulk' XRD	**Microbeam synchrotron XRD**	**3D Synchrotron XRD**
Resolution	Sample average	Depth resolved [~2um]	Grain resolved
Preparation time	~1 hrs	~2 hrs	~8 hrs
Measurement time	~16 hrs	~1 hr	~8 hrs
Analysis time	~10 mins	~days	~weeks

→ More complex experiments, more information

Line broadening



Line profile analysis

$$A^{D}_{hkl}(L) \cong \exp[-2\pi^2 g^2 L^2 \langle \varepsilon^2_{g,L} \rangle],$$
$$\langle \varepsilon^2_{g,L} \rangle = \rho \frac{Cb^2 4}{\pi} f(\eta),$$

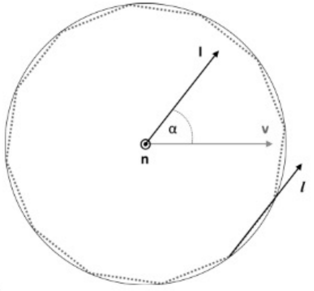
Measured pattern is a convolution of different effects:

Non-monochromatic source, $K\alpha_1 \backslash K\alpha_2$, beam divergence, detector

Dislocations, loops, coherent precipitates

$$I_{Measured} = I_{Instrument} * I_{Size} * I_{Strain} + I_{Background}$$

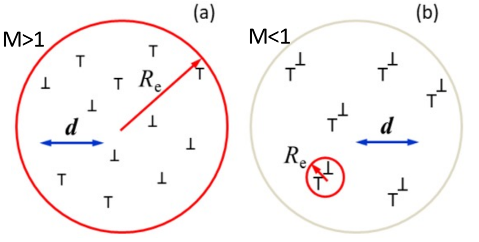
If crystallite size < 1 μm



L. Balogh et al., Contrast factors of irradiation-induced dislocation loops in hexagonal materials, Journal of Applied Crystallography, 49 (2016) 2184-2200

Physical parameters:

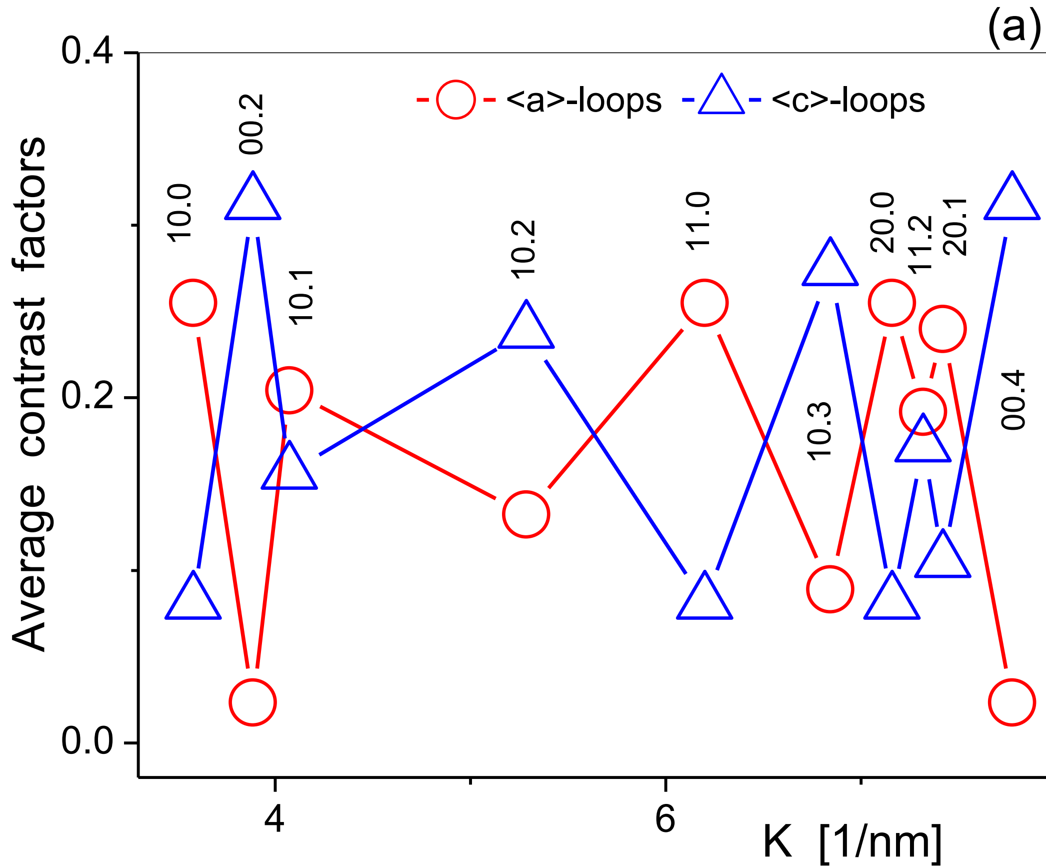
- a1, a2: Contrast factors
- b, c: Size broadening (median, sigma)
- d: Dislocation density
- e: Dislocation arrangement



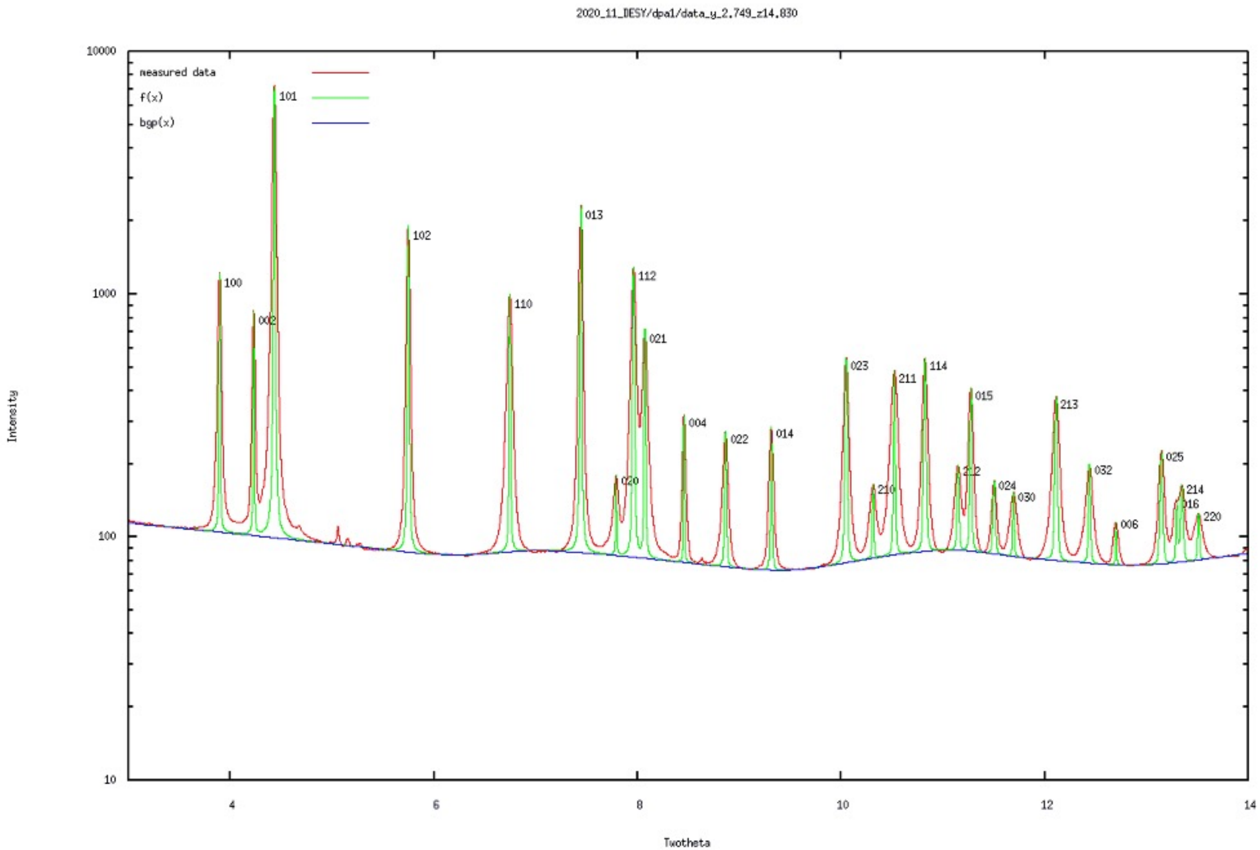
G. Ribárik et al., The Convolutional Multiple Whole Profile (CMWP) Fitting Method, a Global Optimization Procedure for Microstructure Determination, Crystals, 10(7)623

Contrast factor

- Describes how visible a given dislocation structure is on a given reflection (in an analogous way to TEM)
- <a>-loops and <c>-loops have a very different contrast factor
- Can use this fact to get a partial dislocation density and arrangement parameter



MIDAS CMWP Procedure



Start with initial guess

- Physical (Monte Carlo)

↓

- Position (Levenberg–Marquardt)

↓

- Intensity (Levenberg–Marquardt)

↓

- Physical (Levenberg–Marquardt)

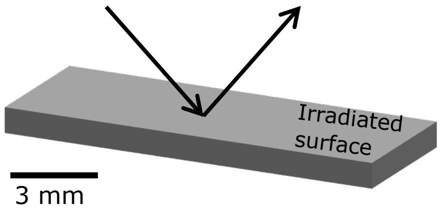
↓

- Background (Levenberg–Marquardt)

↓

- All (Levenberg–Marquardt)

Bulk measurement

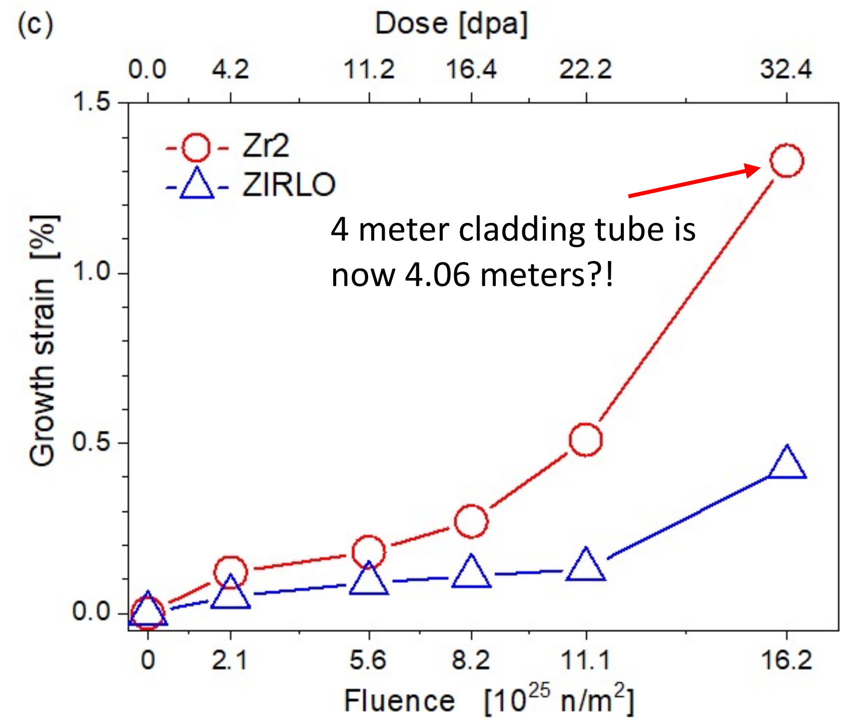


Sample resolved

BOR-60 Samples

- Zr-2 RXA & Low-Sn ZIRLO RXA

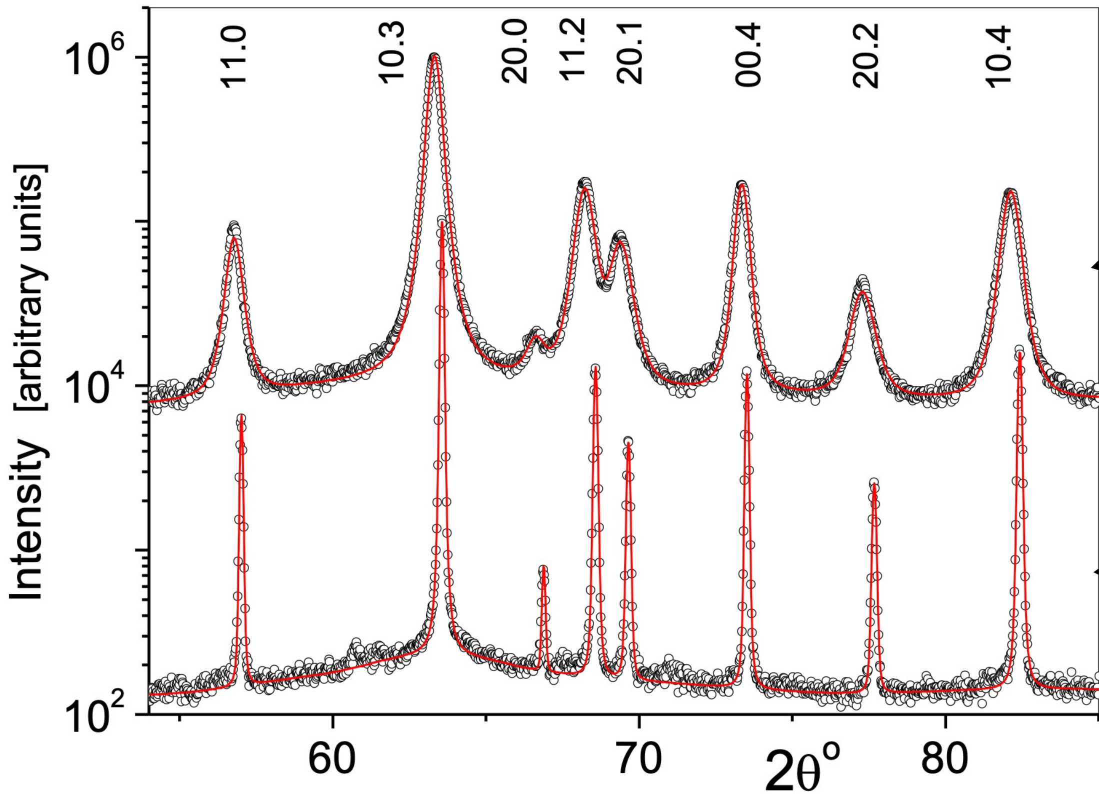
- Neutron irradiated in BOR-60 reactor at to 2.1, 5.6. 8.2, 11.1 & 16.2 × 10^{25} n/m^2 at 320 °C



Aim: Correlate growth behaviour with dislocation loop density

Yagnik, S. et al.., "Effect of Alloying Elements, Cold Work, and Hydrogen on the Irradiation-Induced Growth Behavior of Zirconium Alloy Variants," Zirconium in the Nuclear Industry: 18th International Symposium, ASTM STP1597, R. J. Comstock and A. T. Motta, Eds., ASTM International, West Conshohocken, PA, 2018, pp. 748–795

MIDAS Diffraction patterns



Zircaloy-2 RXA irradiated with neutrons in the BOR-60 reactor

16.2×10^{25} n/m^2 (32.4 dpa)

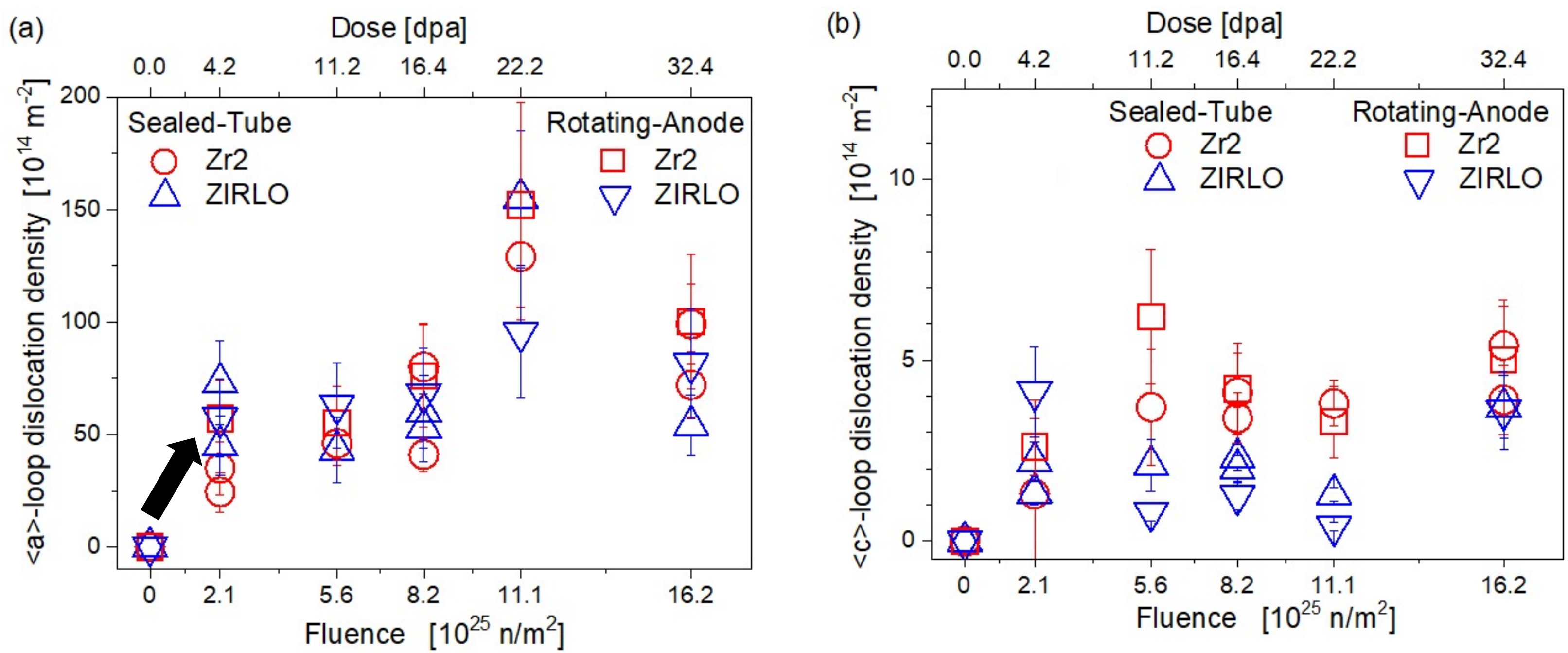
$\rho_{<a>} = 49 \times 10^{14}\ m^{-2}$

$\rho_{<c>} = 3.6 \times 10^{14}\ m^{-2}$

Non-irradiated

$\rho_{total} = < 1 \times 10^{14}\ m^{-2}$

MIDAS Dislocation density



- <a>-loop density saturates at lowest dose
- <c>-loop density increases faster for Zr-2 compared to low-Sn ZIRLO

Transmission Electron Microscopy

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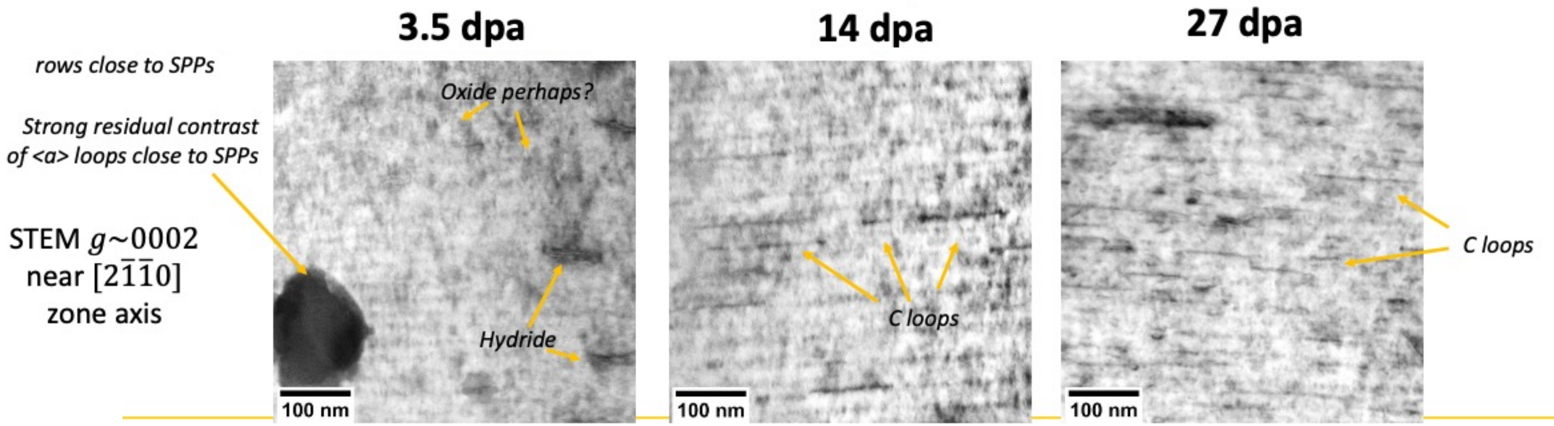
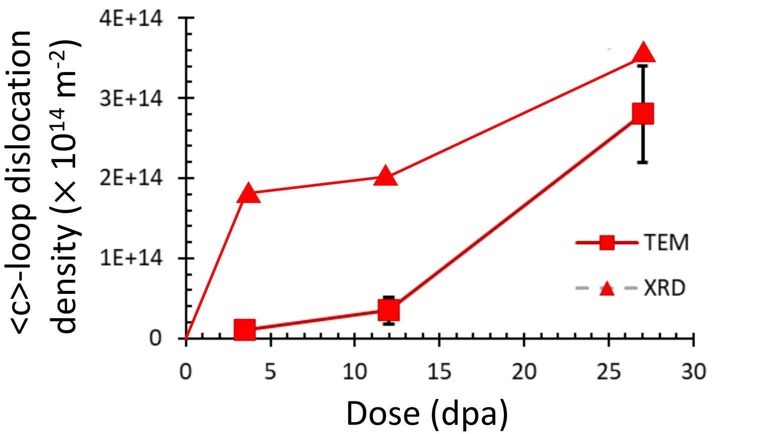
Zircaloy-2





- <c>-loop present at lowest dose, but very low density

Jack Haley, University of Oxford

MIDAS <c>-loop density vs growth strain

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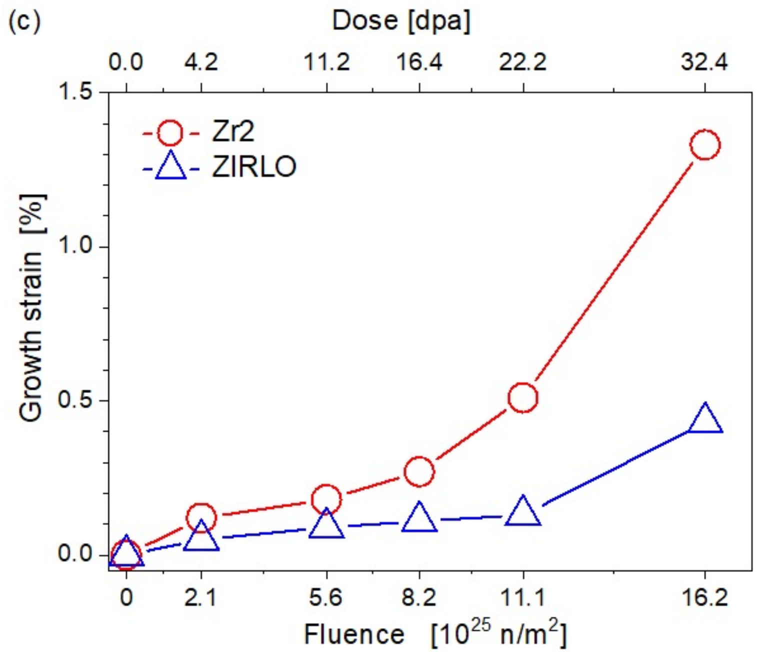
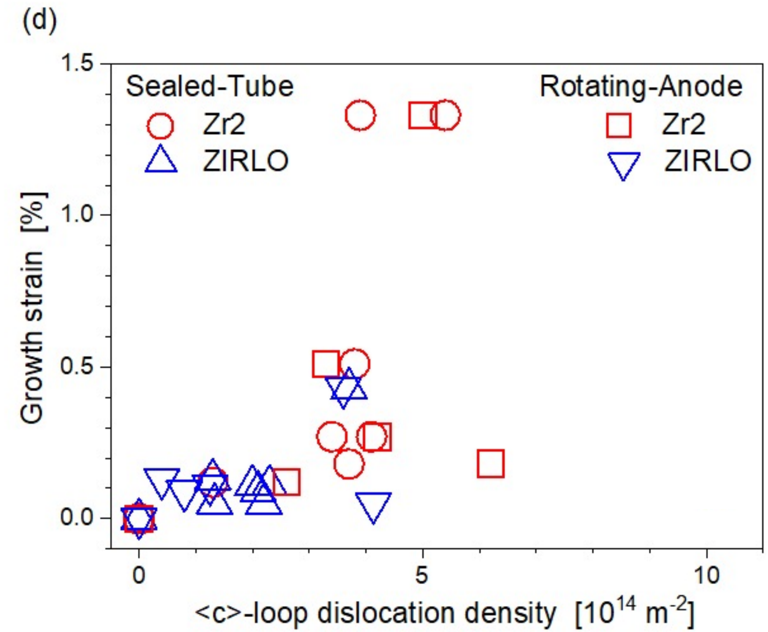
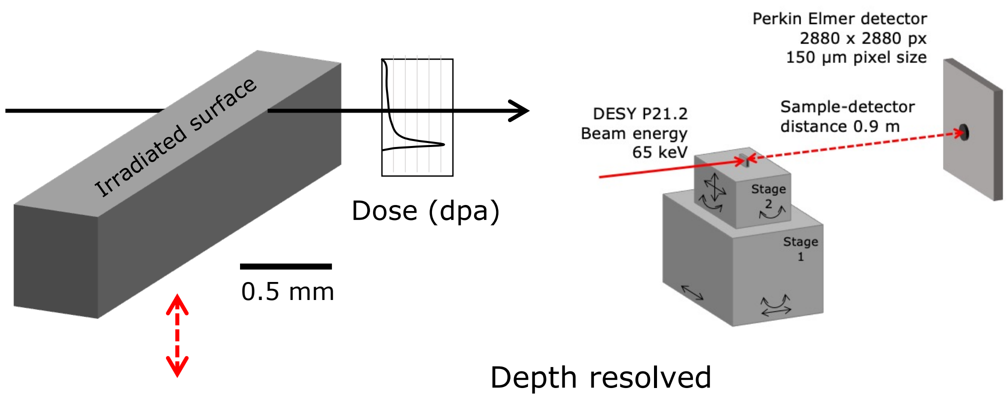




- Improved axial growth for low-Sn ZIRLO compared to Zr-2
- Weak correlation of <c>-loop density to axial growth strain
- Increase in growth at $\rho_{<c>} \approx 2 \times 10^{14}$ m^{-2}

Depth profiling with microbeam synchrotron XRD



MIDAS Proton irradiation

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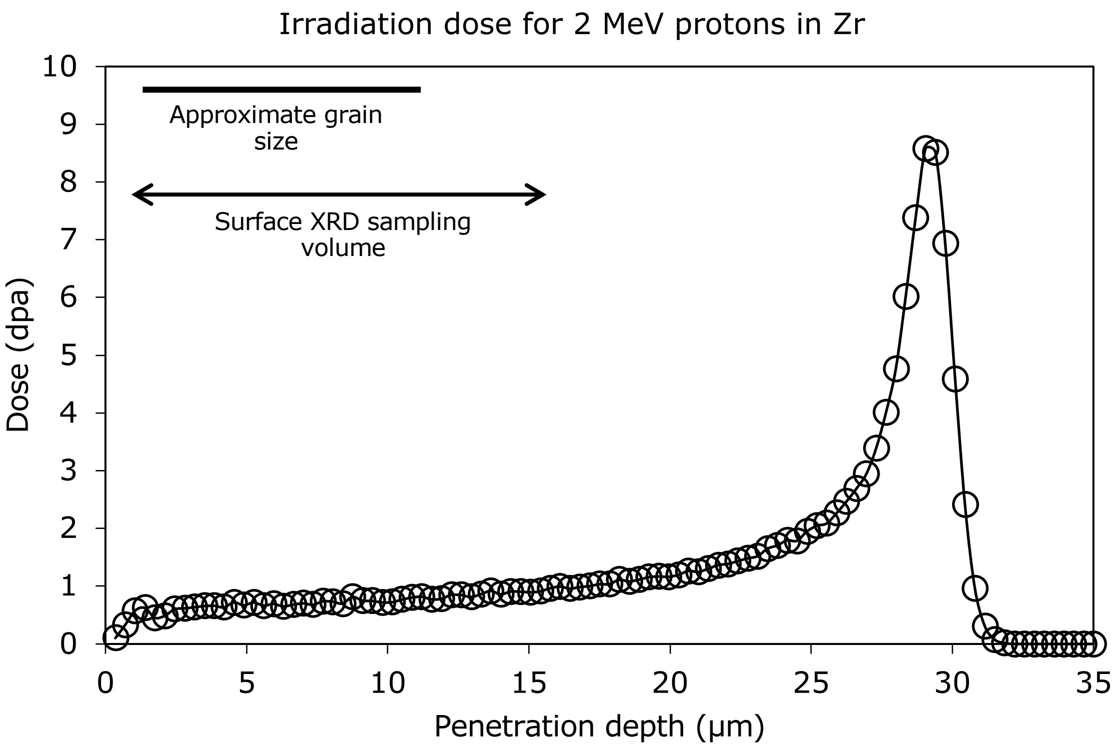
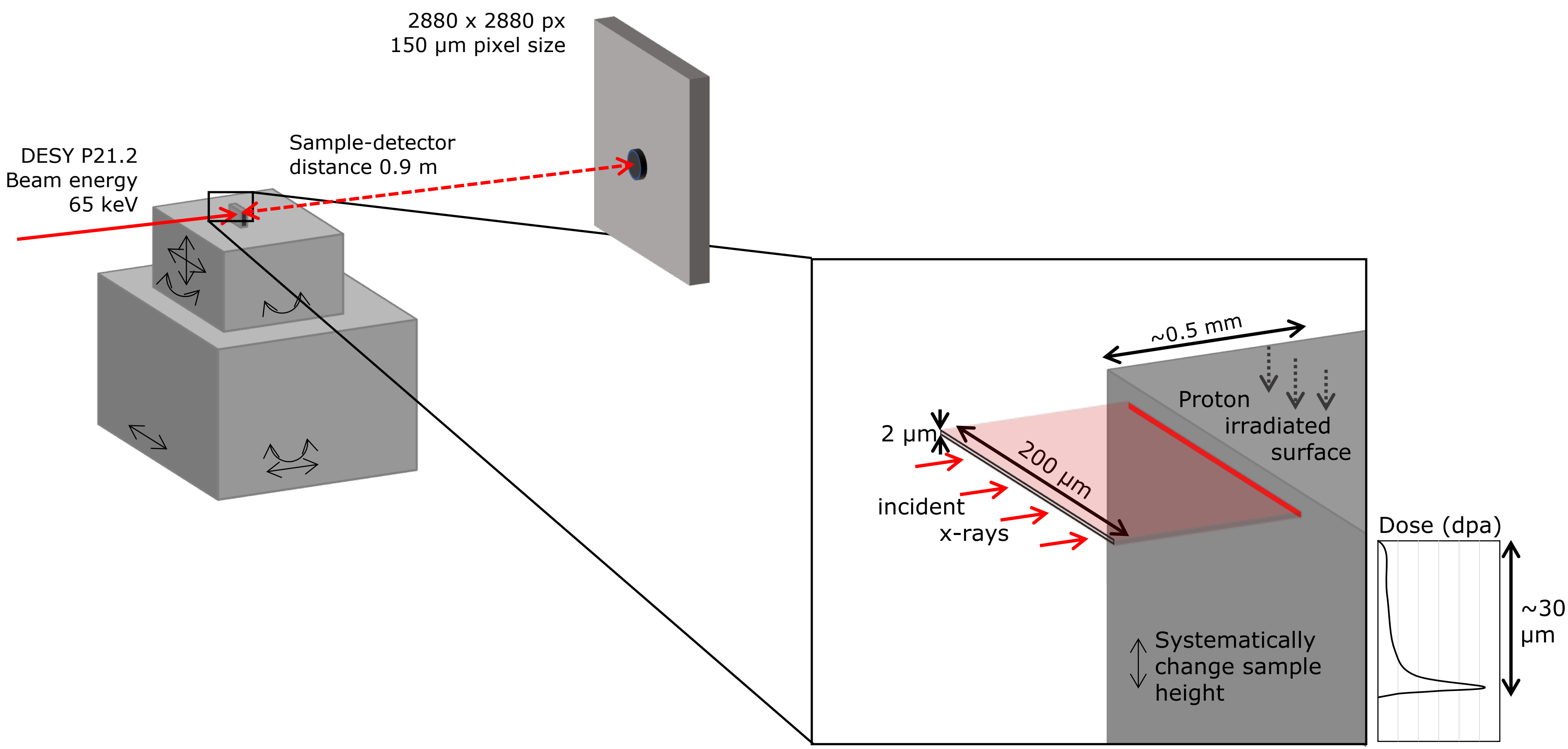


Aim: Measure dislocation density as a function of predicted dose

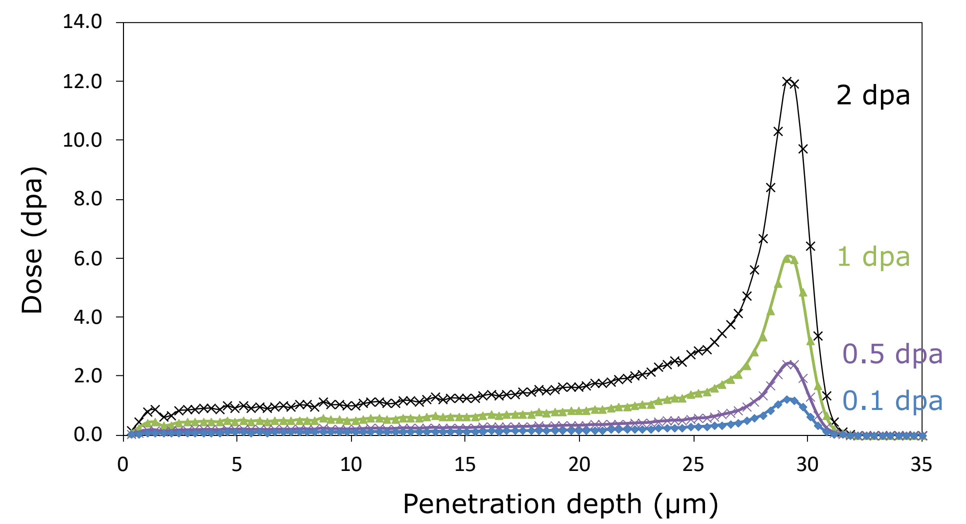
MANCHESTER 1824 The University of Manchester
Imperial College London
UNIVERSITY OF OXFORD
CCFE CULHAM CENTRE FOR FUSION ENERGY
NATIONAL NUCLEAR LABORATORY

Synchrotron XRD



Material

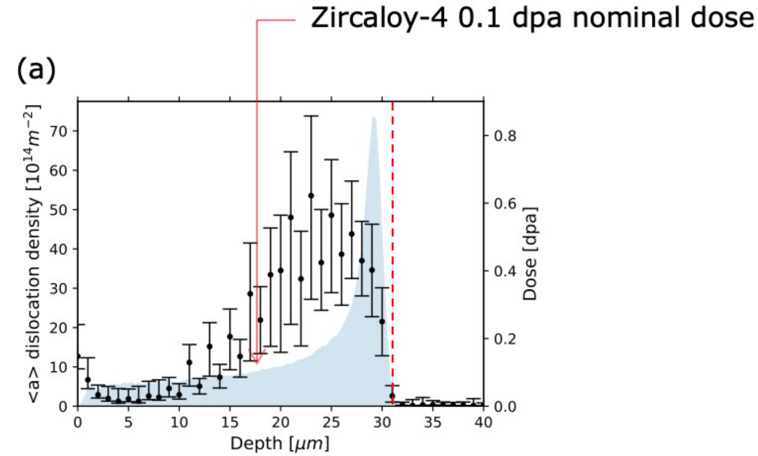
	Dose (DPA)	Temperature (°C)	Dose rate (dpa s^{-1})
Zircaloy-4	0.15	350	7.2×10^{-6}
Zircaloy-4	0.5	350	1.7×10^{-5}
Zircaloy-4	1.0	350	1.3×10^{-5}
Zircaloy-4	2.0	350	1.7×10^{-5}

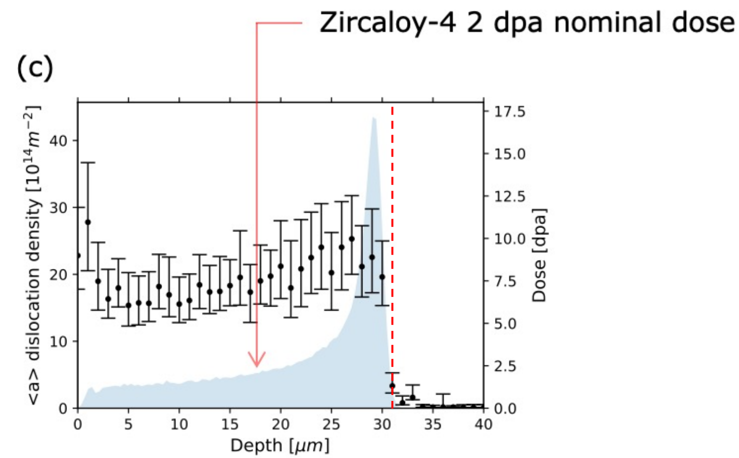


- Initial dislocation density <1 × 10^{14} m^{-2}

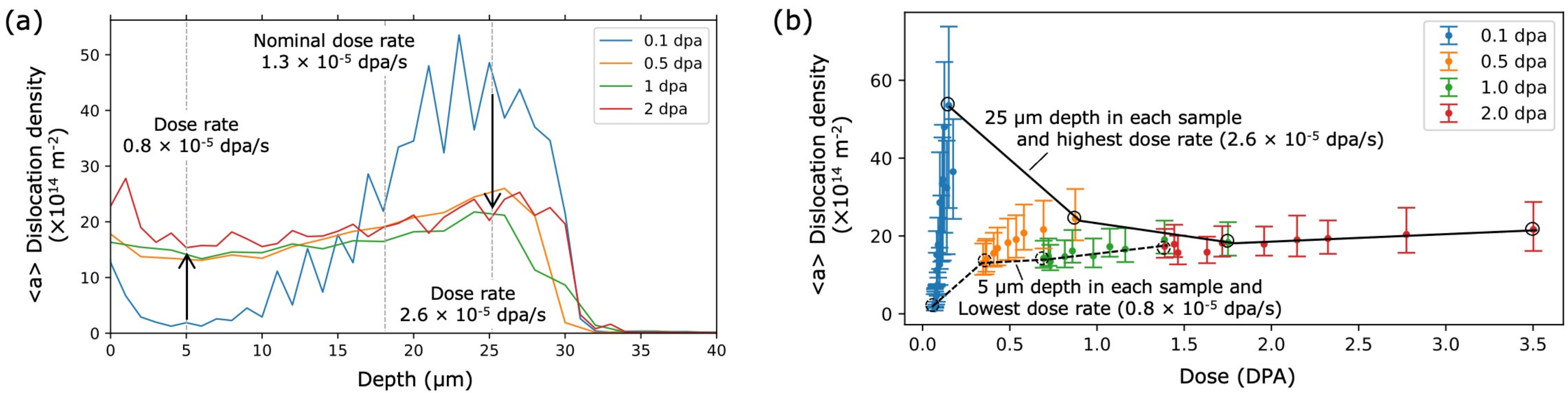
MIDAS Depth dependance

- SRIM range prediction accurate
- Varying dislocation density with depth for lowest dose (0.1 dpa)
- Saturation at higher doses (above 0.5 dpa)
- Increasing dislocation arrangement parameter with dose

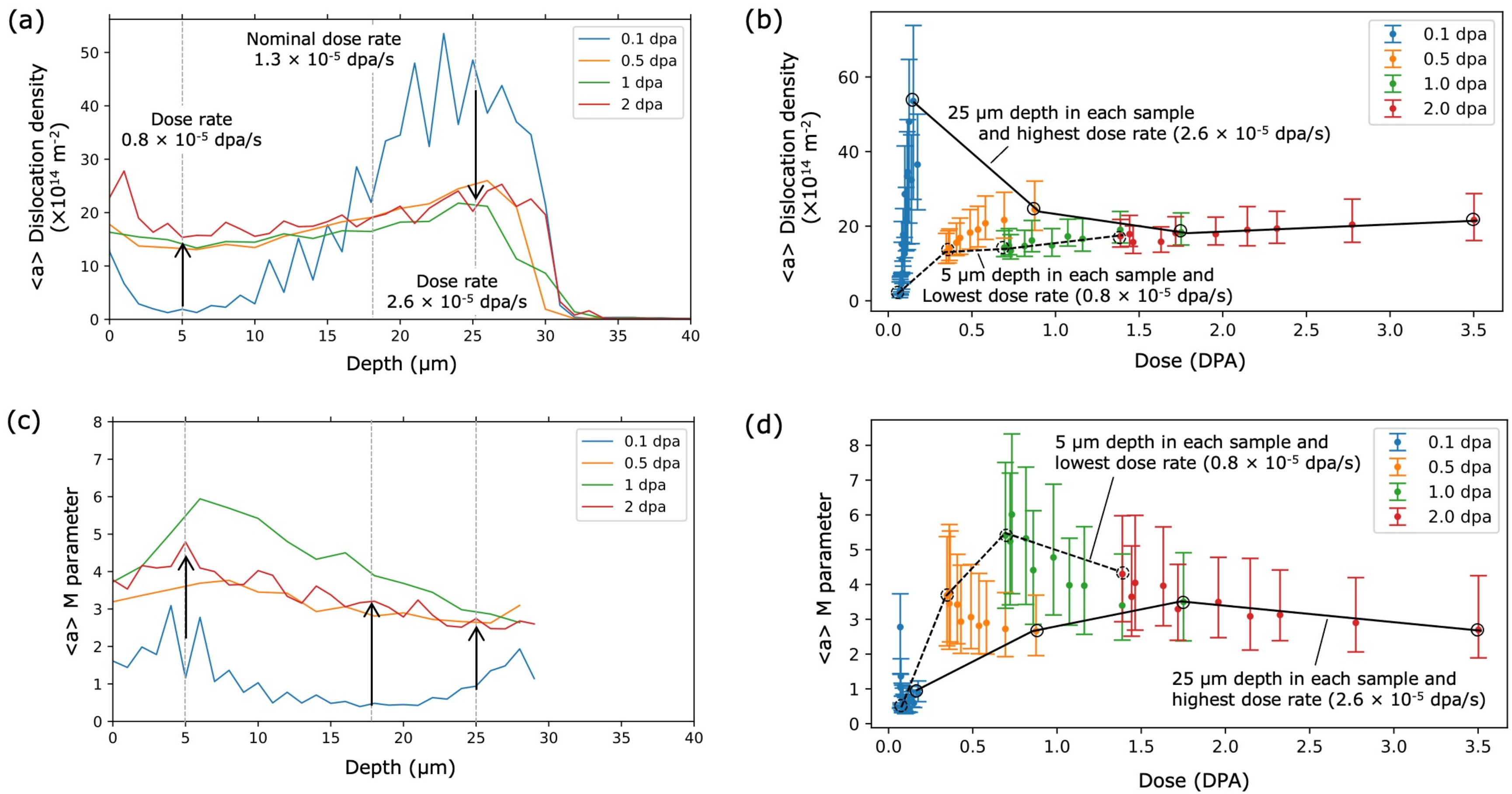




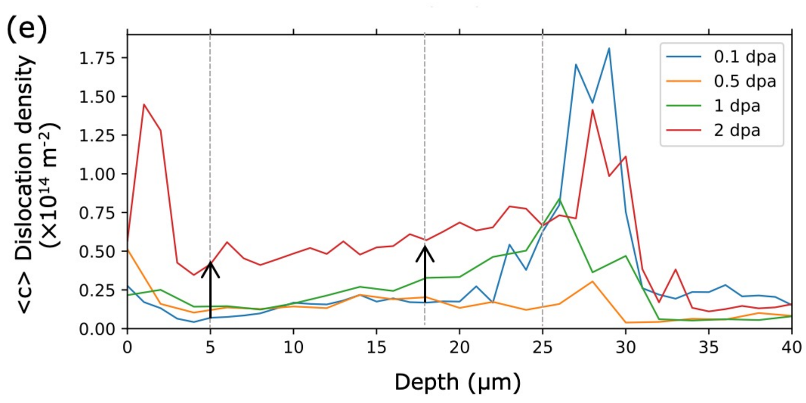
MIDAS Correlation with dose

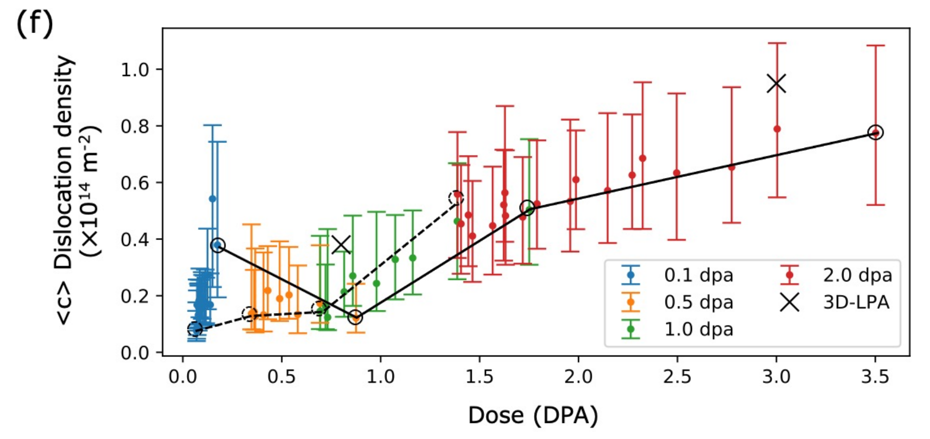


Correlation with dose



MIDAS Correlation with dose





Grain resolved synchrotron XRD

MIDAS Experimental schematic

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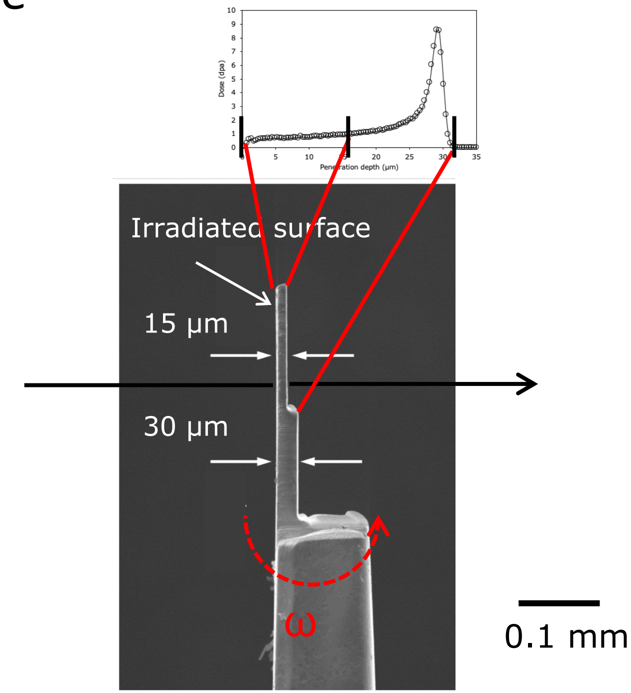
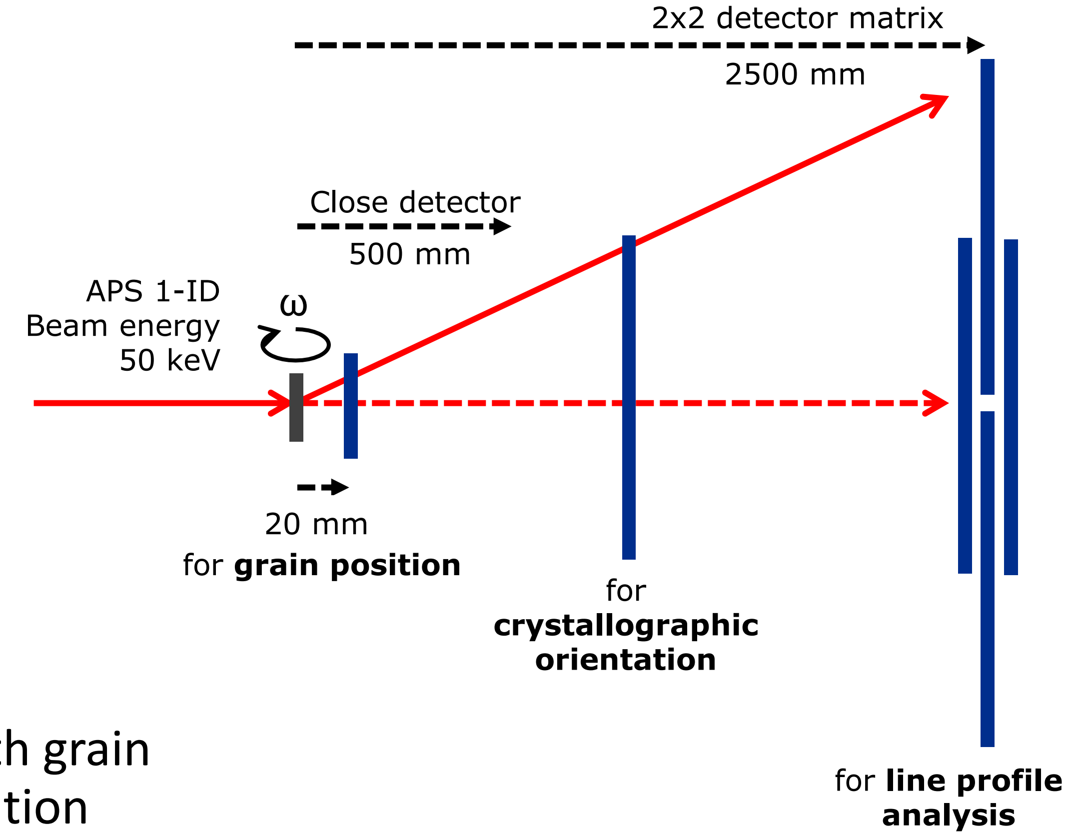
Zr-0.1Fe





Aim: Correlate dislocation density with grain position, size, crystallographic orientation

Dislocation density

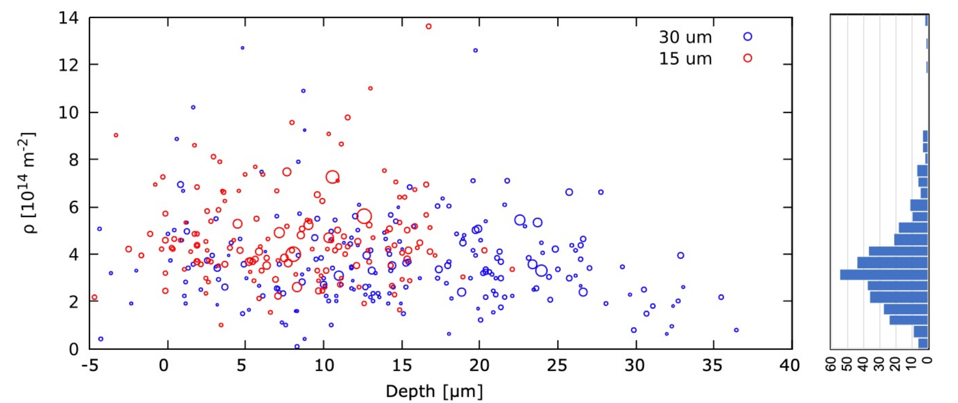
- Dislocation density per grain as a function of depth shows grain-to-grain variability in dislocation density

- 0.8 dpa = $4.2 \pm 2.0 \times 10^{14}$ m^{-2}
- 3 dpa = $3.2 \pm 1.8 \times 10^{14}$ m^{-2}

- Marker size = grain volume ^ 1/3

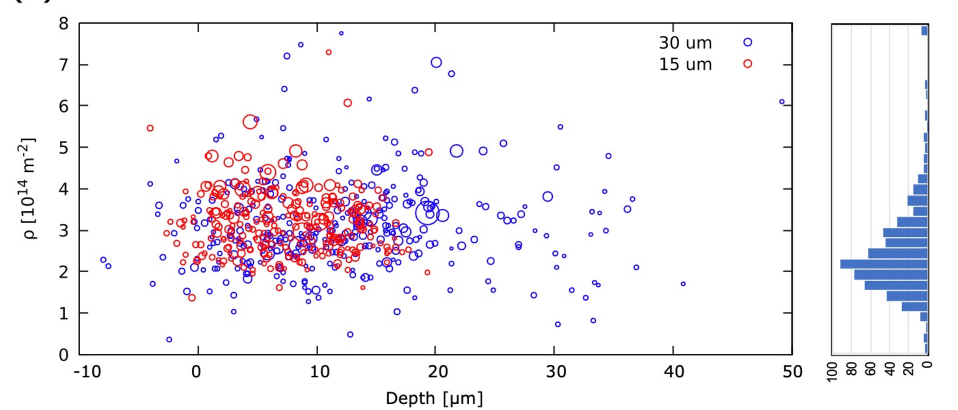
(a)

0.8 dpa



(b)

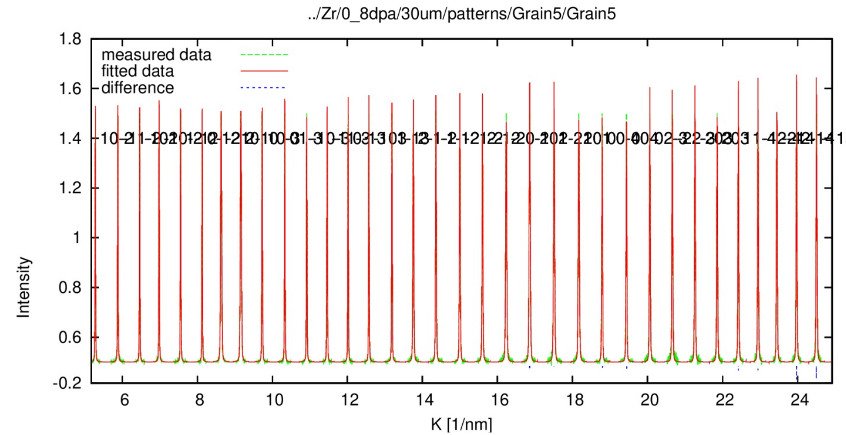
3 dpa



MIDAS Loop types

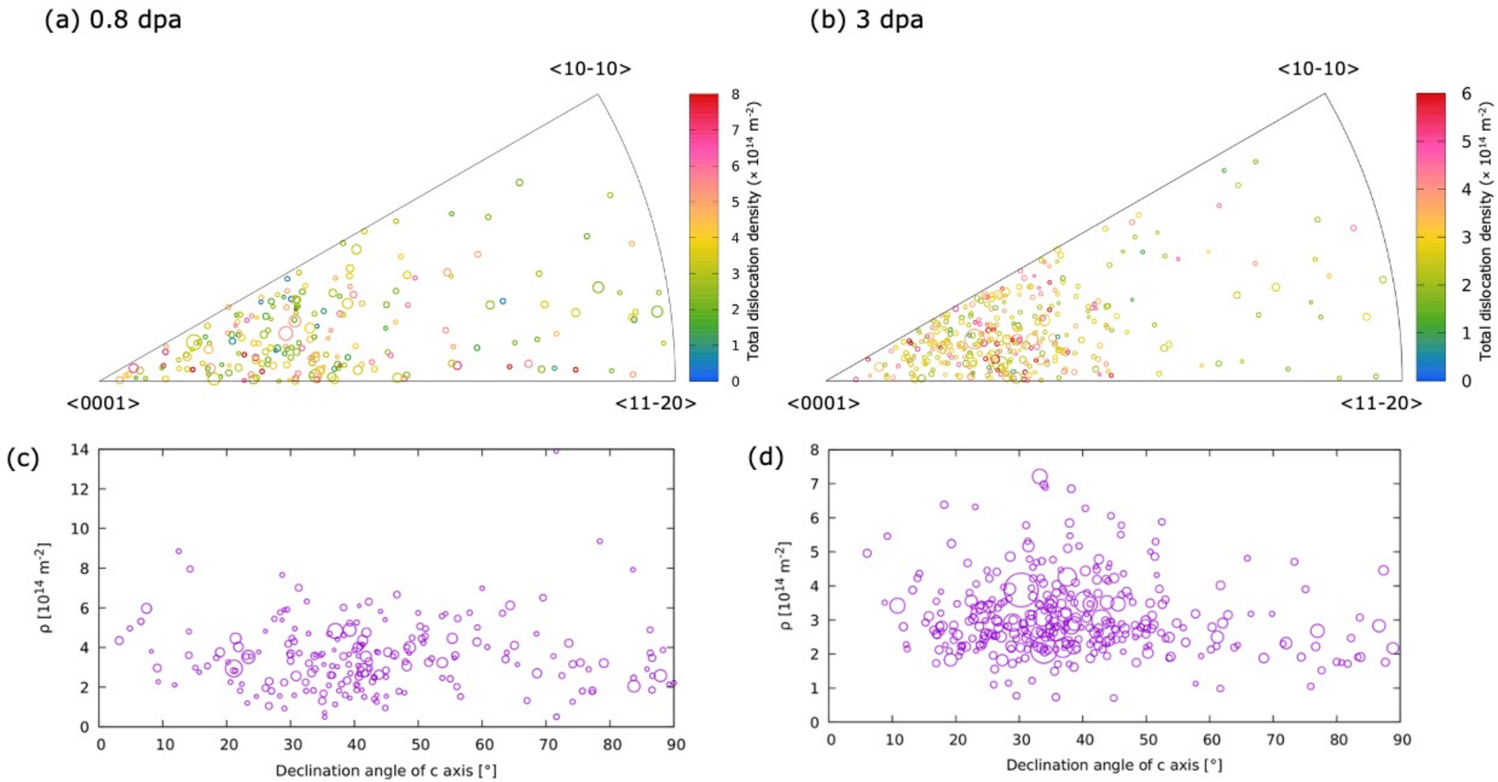
- With each signed hkl reflection, can look at habit plane and Burgers vector

- Two predominant loop types highlighted below



		Average dislocation density per grain in 0.8 dpa sample ($\times 10^{14}$ m^{-2})	Average dislocation density per grain in 3.0 dpa sample ($\times 10^{14}$ m^{-2})
1/3 <11-20> [11-20]	<a>	0.37	0.19
1/3 <11-20> [10-10]	<a>	3.09	2.15
1/3 <10-10> [10-10]	<a>	0.27	0.25
1/3 <11-23> [10-11]	<c>	0.05	0.05
1/3 <11-23> [10-10]	<c>	0.02	0.02
1/6 <20-23> [0001]	<c>	0.38	0.95

MIDAS Orientation dependance



- No clear correlation between grain orientation and dislocation density

MIDAS Conclusions

- **<a>-loop dislocation densities** in neutron-irradiated BOR-60 samples **saturate at low dose**
- Both **<a>- and <c>-loop density is higher in Zircaloy-2** compared to low Sn ZIRLO at 32 dpa
- **Axial growth strain only weakly correlated to <c>-loop** density in neutron-irradiated BOR-60 samples Extent of the damaged region in Zircaloy-4 irradiated with 2 MeV protons is **consistent with prediction from simulation**
- **For low doses, a variable irradiation damage profile** is observed as a function of depth, whereas saturation is observed for higher doses.
- There exists a **grain-to-grain dislocation density variability** in proton-irradiated Zr-0.1Fe
- **No trend between dislocation density and crystallographic orientation** relative to the irradiation direction was observed, suggesting that channelling is not significant for the crystallographic orientations sampled
- Mostly, **<11-20> [10-10] <a>-loops** and **<20-23> [0001] <c>-loops** were observed, with all other loop types having an order of magnitude lower dislocation densities.

Thank you for your attention

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MIDAS Acknowledgments

- Hattie Xu, Sheng Cao, Matthew Topping, Alistair Garner, Albert Smith
- BOR-60 samples provided by the EPRI led NFIR programme
- Manchester Zr group

- MIDAS EPSRC programme grant (EP/S01702X/1)
- DESY (Hamburg, Germany)
- Advanced Photon Source
- Part-funded by the EPSRC Energy Programme grant number EP/T012250/1
- G.R. is grateful for the support of OTKA grant K124926 funded by the Hungarian National Research, Development and Innovation Office (NKFIH)