Detection of defects in displacement-damaged tungsten and iron

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EUROfusion Enabling research project - DeHydroC - ENR-MAT-01-JSI

Outline

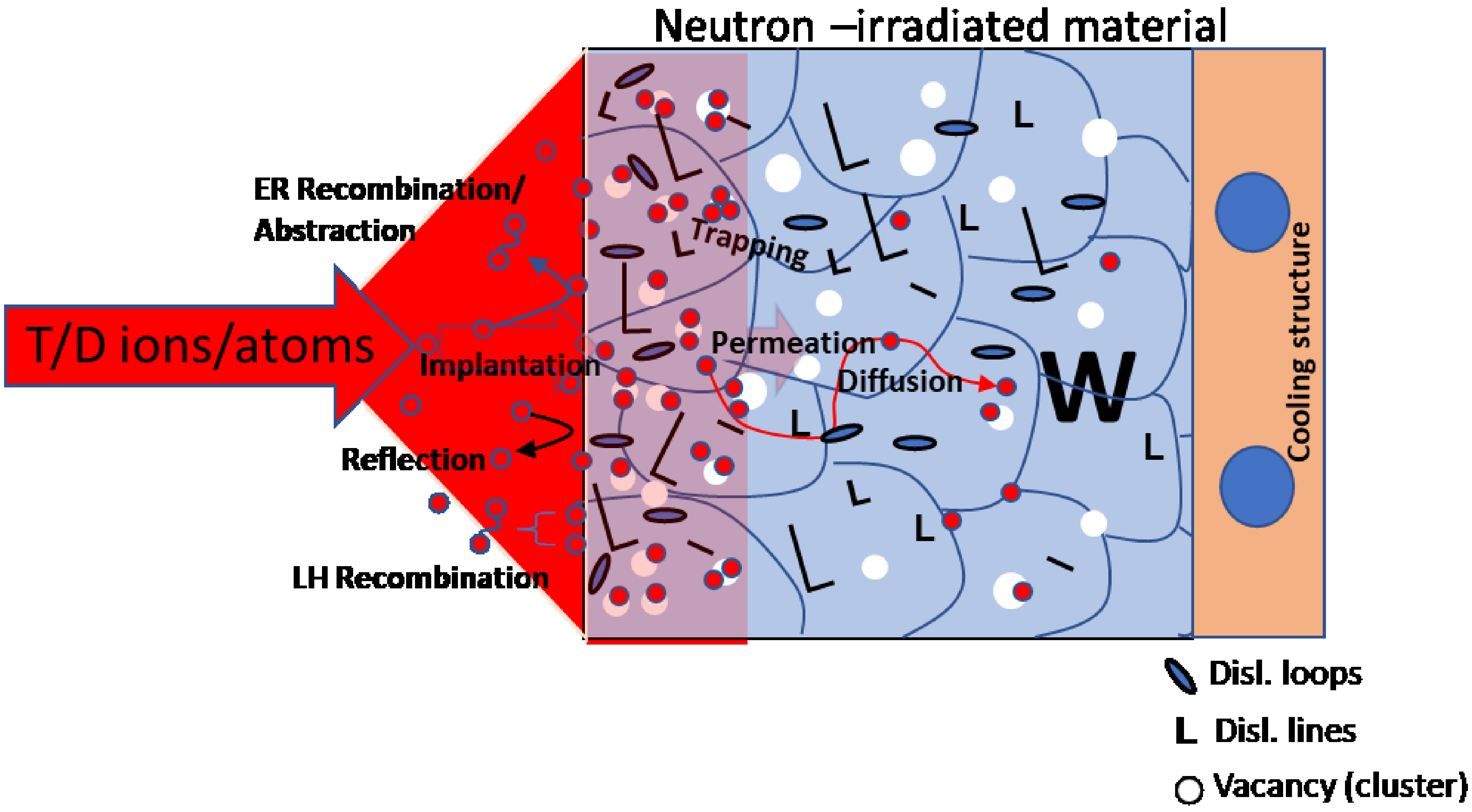
- Methodology
- Detection of **defects** by **Rutherford Backscattering Spectroscopy** in **Channeling** configuration (RBS-C) in W
- RBSADEC modelling
- Detection of **defects** by **Rutherford Backscattering Spectroscopy** in **Channeling** configuration (RBS-C) in Fe
- Conclusion

Motivation

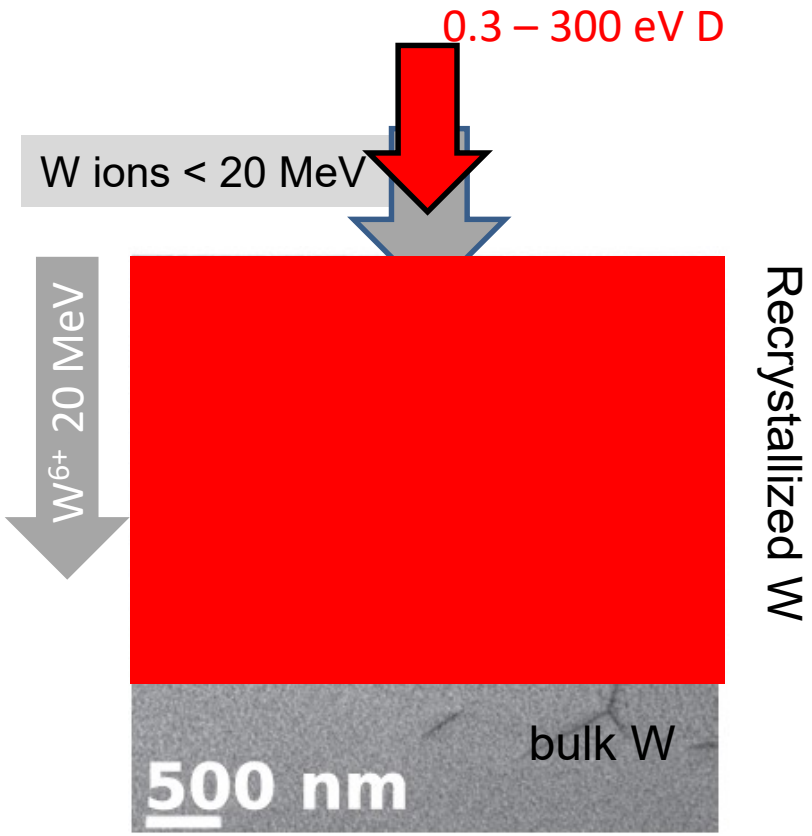
Hydrogen isotope interaction with displacement damage material

Tungsten and steels – first wall and divertor

- We need to be able to predict tritium loss in the first wall of a future fusion reactor
- Focus on the bulk and the influence of neutrons
- Last ten years the focus was on displacement damage



Separating individual aspects of the interaction



Damaged layer characterization by Scanning Transmission Electron Microscopy (STEM) [Založnik et al. Phys Scr. T167 (2016) 014031]

e.g.: S. Markelj JNM 469 (2016) 133–144; Založnik et al. Phys Scr. T167 (2016) 014031; S. Markelj et al., Nucl. Fusion 59 (2019) 086050; M. Pečovnik et al., Nucl. Fusion 60 (2020) 036024; etc.

W ion irradiation by MeV W ions
- Creation of displacement damage
- Dense cascades

- Exposure to D atoms/ions – to only populate the existing traps without producing new ones
- Open volume defects are traps for hydrogen isotopes [S. M. Myers et al., JNM 165 (1989) 9–64]

Methodology to quantify D retention, defect evolution and defect concentration:

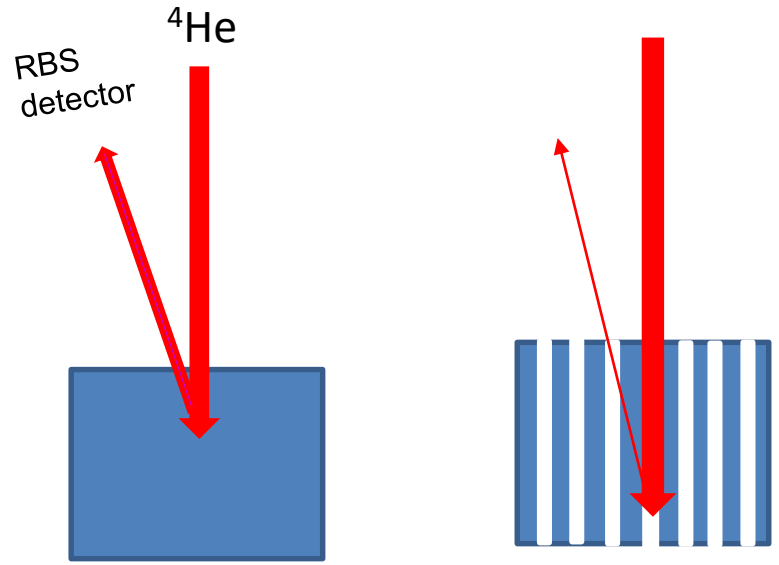
- **Measure D concentration by nuclear reaction analysis (NRA) via $D(^3He,p)^4He$**
- **Desorption kinetics by thermal desorption spectroscopy (TDS)**
- **Use macroscopic rate equation modelling**

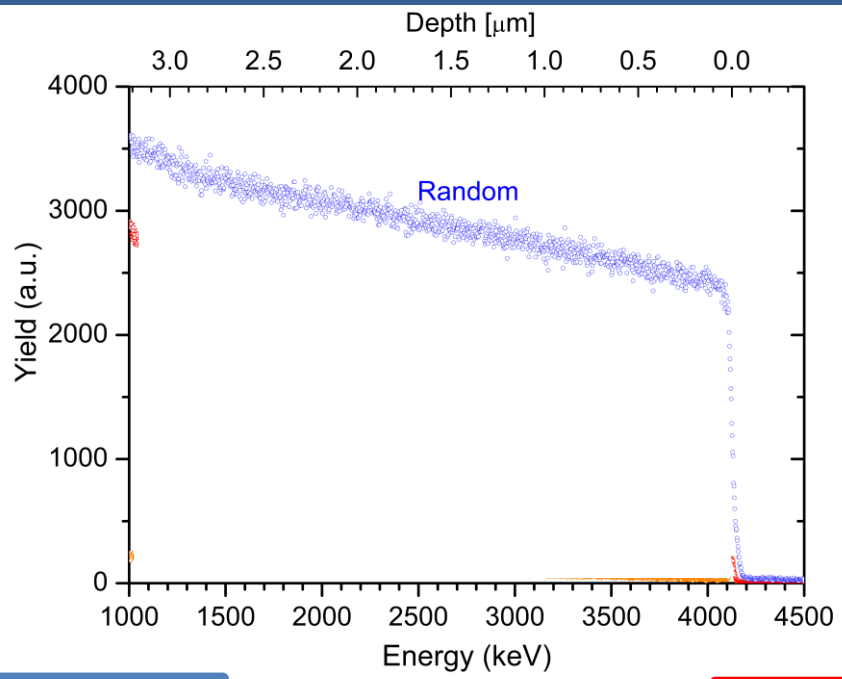
Which defects are really responsible for hydrogen isotope retention?
Where does hydrogen sit?
Can we get more information about the defects?

Methodology

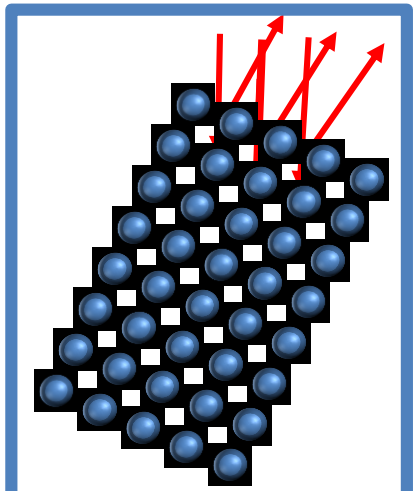
Rutherford Backscattering Spectroscopy (RBS)

In Channeling (RBS - C)

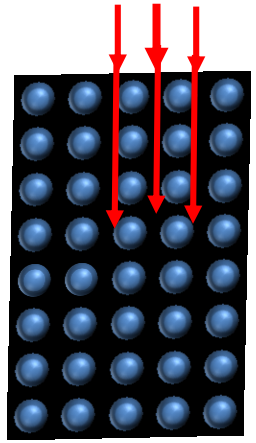




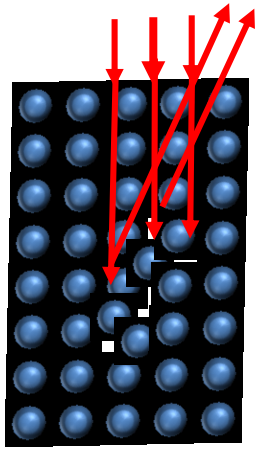
- Ion beam technique
- Non-destructive!
- In situ!
- Study disorder in materials due to irradiation (depth resolved)
- Defect type? Sensitive mainly to dislocation structure (loops, lines)



Amorphous material
Random direction



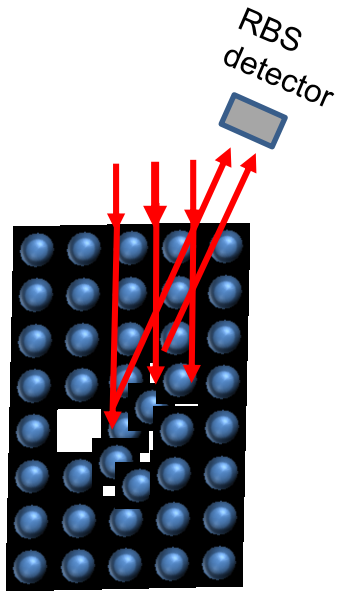
Pristine crystal - aligned



Dis-ordered crystal - aligned

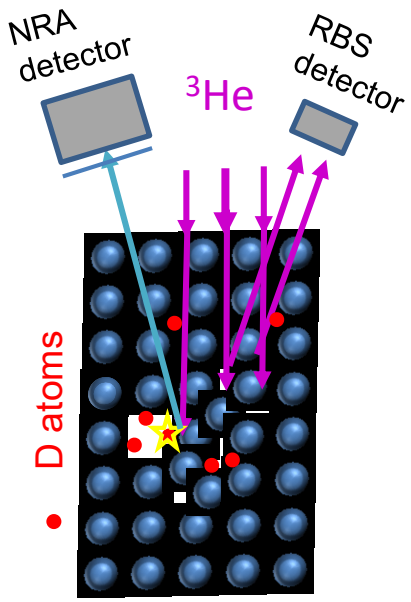
Methodology

Channeling Rutherford Backscattering Spectroscopy (RBS - C)



Nuclear Reaction Analysis (NRA - C)

$D(^3He,p)^4He$



- Ion beam techniques
- Non-destructive!
- In situ!

- Disorder in materials due to irradiation (depth resolved)

Defect type?

- Sensitive mainly to dislocation structure (loops, lines)

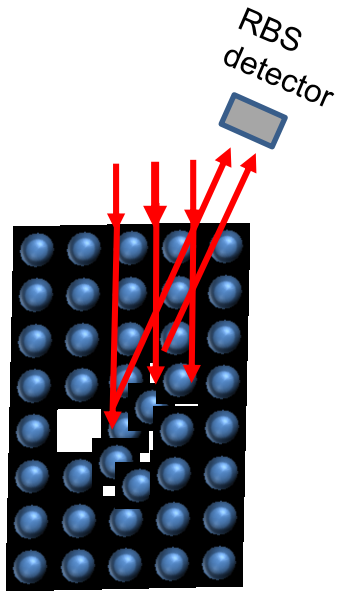
- Deuterium populates defects in materials – analysis depth resolved

Defect type?

- D traps mainly in open volume defects (vacancies, vacancy clusters)

Methodology

Channeling Rutherford Backscattering Spectroscopy (RBS - C)



- Disorder in materials due to irradiation (depth resolved)

Defect type?

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Nuclear Reaction Analysis (NRA - C)

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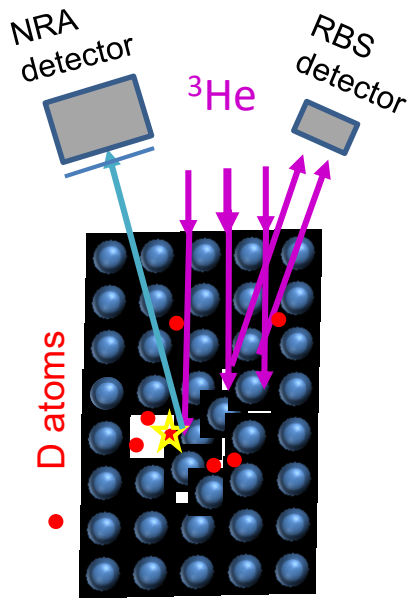

- Ion beam techniques
- Non-destructive!
- In situ!

- Deuterium populates defects in materials – analysis depth resolved

Defect type?

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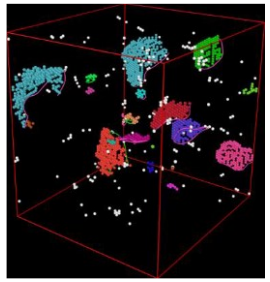
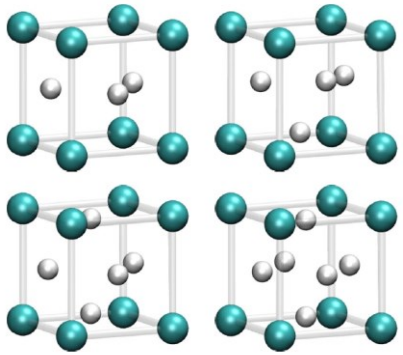
Molecular Dynamics (MD)



- Collision cascades $(\sim 10-100\ \mathrm{nm})$
- High radiation dose

Density Functional Theory (DFT)



Heinola et al. PR B (2010)

- Hydrogen atom positions around vacancy/vacancy cluster

Methodology

RBS-C experiment

+

NRA-C experiment

$D(^3He,p)^4He$

+

Molecular Dynamics (MD)
Density Functional Theory (DFT)

RBS-C / NRA-C simulations
RBSADEC code

[S. Zhang *et al.*, *Phys. Rev. E*, 94 (2016) 043319]

UNIVERSITY OF HELSINKI

Combination of MD, DFT
RBS-C, NRA-C experiments and simulations

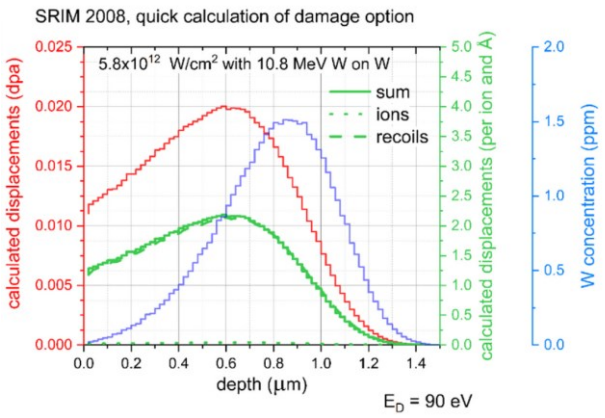
Defect types and deuterium locations!

Creation of defects in tungsten – different dominating defect

Samples: tungsten single crystals (111) and (100)
RBS-C NRA-C

Irradiation: 10.8 MeV W ions

@ two damage doses and two temperatures to create different dominating open volume defect types



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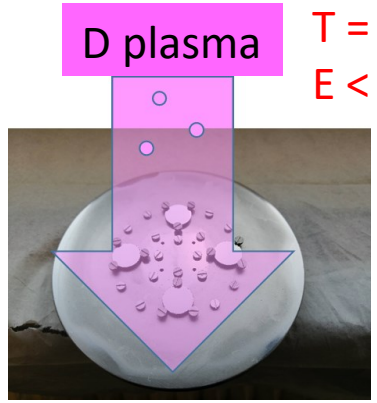
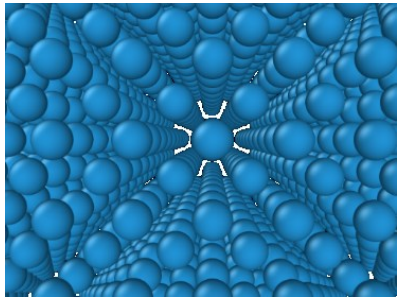


T = 370 K,
E < 5 eV/D

'gentle' loading = 'decoration' of defects
ion flux: 6 ×10^{19} D/(m^2s)
ion fluence: 1·10^{25} D/m^2 (48 h)

Sample	Irradiation conditions	Predominant defect expected	Vacancy cluster size by PAS
78g / #1	0.02 dpa, 290 K	single vacancies	v = 2
78f / #5	0.2 dpa, 290K	heavily damaged standard	v = 2 - 4
78c / #3	0.02 dpa, 800 K	small vacancy clusters	v = 25
78b / #2	0.2 dpa, 800 K	big vacancy clusters	v = 50

- ✓ Initial assumptions on open volume defects based on Hu et al. JNM 556 (2022) 153175
- ✓ Confirmed by Positron Annihilation Spectroscopy (PAS)



W (100)

See: J. Zavašnik et al. Materials Characterization 224 (2025) 115050.

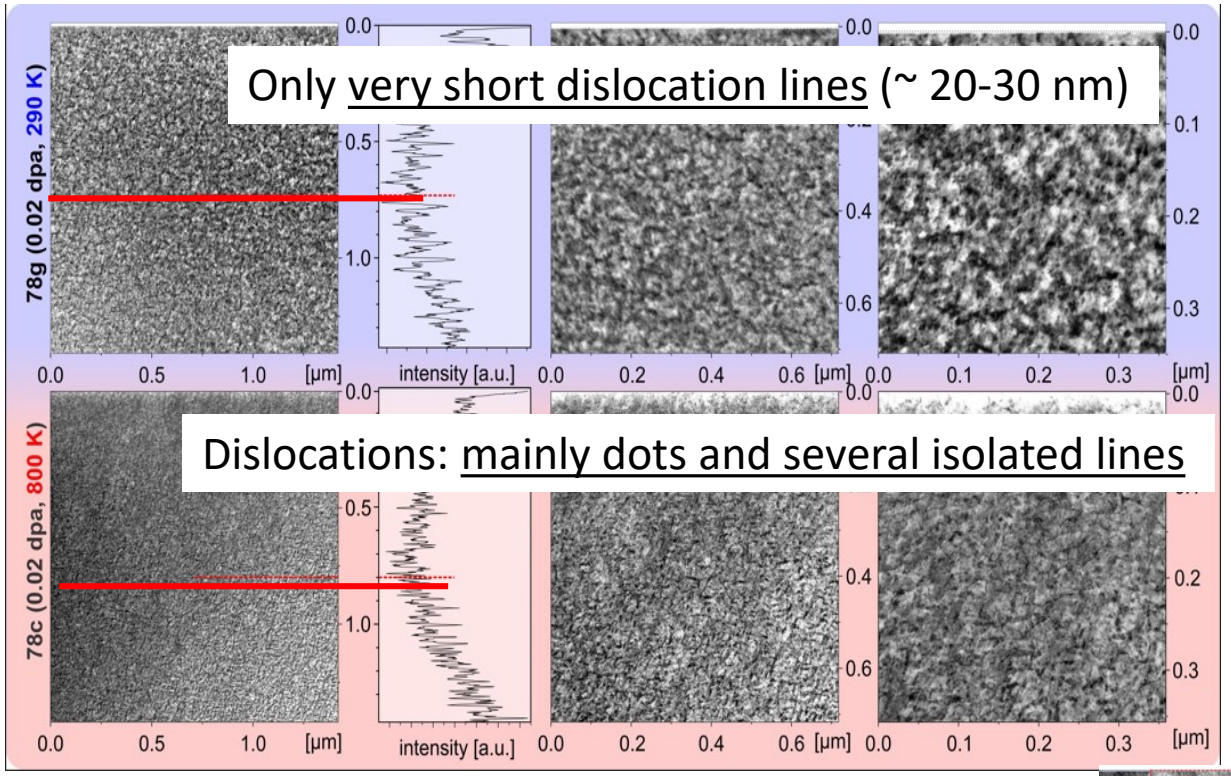
Transmission electron microscopy (TEM) analysis – dislocations, voids

Low dose 0.02 dpa

High dose 0.2 dpa

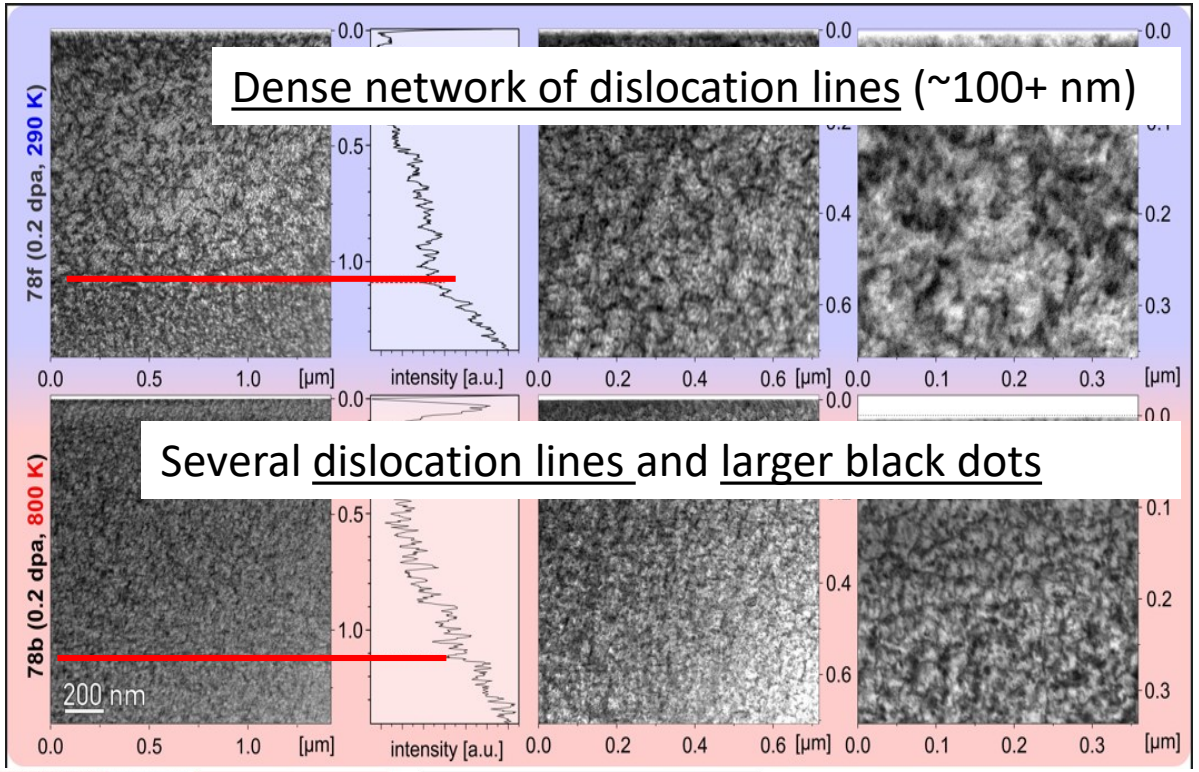
290 K

800 K



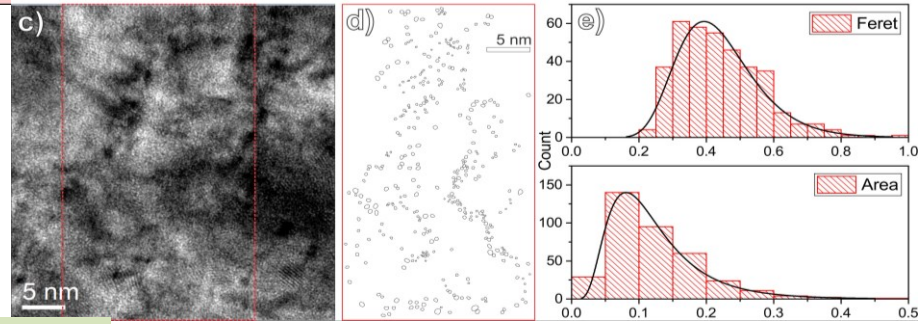
Only very short dislocation lines (~ 20-30 nm)

Dislocations: mainly dots and several isolated lines



Dense network of dislocation lines (~100+ nm)

Several dislocation lines and larger black dots

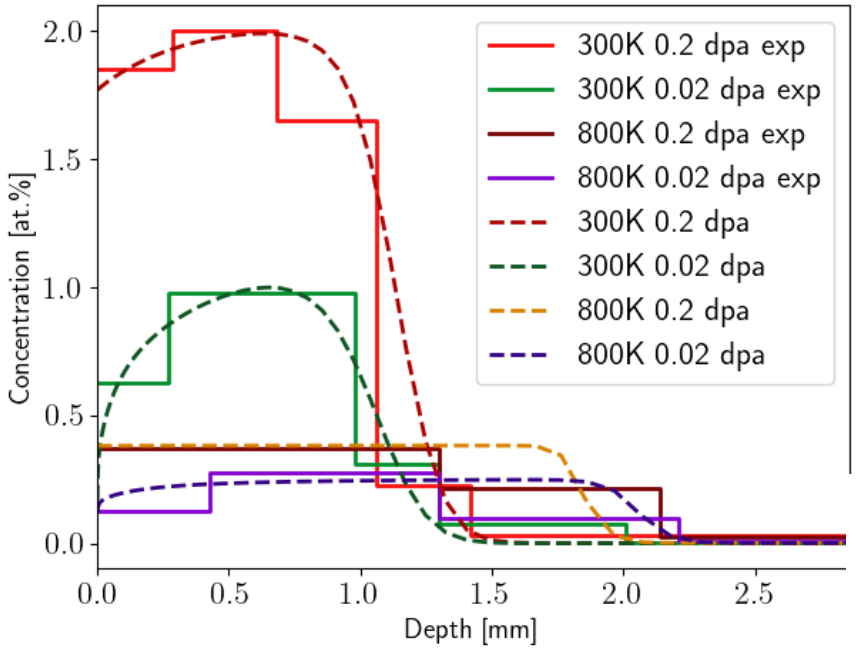


Detection of small voids in 0.2 dpa / 800 K sample

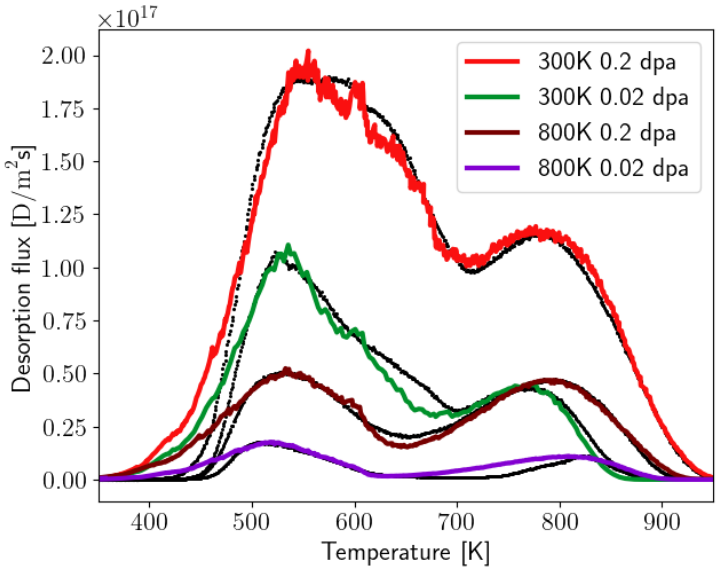
See: J. Zavašnik et al. Materials Characterization 224 (2025) 115050.

D retention in W damaged samples

W(111) samples analysed by PAS
NRA

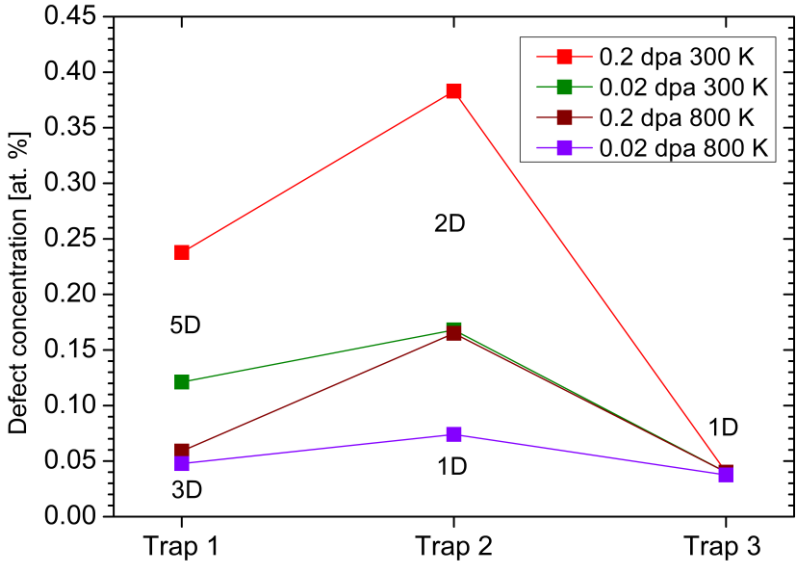


TDS



[J. Zavašnik et al. Materials Characterization 224 (2025) 115050.]

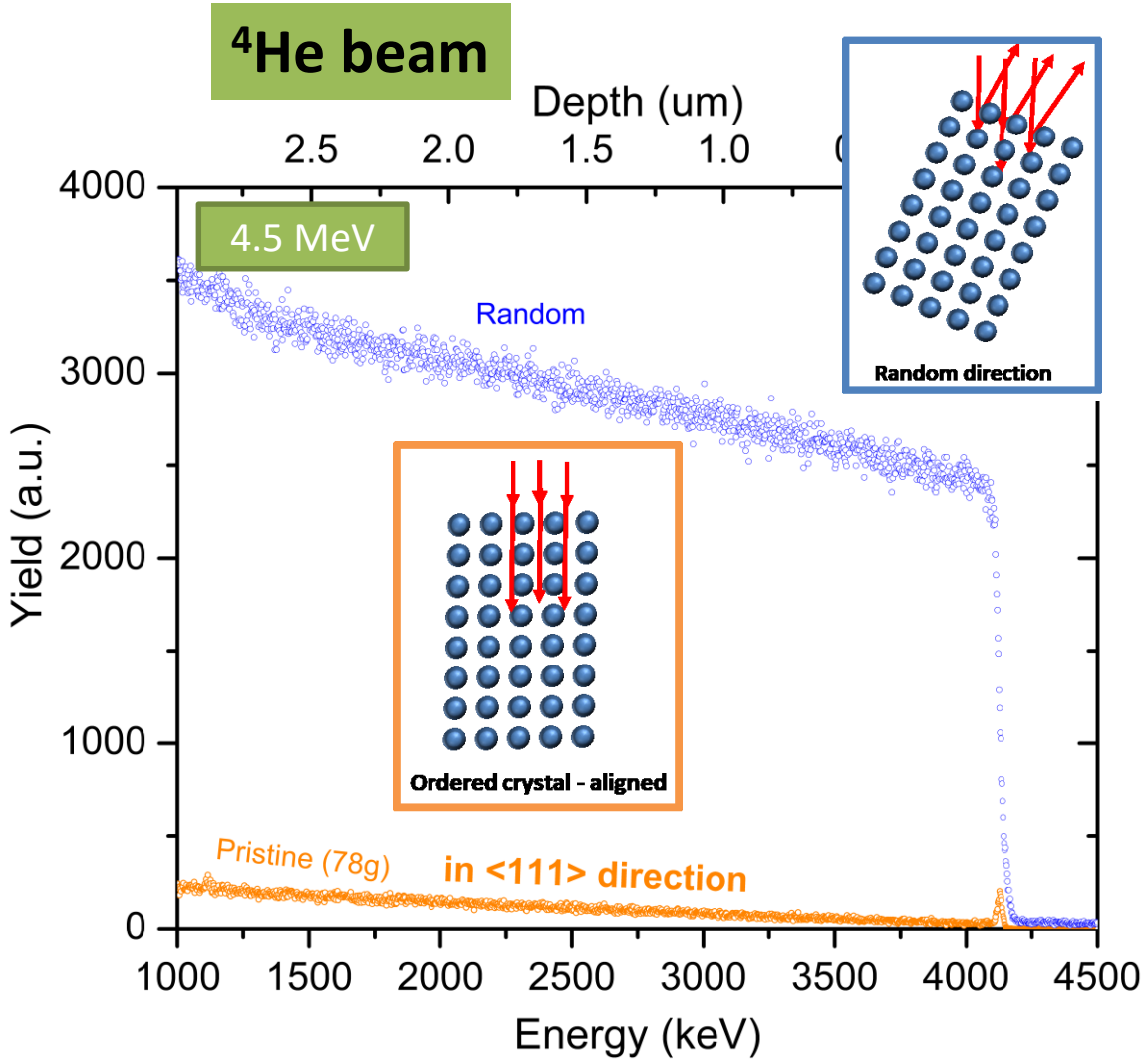
➢ From rate equation modelling



C-RBS – measurements

- Backscattered ^{4}He ions along <111> channel
- Spectrum from pristine W @ 4.5 MeV
- Random = Σ100 spectra – nonchanneling
- Ratio of the signal between Pristine/Random after the surface peak = 1.3 %

✓ Starting material (pristine sample) has a very good crystallinity



Measurements were preformed at CMAM, Madrid

[S. Markelj, et al., Acta Mater., 263 (2024) 119499]

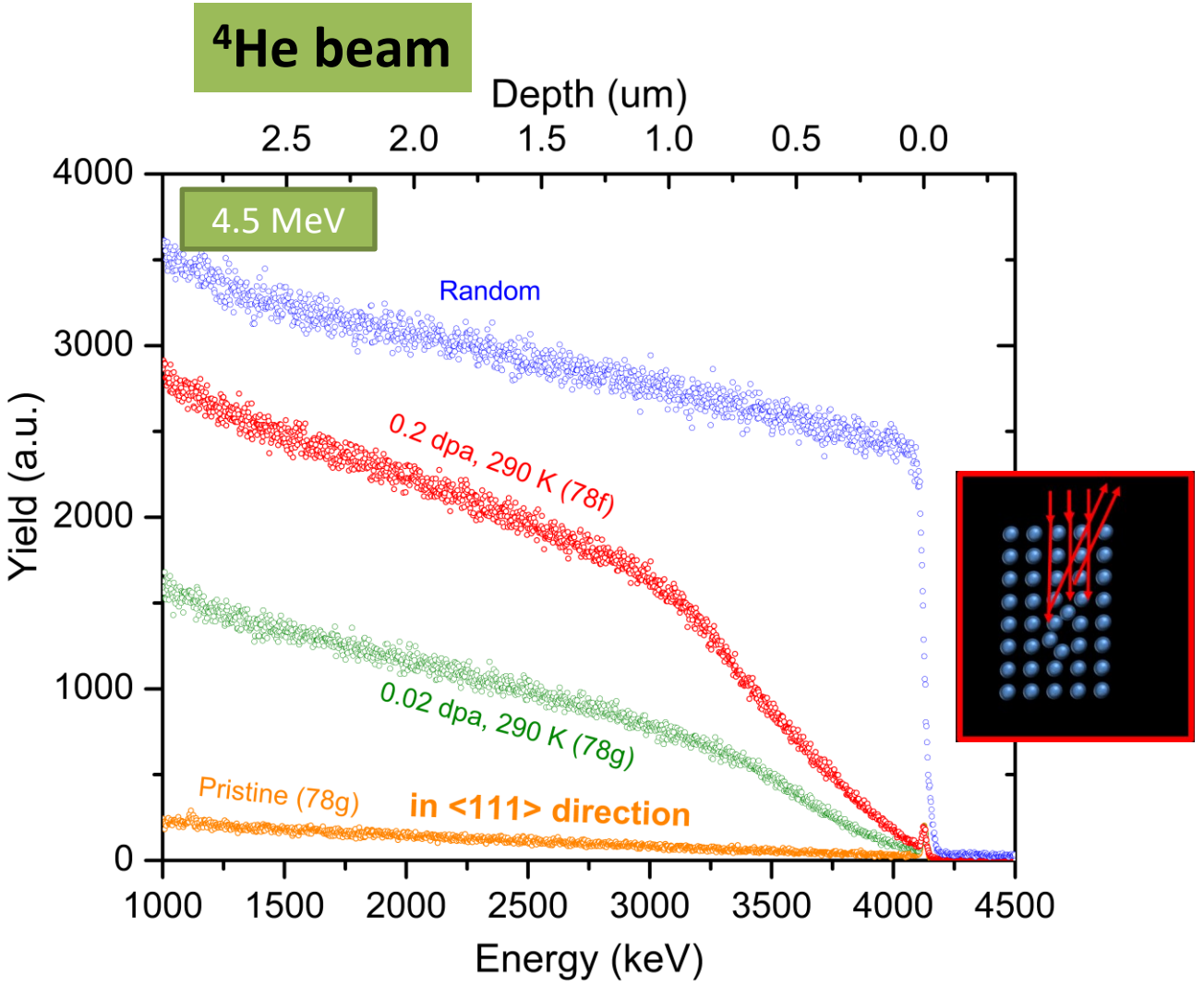
C-RBS - measurements

- Backscattered ^{4}He ions along <111> channel

➢ RBS-C spectra @ 4.5 MeV

Irradiation at 290 K:

- 78f : 0.2dpa, 290 K (heavily damaged standard)
- 78g : 0.02 dpa, 290 K (single vacancies)



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[S. Markelj, et al., Acta Mater., 263 (2024) 119499]

C-RBS - measurements

- Backscattered ^{4}He ions along <111> channel

➢ RBS-C spectra @ 4.5 MeV

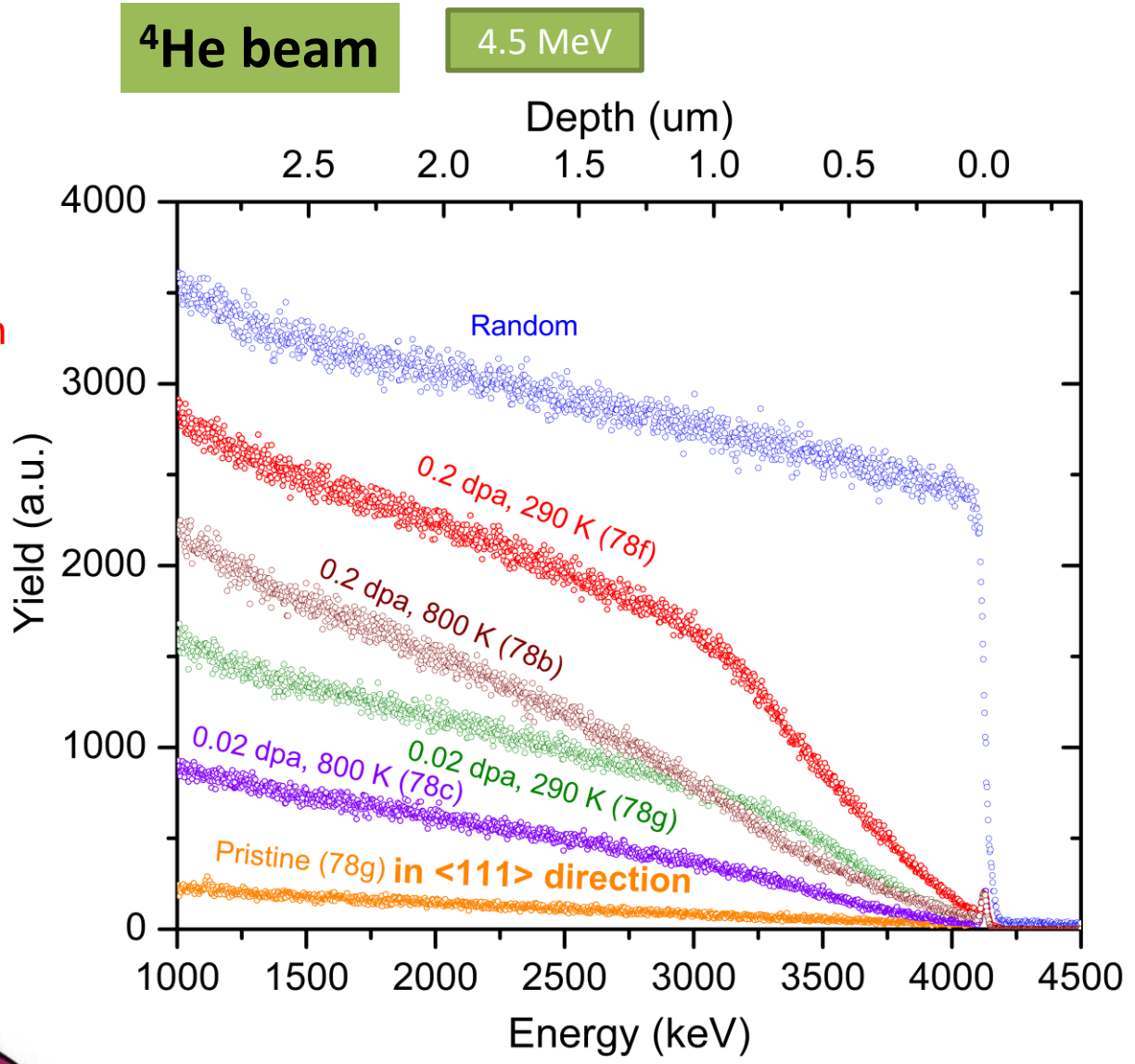
Irradiation at 290 K:

- 78f : 0.2dpa, 290 K (heavily damaged standard) – 1.1 µm
- 78g : 0.02 dpa, 290 K (single vacancies) – 0.8 µm

Irradiation at 800 K:

- 78c : 0.02 dpa 800 K (small vacancy clusters) – ? µm
- 78b : 0.2 dpa 800 K (big vacancy clusters) – 1.3 µm

- **Clear differences between the irradiation damage treatments**

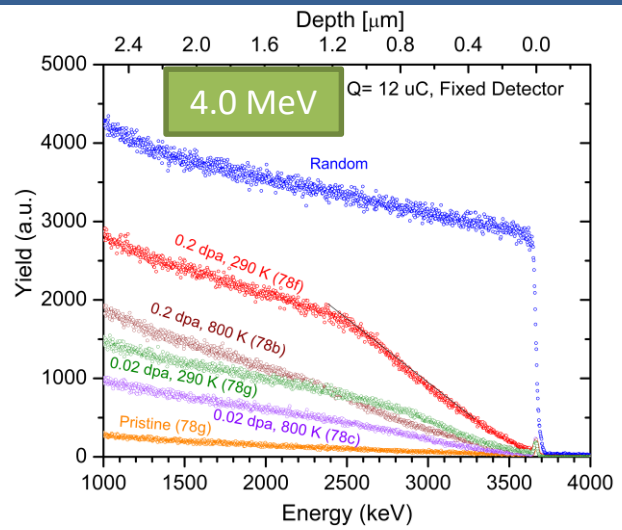


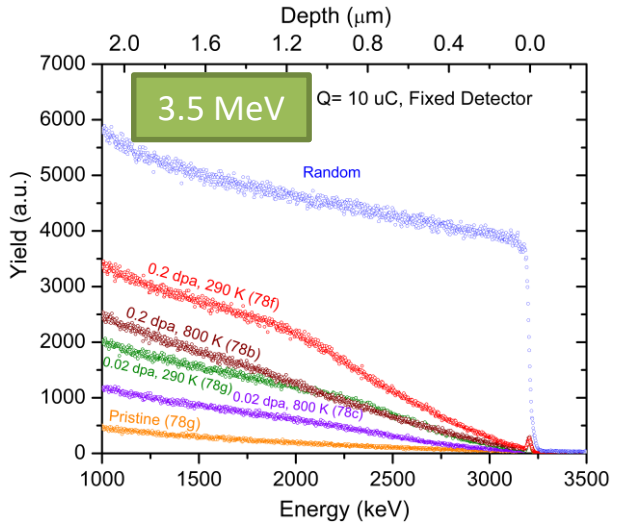
[S. Markelj, et al., Acta Mater., 263 (2024) 119499]

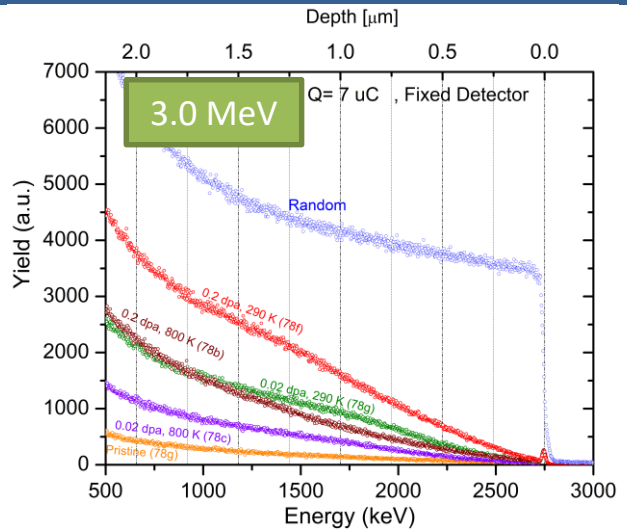
[J. Zavašnik et al. Materials Char. 224 (2025) 115050.]

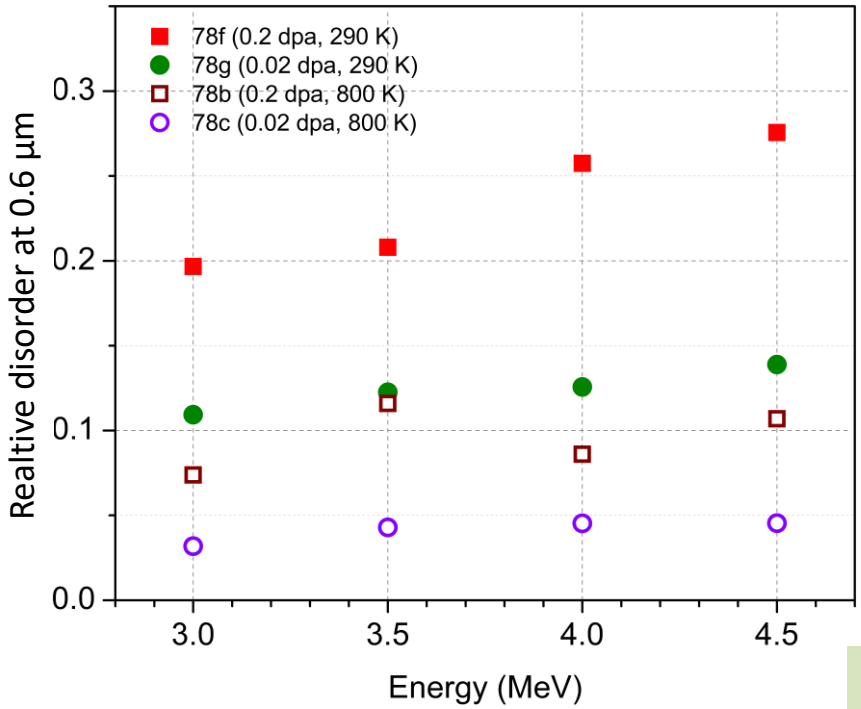
Measurements were preformed at CMAM, Madrid

Multi-energy RBS-C along <111> channel









- **Following:** ***FELDMAN, MATERIALS ANALYSIS BY ION CHANNELING. Submicron Crystallography, 1982, Academic press***

78f (0.2 dpa, 290 K): positive slope –extended defects – dislocation lines (TEM)

78g (0.02 dpa, 290 K): no slope – localized defects – small dislocation loops (TEM)

78b (0.2 dpa, 800 K): no slope – localized defects – dislocation lines and black dots (TEM)

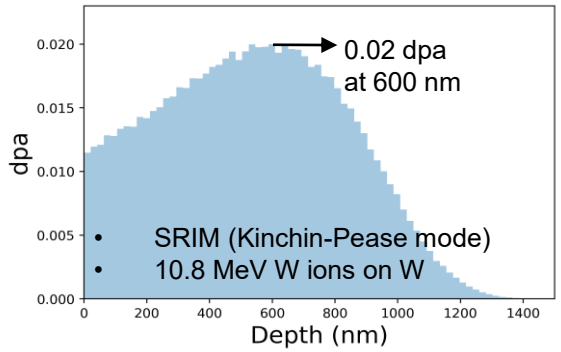
78c (0.02 dpa, 800 K): no slope – localized defects – dots and isolated lines (TEM)

- **Good qualitative agreement with TEM**

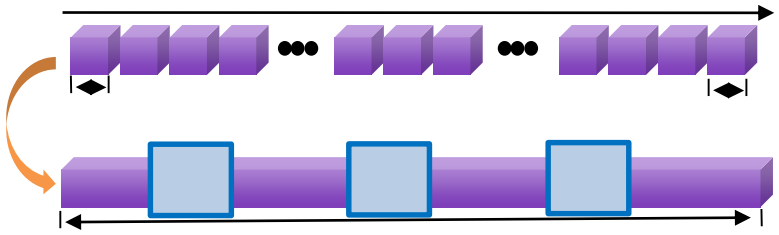
[J. Zavašnik et al. Materials Char. 224 (2025) 115050.]

Simulation of RBS-C spectra using MD – samples irradiated at R.T.

A. Get depth distribution of damage level (SRIM)



B. MD simulations and assemble of MD cells

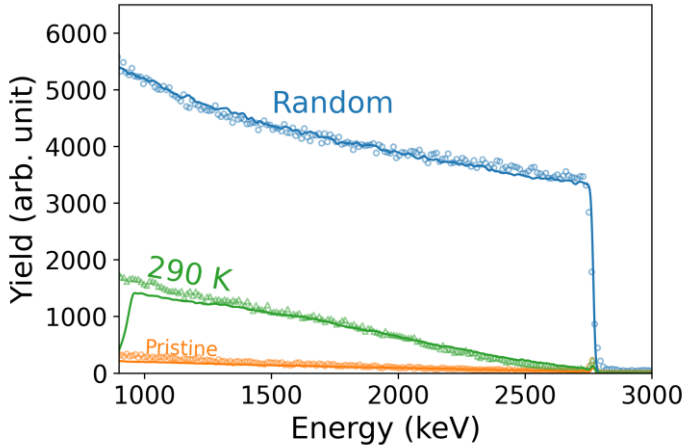


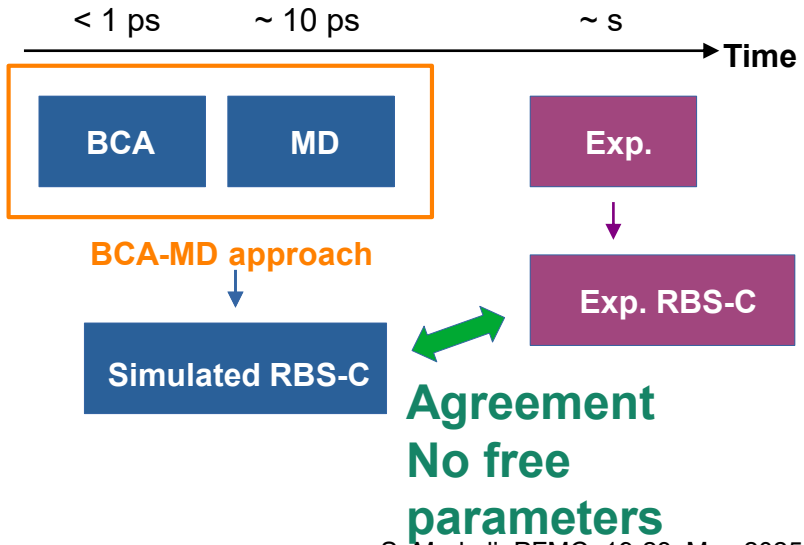
Creation of radiation defects by Molecular Dynamics (MD) calculation

MD cells with certain number of collision cascades
Primary knock-on atom: 10 keV
Size of MD cells: ~ 20nm
Overlapping of cascades: High damage dose (evolution of defects)
[F. Granberg et al., J. Nucl. Mater. 556 (2021) 153158]

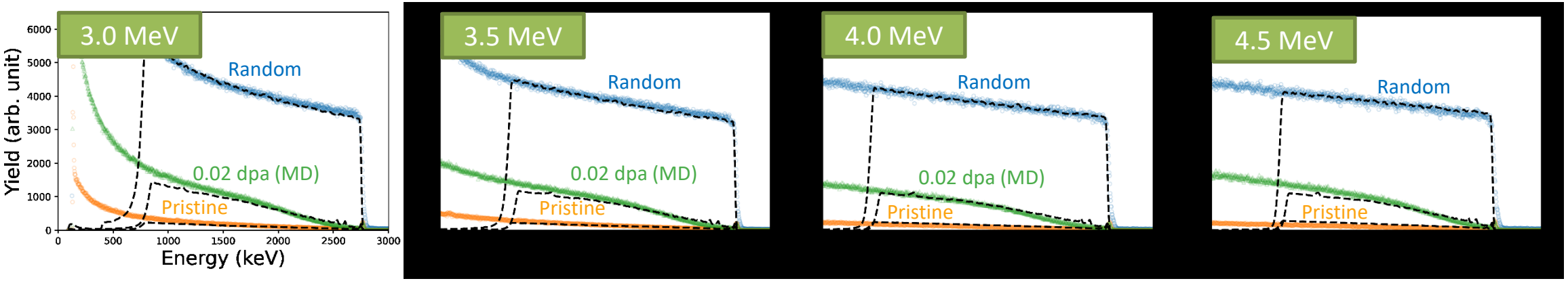
Incorporation of merged MD cells into RBSADEC simulation code

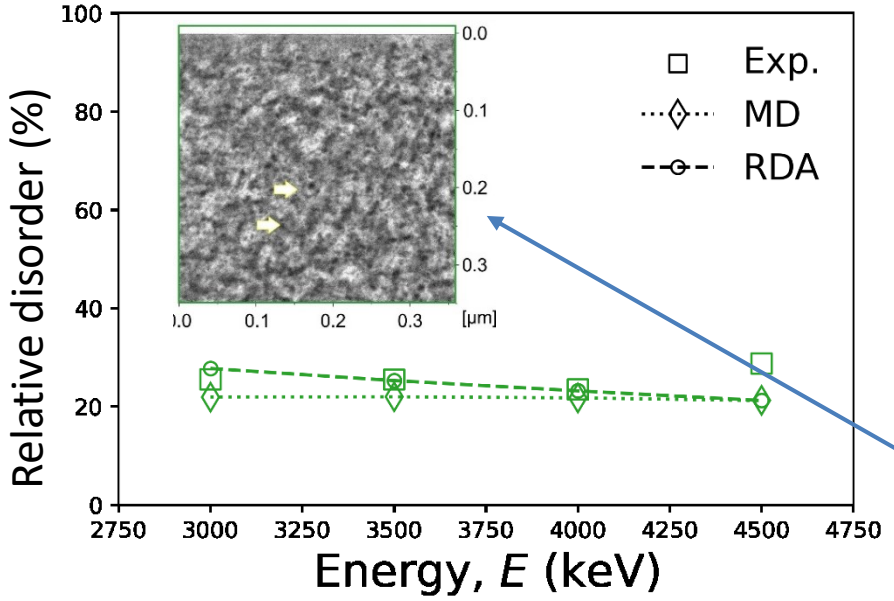
C. Simulation and comparison of RBS-C spectra





Multi-energy RBS-C (0.02 dpa, 290 K) – MD input





(Dechannelling calculated at $1.2 - 1.3\ \mu m$)

❑ **Dechanneling (energy)**

- **Experiments: No obvious trend**
- **Simulations (MD): Constant**
- Simulations (RDA): Negative slope

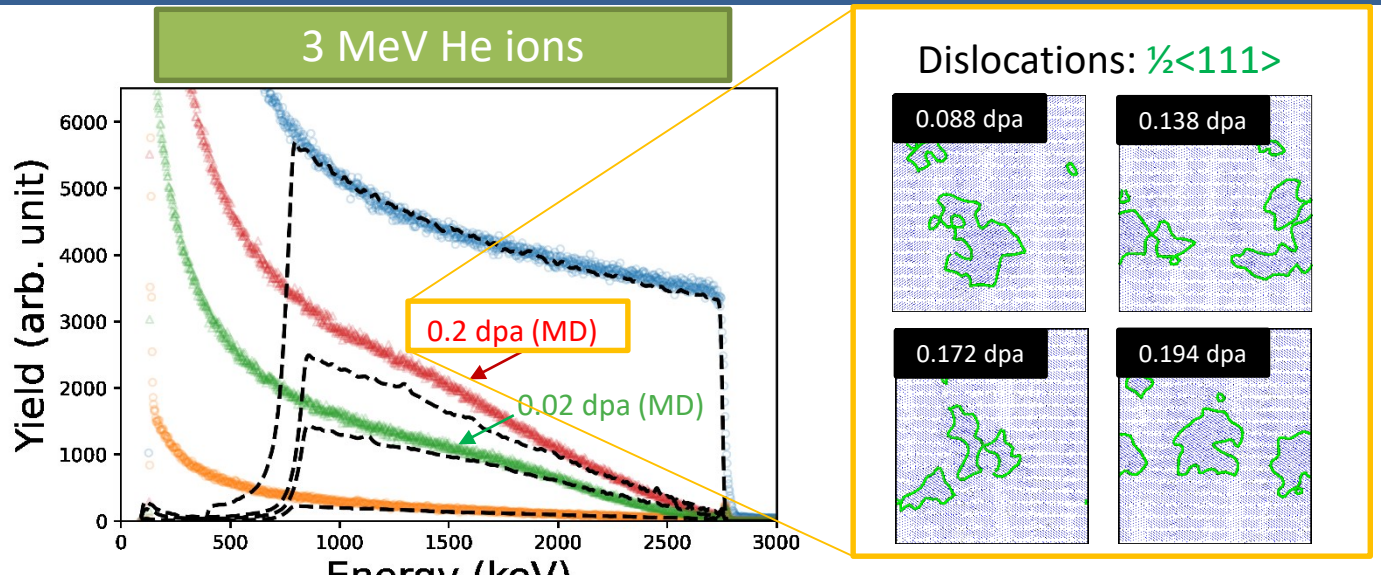
Defect nature: Dislocation loops

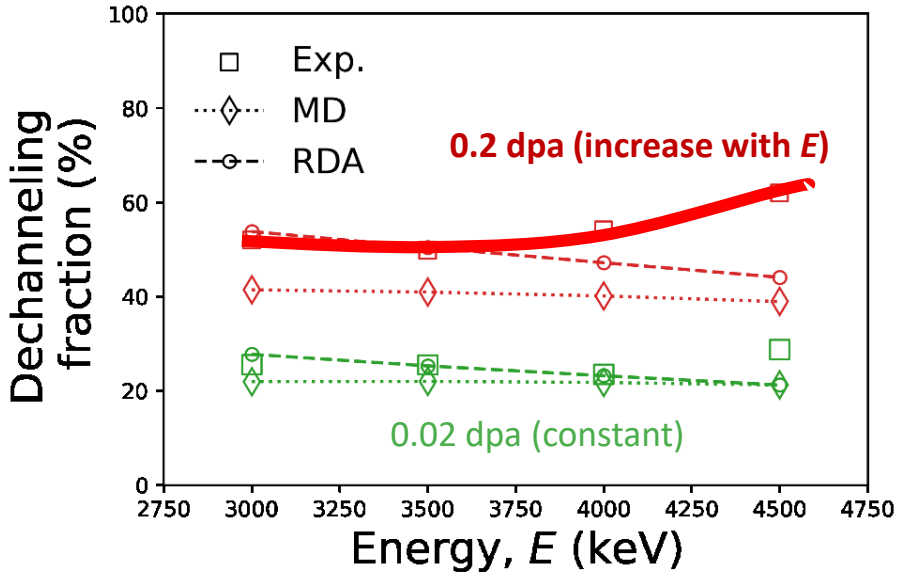
- **STEM results:**

U-shaped dislocation loops around "black dots" (size ~ 10 nm).

[S. Markelj, et al., Acta Mater. 263 (2024) 119499]

Multi-energy RBS-C (0.2 dpa, 290 K) – MD



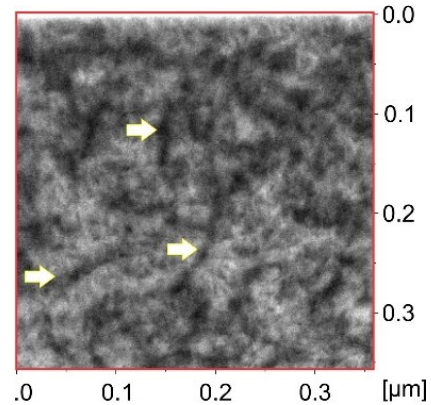


(S. Markelj, et al., Acta Mater., 263 (2024) 119499)

- **RBS-C experiments (high damage, 0.2 dpa)**
  - Higher yield than that of 0.02 dpa
- **RBS-C simulations (MD cells)**
  - **Dechannelling as a function of *E***

	MD	Exp.	RDA
Dechan.	Constant	Increases with E	Decrease
Defect nature	Dislocation loops	Dislocation lines	
Strain field	$1/r^3$	$1/r$	

Lacking agreement between experiment and simulation due to limited size of MD cells.



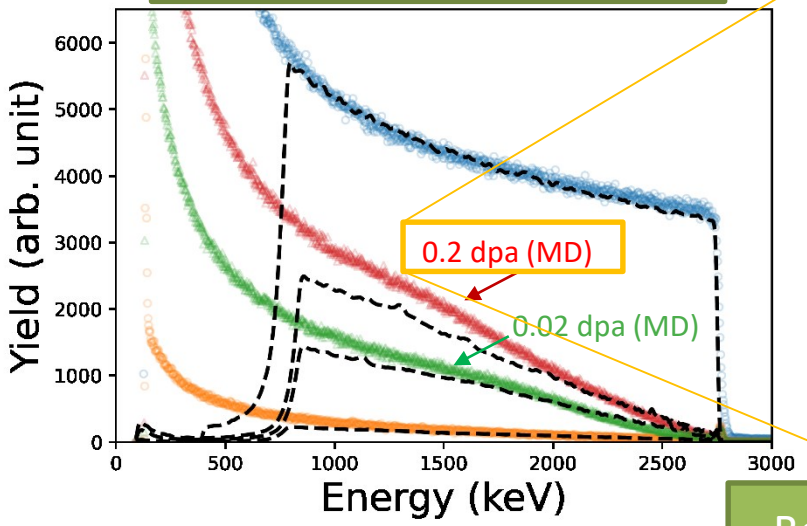
STEM results:

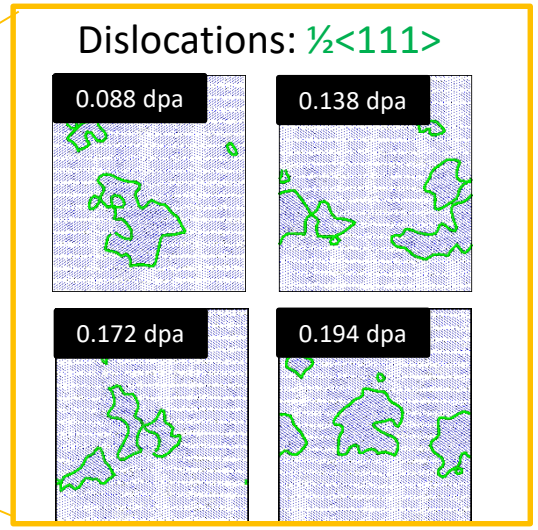
- Dense networks of dislocation lines (length > 100 nm).
- No lines in MD cells: limited size of cells

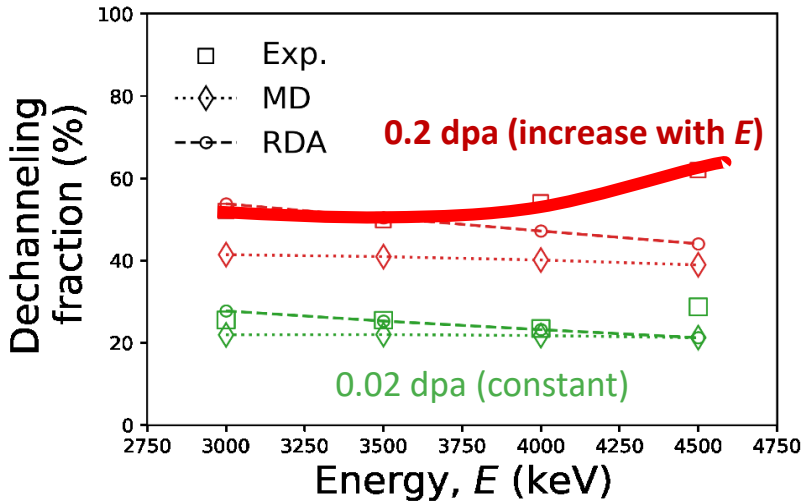
Multi-energy RBS-C (0.2 dpa, 290 K) – effect of MD potentials

Ackland and Thetford potentials (AT-ZN)

3 MeV He ions



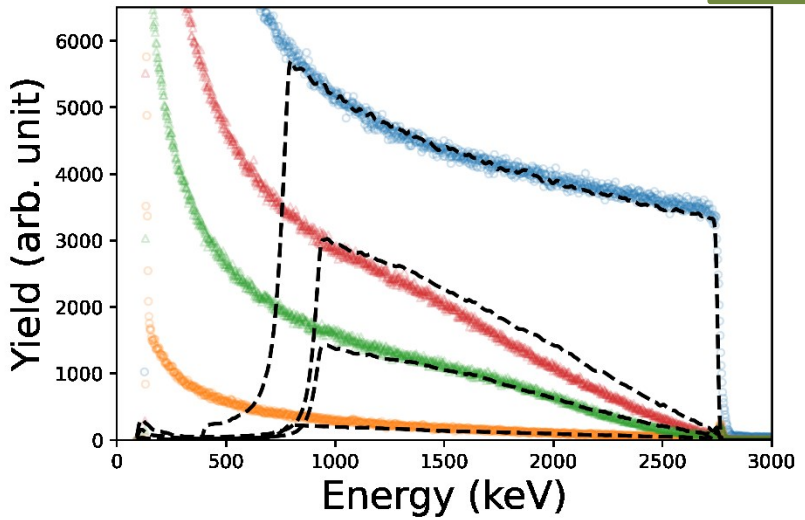


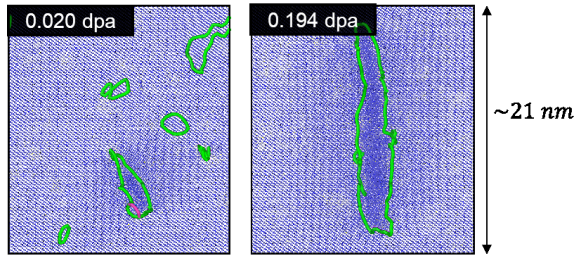


[G. Ackland and R. Thetford, Philos. Mag. A, 56, 1987]

Better agreement with Mason potential

MNL potentials





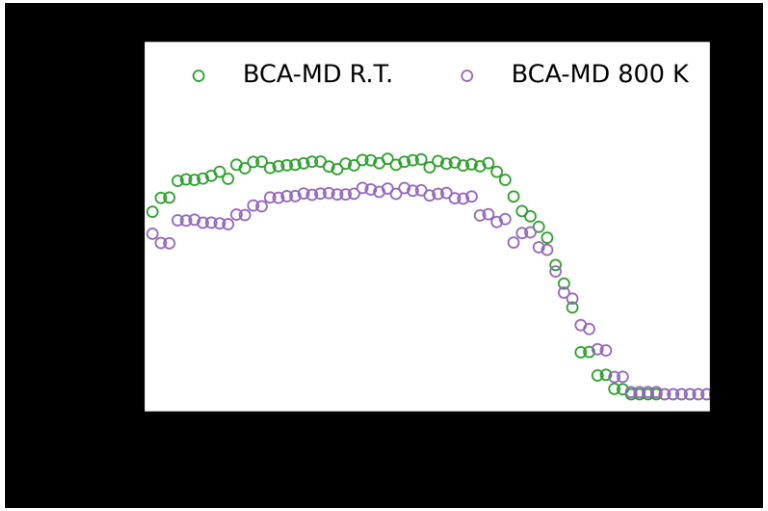
Larger MD cells produced and work ongoing

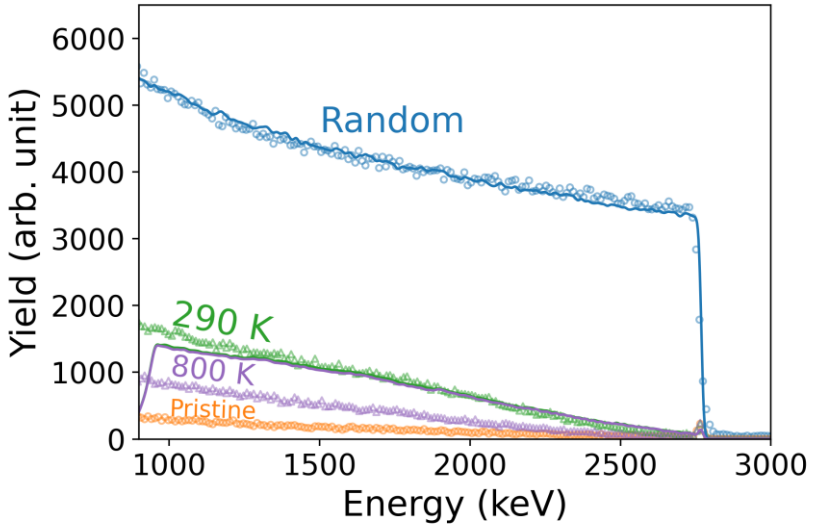
- From 0.02 dpa to 0.2 dpa, the dislocation density is similar, but the size becomes larger
- Larger dislocation loops have a stronger ability of inducing dechanneling

[D. Mason, et al., J. Phys. Condens. Matter, 35, 2023]

Multi-energy RBS-C (290 K vesus 800 K, 0.02 dpa)

The BCA-MD approach (dots)





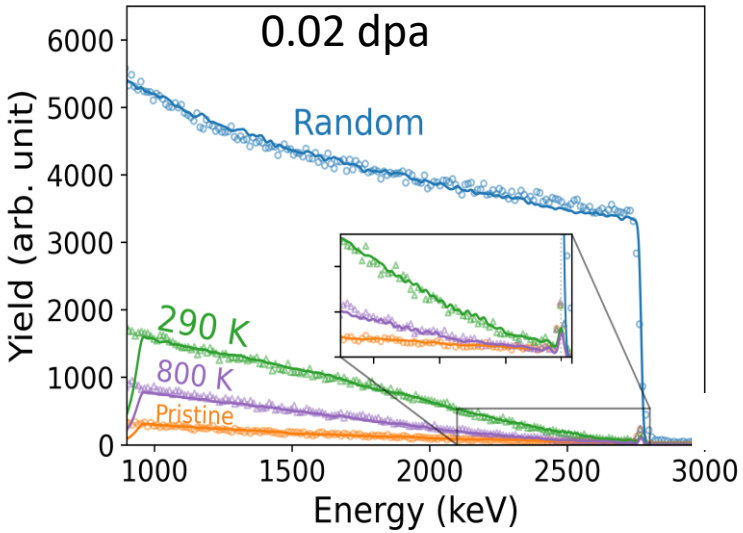
Agreements are not optimal at 800 K:

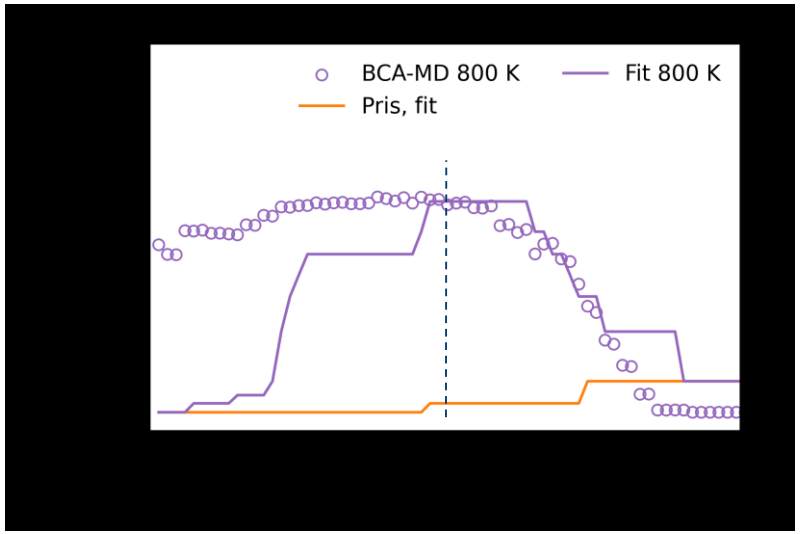
1. With a higher temperature, the experimental yield is smaller. But in simulations using the BCA-MD approach, the yields are almost the same.
2. Dislocation densities in MD cells only decrease a bit.

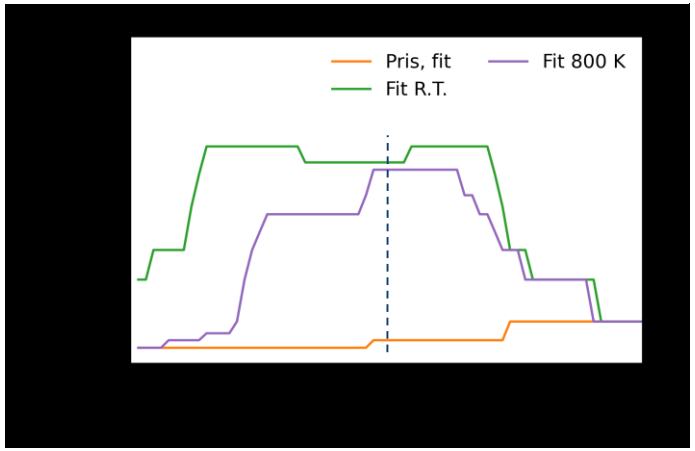
- **No agreement with experiment**
- **MD – no time/temperature evolution of the defects**

Simulation of RBS-C spectra at 800 K

R.T. and 800 K from fitting

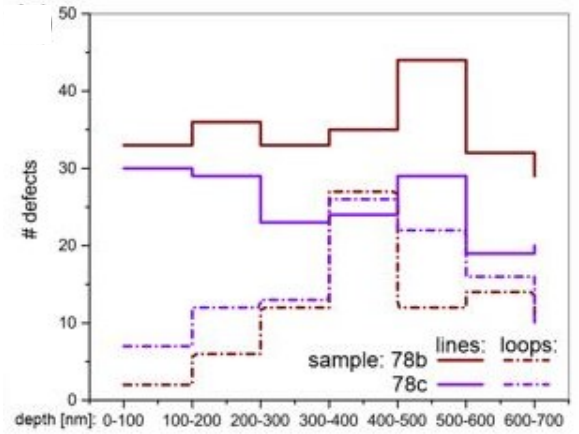


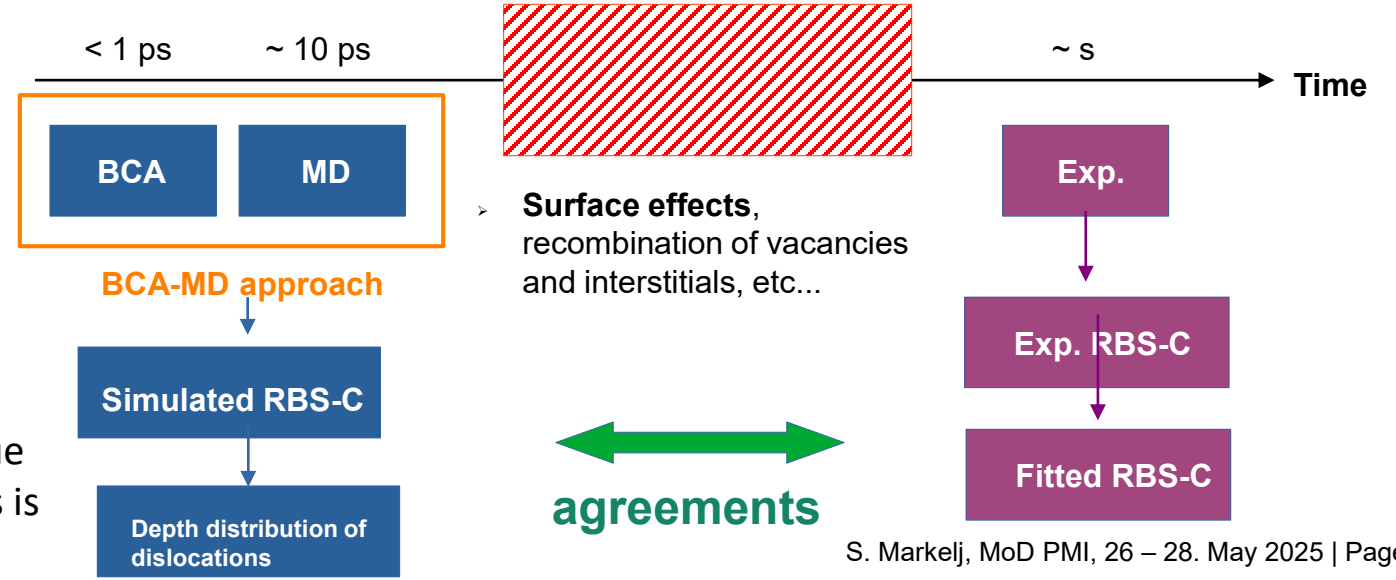




- From R.T. to 800 K, the decrease of RBS-C yield is due to a significant decrease of dislocation density. (This is not reproduced in BCA-MD approach)

- TEM analysis indeed shows the same trend of loop distribution





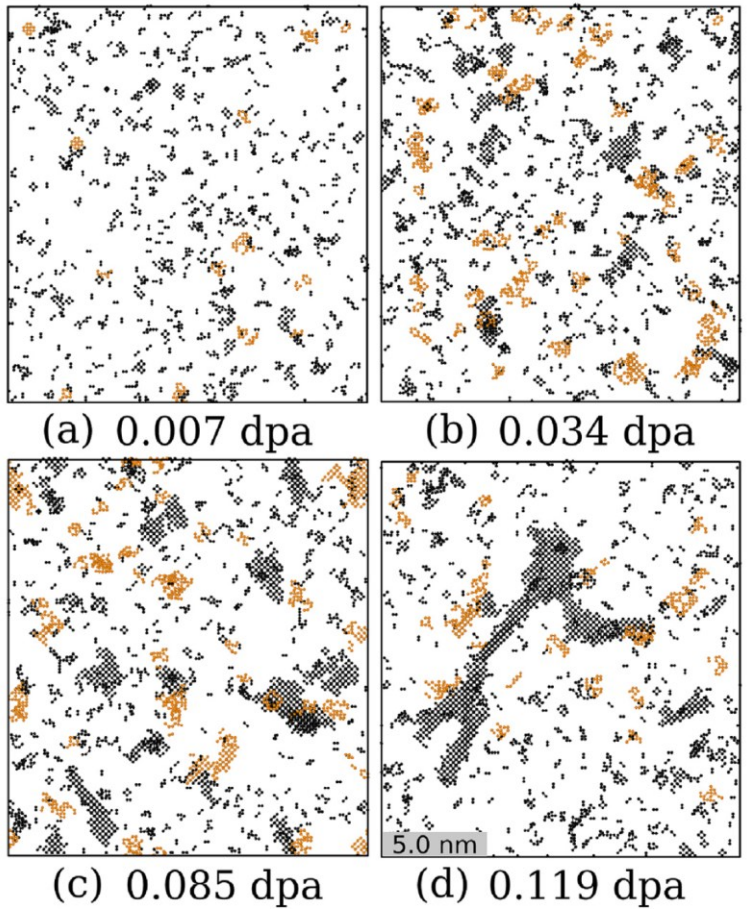
Outline

- Methodology
- Detection of **defects** by **Rutherford Backscattering Spectroscopy** in **Channeling** configuration (RBS-C) in W
- RBSADEC modelling
- Detection of **defects** by **Rutherford Backscattering Spectroscopy** in **Channeling** configuration (RBS-C) in Fe
- Conclusion

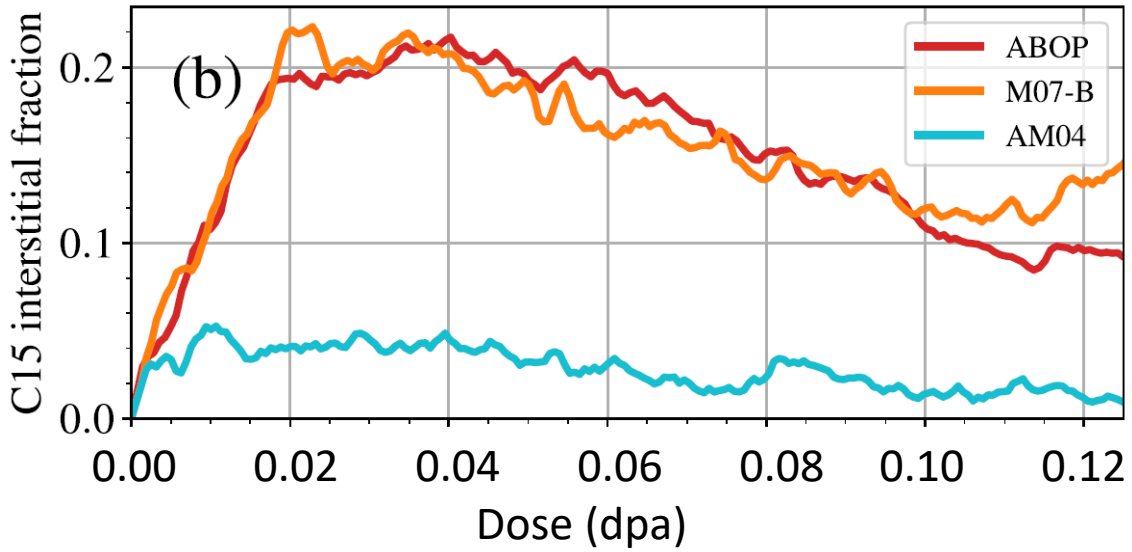
Displacement damage in Fe single crystal

➢ On the experimental evidence of C15 cluster in Fe

- Small SIA clusters are most stable in the form of agglomerates of C15 (1/2 <111> loops for W)
- DFT predicts C15 clusters to be energetically favoured over dislocation loops at small cluster sizes, low doses.



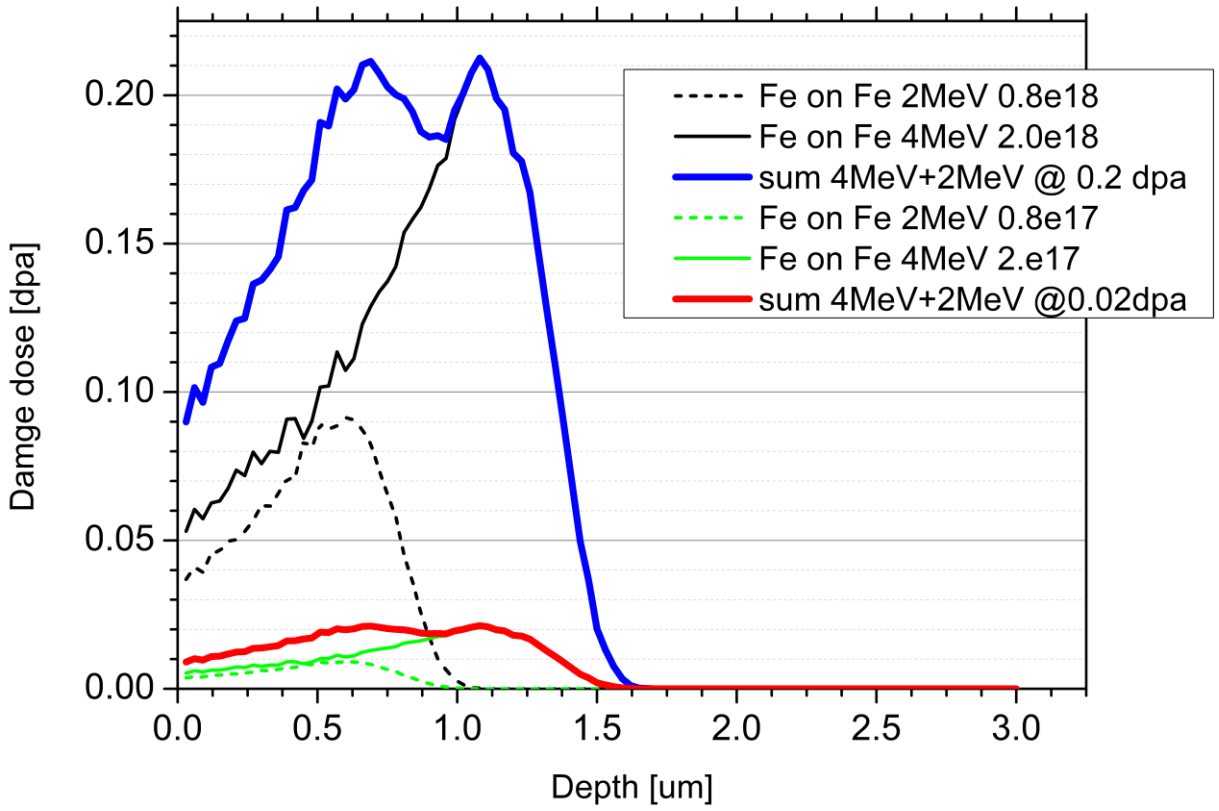
Yellow – C15 clusters



J. Byggmastar and F. Granberg, JNM 528 (2020) 151893

Displacement damage in Fe single crystal - C15

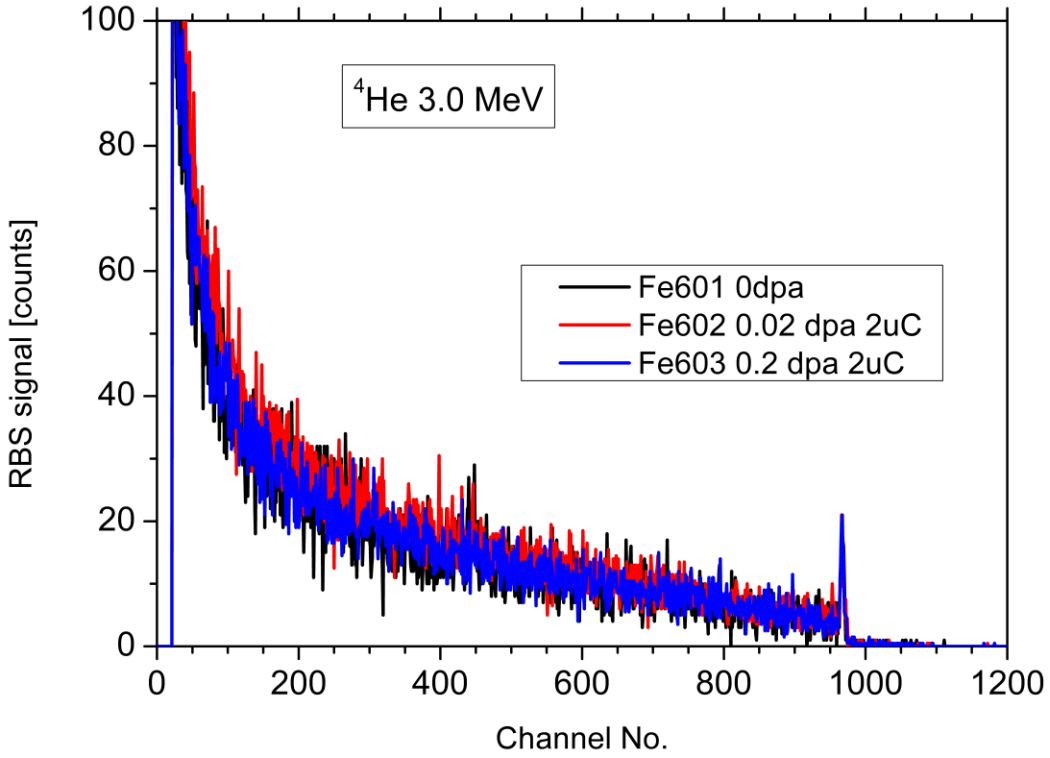
- On the experimental evidence of C15 cluster in Fe – dominant defect at low damage doses

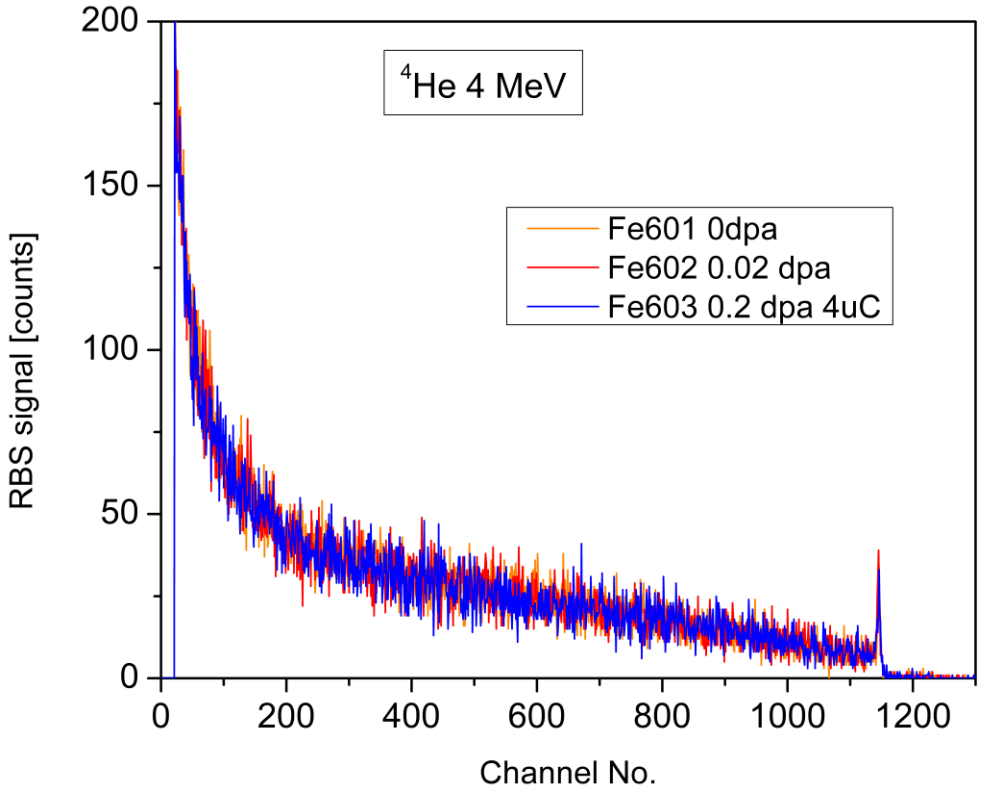


- Irradiate Fe (100) single crystals by Fe ions @ 290 K
  - 0.02 dpa – C15 dominating
  - 0.2 dpa – ½<111> loops dominating

RBS-C on displacement damaged Fe (100)

- On the experimental evidence of C15 cluster in Fe – dominant defect at low damage doses





- No disorder detected
- INTERSTITIAL AND VACANCIES mobile at 290 K – no the case for W – if no impurities (C) are present to stabilize the defects
- MD / DFT calculations at 300 K / 0 K – no defect time evolution
- Next step – irradiation and RBS-C at LN temperatures

Conclusions

- **RBS-C** can provide us a unique way for analyzing the depth distribution of defects.

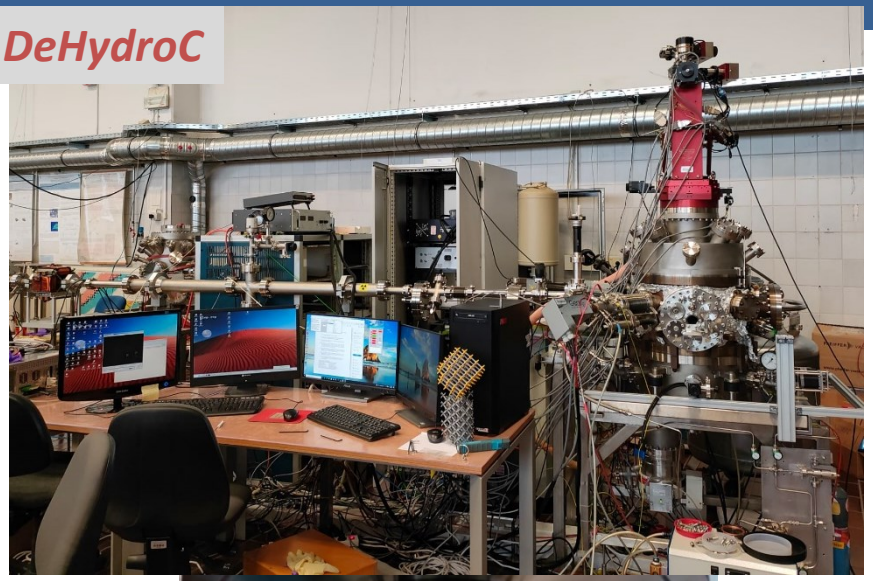
- **Multi-energy RBS-C** is a good tool to study interstitial type of defects in the material (in situ)
  - Combined with RBS-C simulations on W targets, containing realistic defect structures, obtained from molecular dynamics simulations, gives deeper insight into defect structure

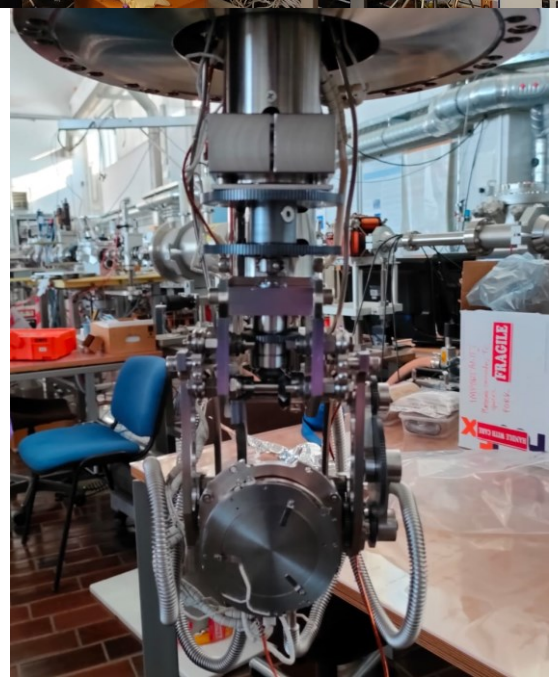
- RBS-C is the method to be used to validate the creation of dislocations by state of the art modelling (test potentials and test if the size of MD cell is adequate)!!

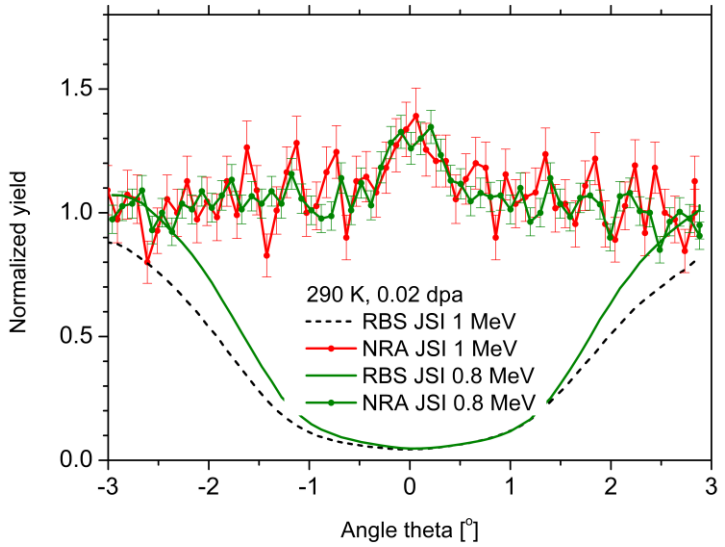
- Only experiment with modelling hand in hand can bring progress in the field

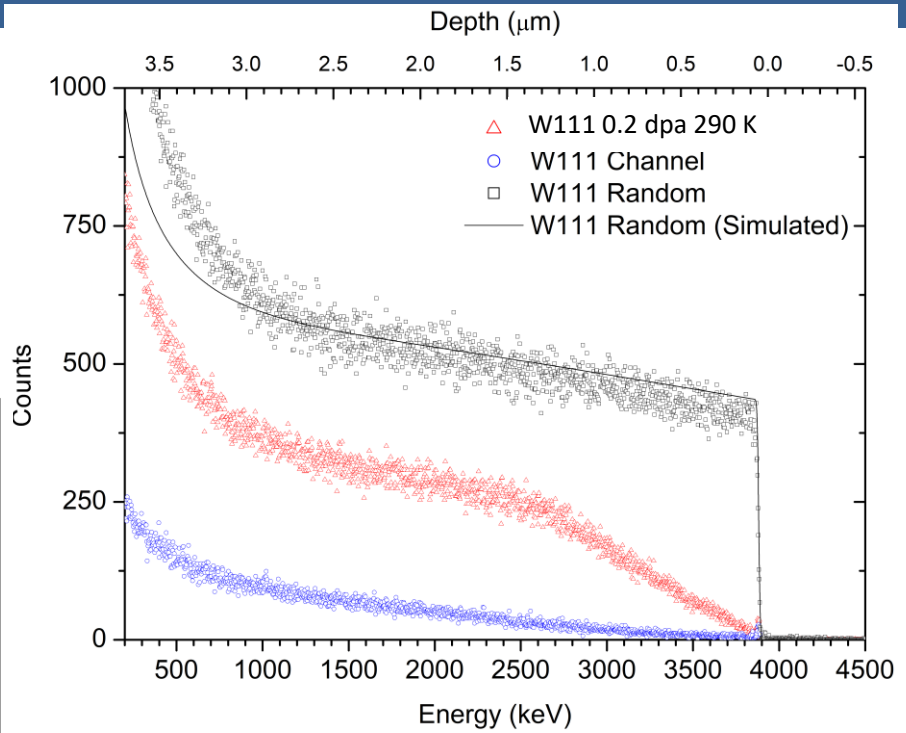
Measurements at JSI with new 6 axis manipulator

DeHydroC









Thank you for your attention