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Atomic scale insights on the microstructure evolution of urania under irradiation

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To cite this version:

A. Chartier, L. van Brutzel, Claire Onofri, C. Sabathier-Devals, O. Dorosh, et al.. Atomic scale insights on the microstructure evolution of urania under irradiation. Nuclear Materials (NuMat - 2016), Nov 2016, Montpellier, France. cea-02438727

HAL Id: cea-02438727

https://cea.hal.science/cea-02438727v1

Submitted on 28 Feb 2020

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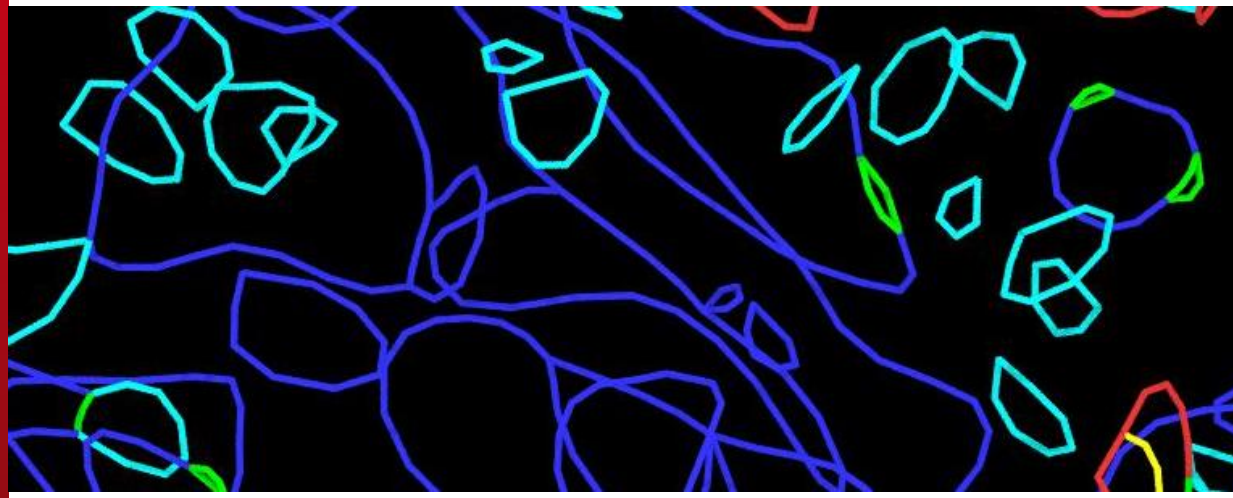
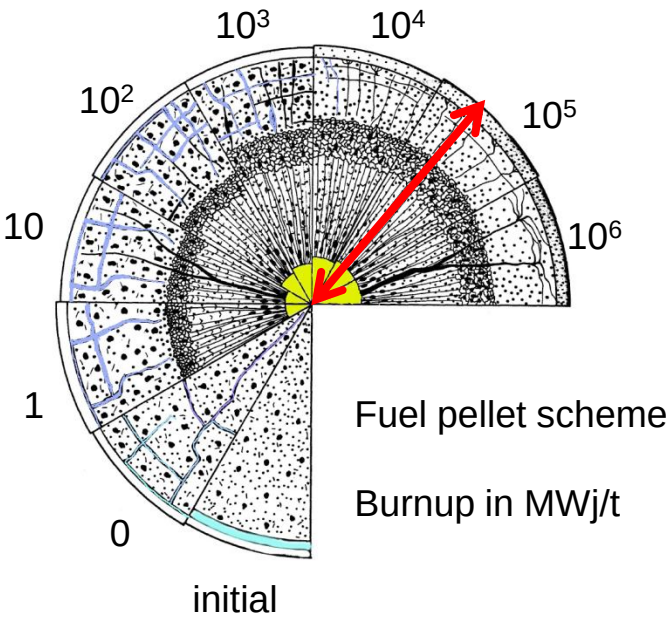
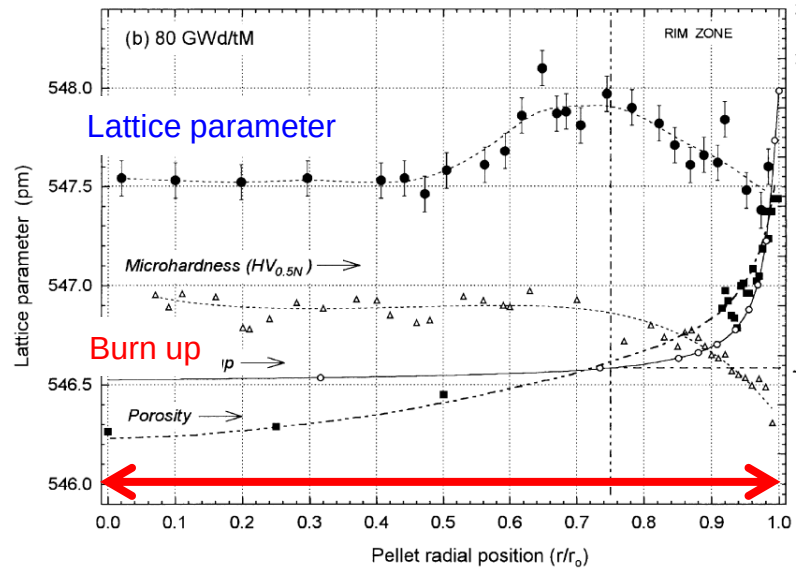
ATOMIC SCALE INSIGHTS ON THE MICROSTRUCTURE EVOLUTION OF URANIA UNDER IRRADIATION

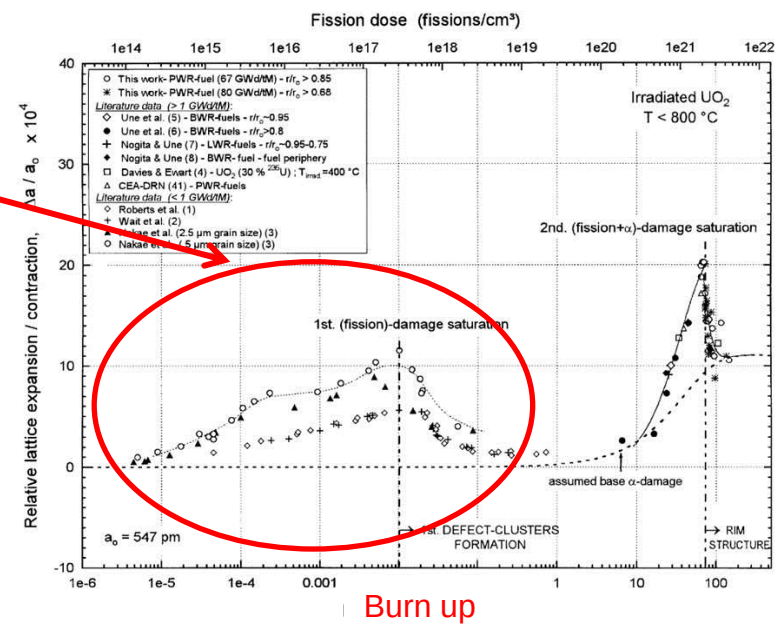


Alain Chartier, Laurent Van Brutzel, Claire Onofri, Catherine Sabathier, Orest Dorosh and Jacek Jagielski

UO_2 nuclear fuel microstructure evolution







First dilatation-contraction peak:

1. point defects induce dilatation
2. Clustering and dislocations induce contraction

Rationale from:

- XRD mapped on rate equations
- TEM observations
- Ion irradiation experiments
- Mesoscopic modelling

But no direct insight of **the early stages ← our goal**

Primary damage in UO_2

Molecular dynamics

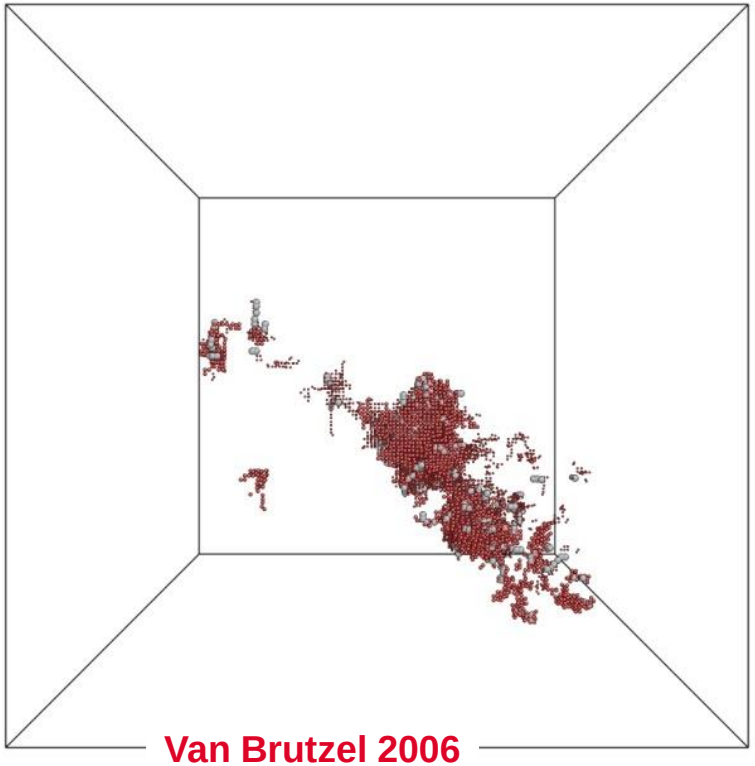
Code: LAMMPS

Empirical potentials: Morelon

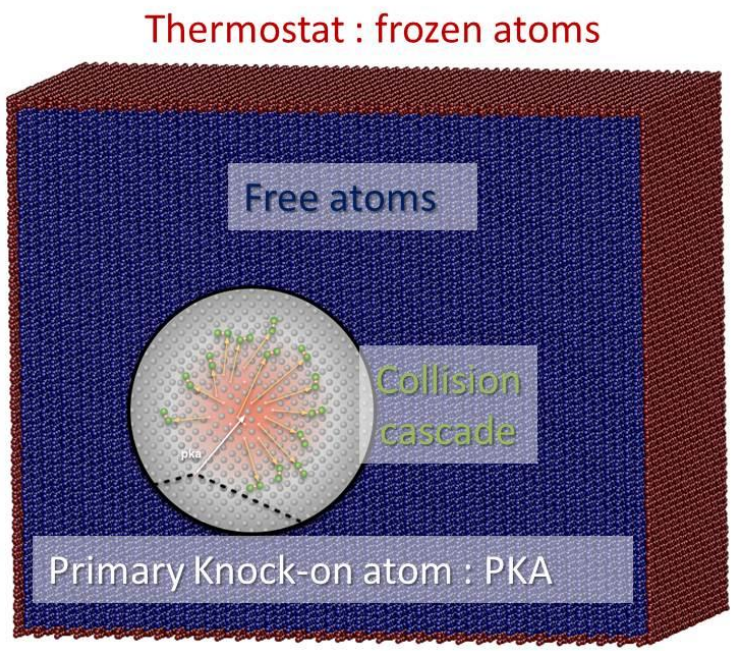
Displacement cascades:

Primary Knock-on Atom: U

E_{PKA} = 80 keV



Van Brutzel 2006



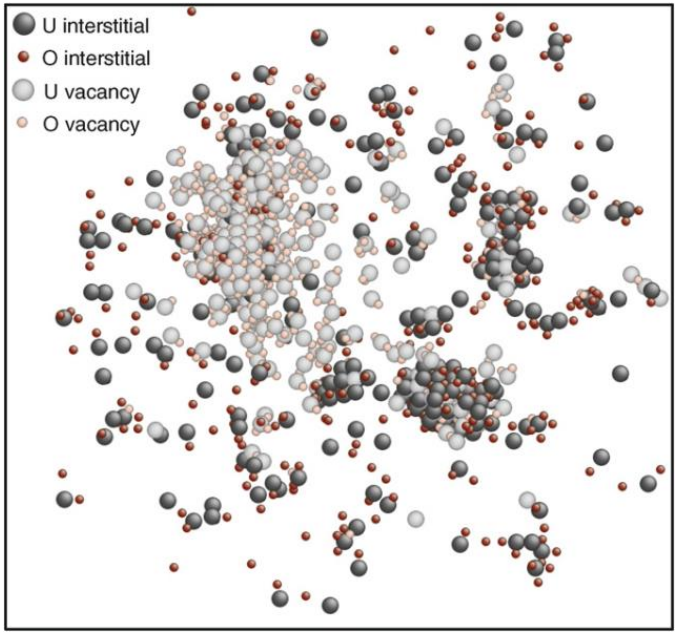
- no direct impact amorphisation

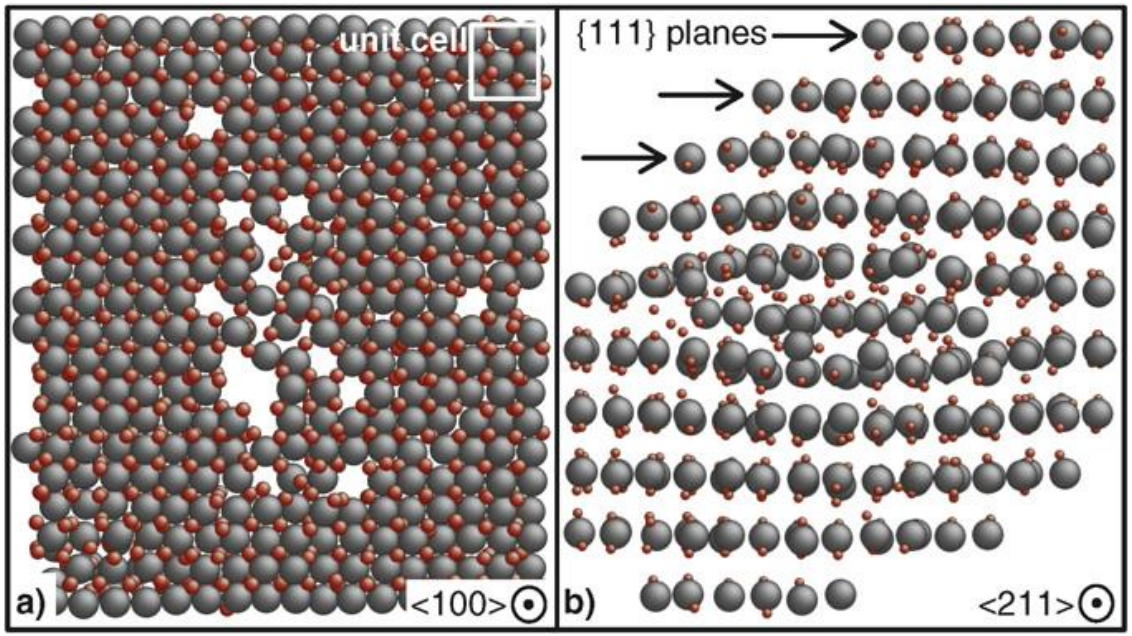
- small defects (interstitials / vacancies)

Primary damage in UO_2

Cascades overlap: E_{PKA} = 10 keV and 36 cascades

Martin 2010





Small defects

Cluster of vacancies

Dislocations

Dose effect modeling

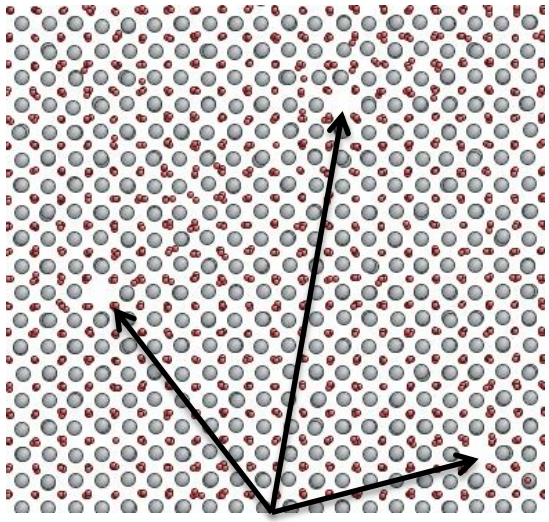
Dose effect: defects accumulation

Creation of Frenkel pairs every 2 ps

Dose rate: 1.6 10^9 dpa/s

Dose: → 10 dpa

80 – 300 – 900 K - with constant pressure and temperature



Frenkel pairs

Analysis:

Dislocations with DXA algorithm

Defects with Voronoï

Lattice expansion – contraction from XRD

HRTEM and SAED

Experiments versus calculations:

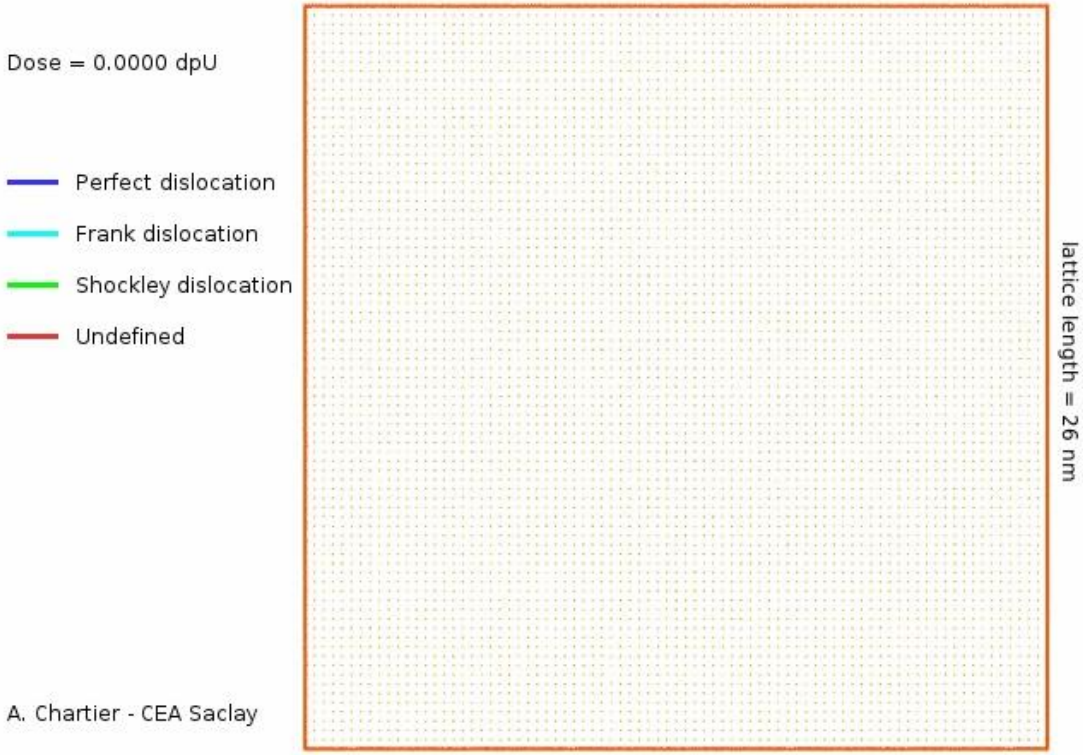
Dose rate : 10^{-3} dpa/s → 34 Å between two displaced Uranium after 1 second

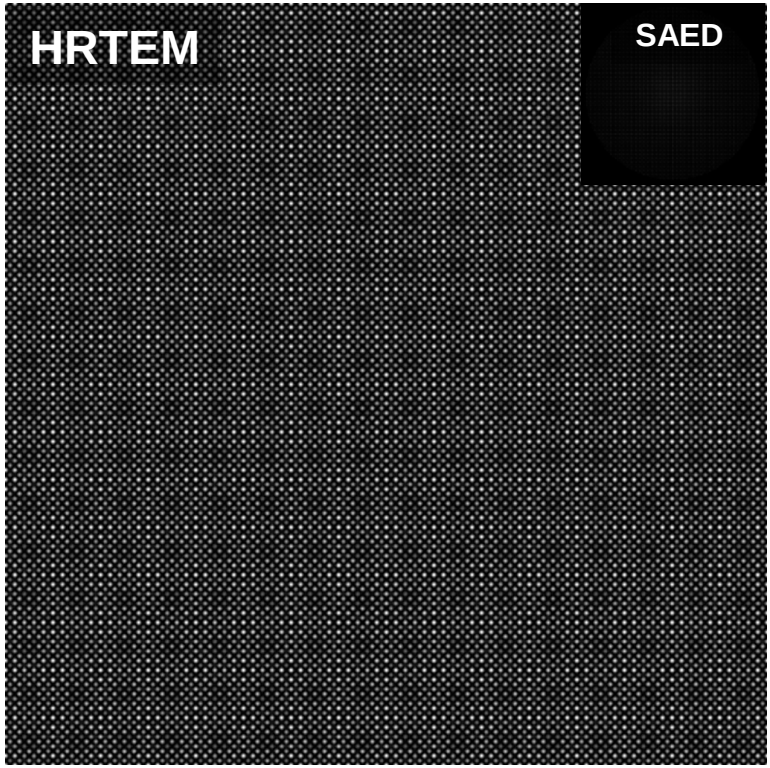
D(U at 900K) = 10^{-31} m^2/s -> 1.18 10^{14}s for travelling 34 Å

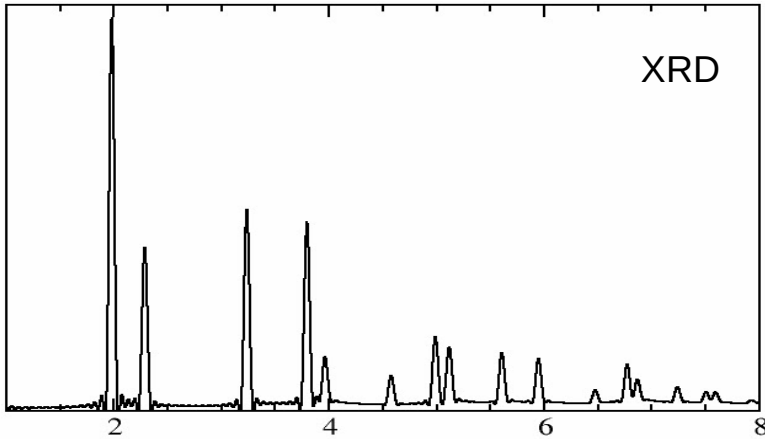
-> **recombination regime !**

Dislocation rescaling (Rest 2004) with $\rho_d \propto {K_0}^{1/6} \rightarrow \rho_{norm} \approx 9.3 \times 10^{-3}\ \rho_{calc.}$

Dose effect from MD - Analysis from curvature criterion, HRTEM, SAED and XRD calculations







→ Vizualization of dislocations – MD and HRTEM

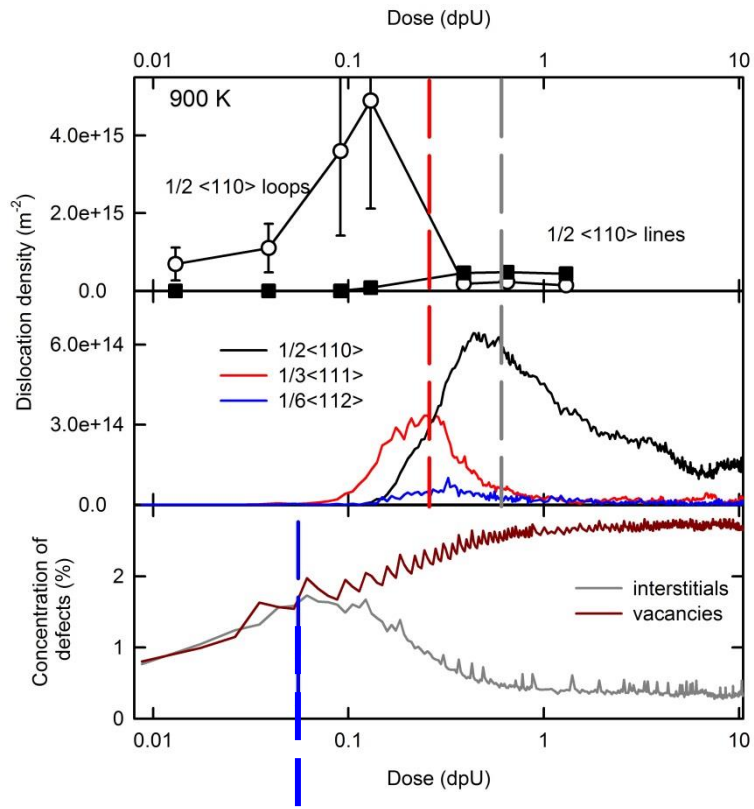
→ Nanodomains – MD and HRTEM

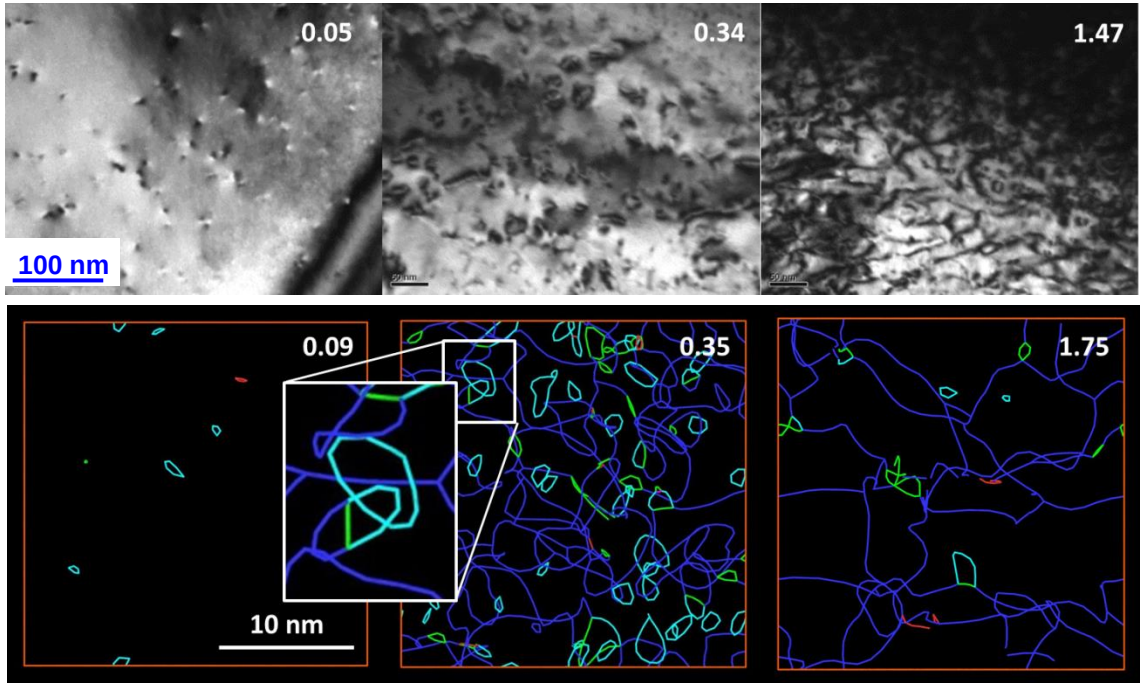
→ Dislocation density – MD

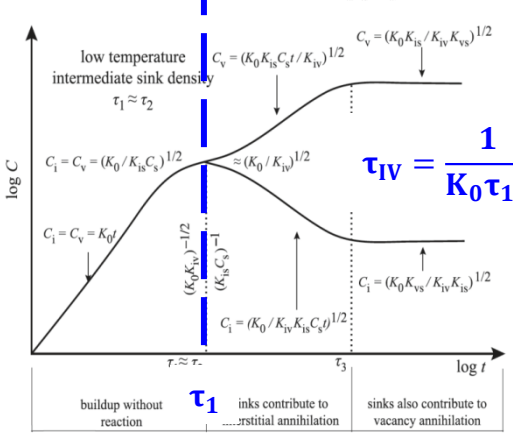
→ Swelling from size of box – XRD

→ no amorphization – SAED and XRD

Irradiation induced dislocations in UO_2







$$\tau_{IV} = \frac{1}{K_0 \tau_1^2} = 1.6 \text{ ps}$$

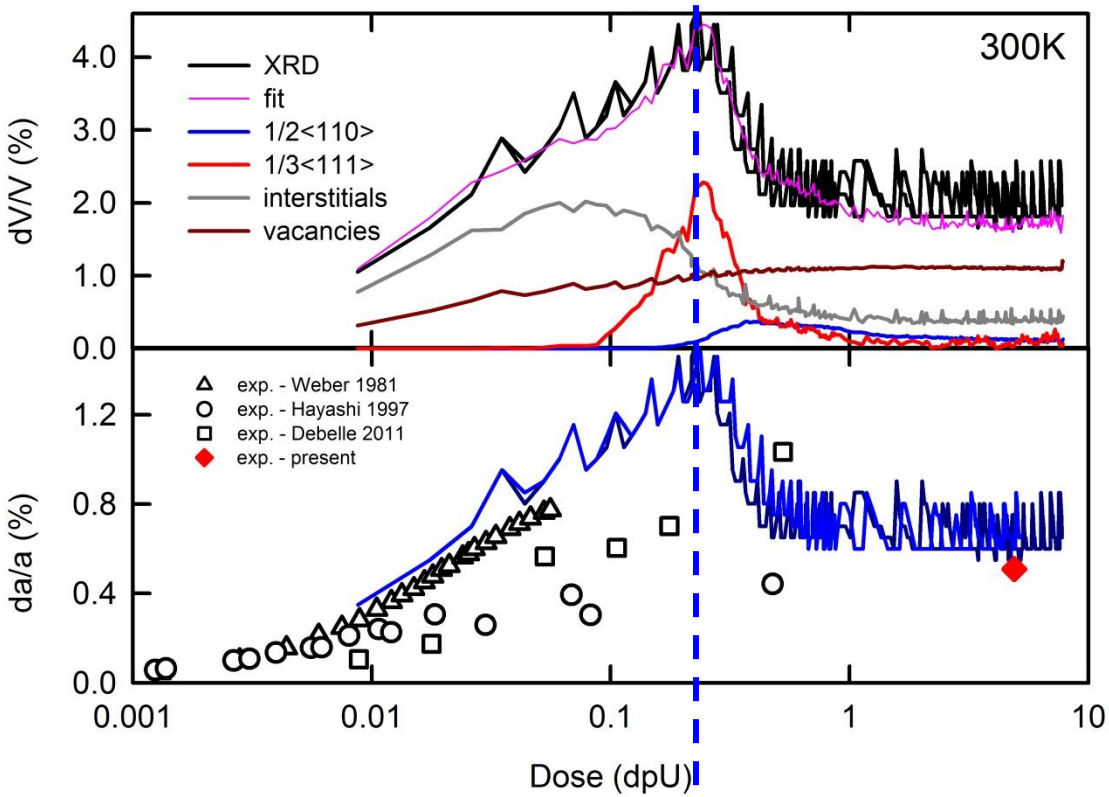
Exp: *not seen ↔ smaller than 2 nm* **½ <110> loops → ½ <110> lines**

MD: point defects → ⅓<111> loops → ½ <110> loops → ½ <110> lines
Frank loops perfect loops

dislocation density: MD = 4 10^{14} m^{-2} and Exp. = 6 10^{14} m^{-2} (rescaled)

Nota:
- point defects behave as expected from rate equations
- 1.6 ps corresponds to spontaneous recombination regime (Van Brutzel 2008)

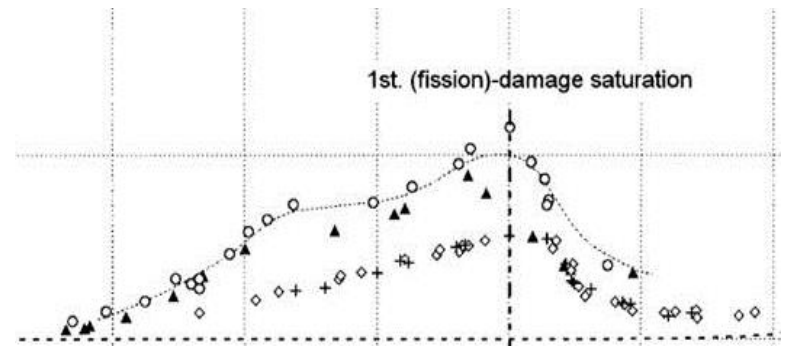
Lattice evolution with irradiation dose



$$\frac{\Delta V}{V} = V_I^f \times \rho_{line} + \frac{\overline{R_{loop}} \times b}{2} \times \rho_{loop} + \frac{V_{rel}^I}{\Omega} \times C_I + \frac{V_{rel}^V}{\Omega} \times C_V$$

$$\text{with } V_I^f = 5.6\ \text{Å}^2;\ \overline{R_{loop}} \sim 3\ \text{nm}\ ;\ V_{rel}^I = V_{rel}^V = 1.\Omega$$

Lattice expansion - contraction comparable to measurements on nuclear fuel



Expansion:

- interstitials and vacancies
- 1/3<111> Frank loops

Contraction:

- transformation of Frank loops into perfect loops
- vacancies at steady state

Summary - Conclusion

Mechanism of irradiation induced dislocations in UO_2:

- Molecular dynamics: point defects → ⅓<111> loops → $\frac{1}{2}$ <110> loops → $\frac{1}{2}$ <110> lines
- Experimental observations: *not seen ↔ smaller than 2 nm* $\frac{1}{2}$ <110> loops → $\frac{1}{2}$ <110> lines

Expansion – contraction peak:

- Expansion : point defects and then Frank loops
- Contraction: transformation of Frank loops in unfaulted dislocations.

→ re-interpretation of accepted picture

Recombination regime arguments:

- Comparison between dose rates and characteristic time for diffusion => recombination regime

- Link between MD and exp. dislocation density with renormalisation formula => recombination regime

A. Chartier, C. Onofri, L. Van Brutzel, C. Sabathier, O. Dorosh, and J. Jagielski, Appl. Phys. Lett. in press

Thank you for listening

Questions ?

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