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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-02438727>**

Submitted on 28 Feb 2020

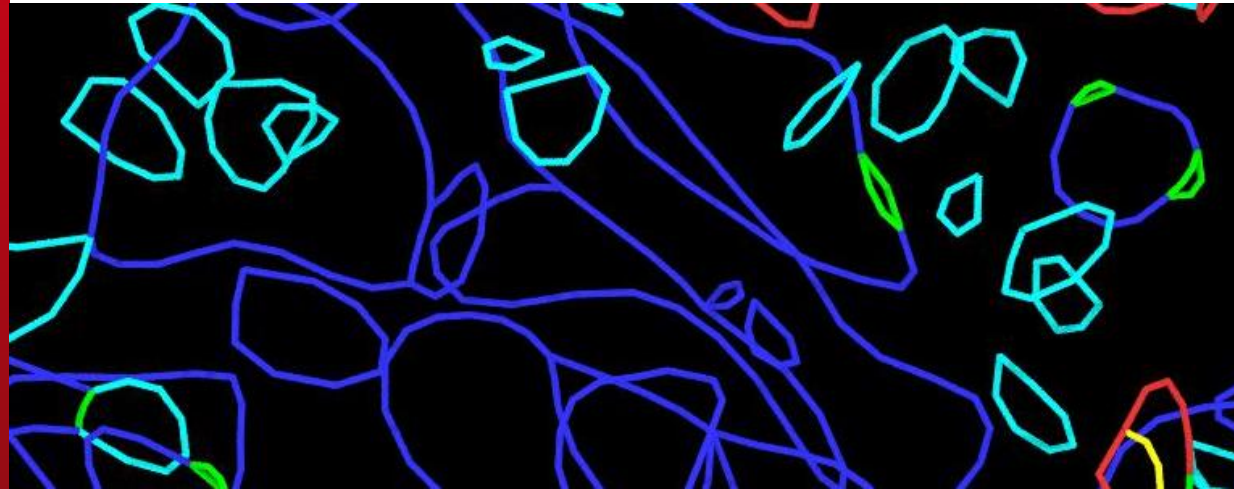
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ATOMIC SCALE INSIGHTS ON THE  
MICROSTRUCTURE EVOLUTION OF URANIA  
UNDER IRRADIATION

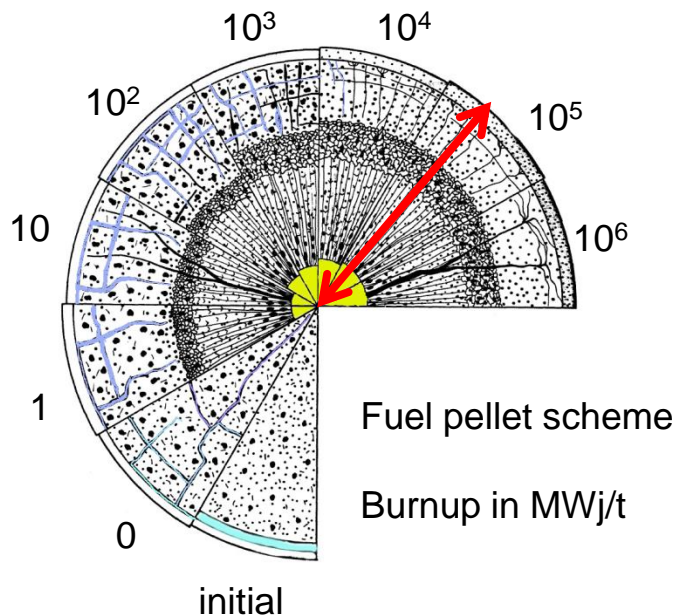
DE LA RECHERCHE À L'INDUSTRIE  
cea den

université  
PARIS-SACLAY



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

# UO<sub>2</sub> nuclear fuel microstructure evolution



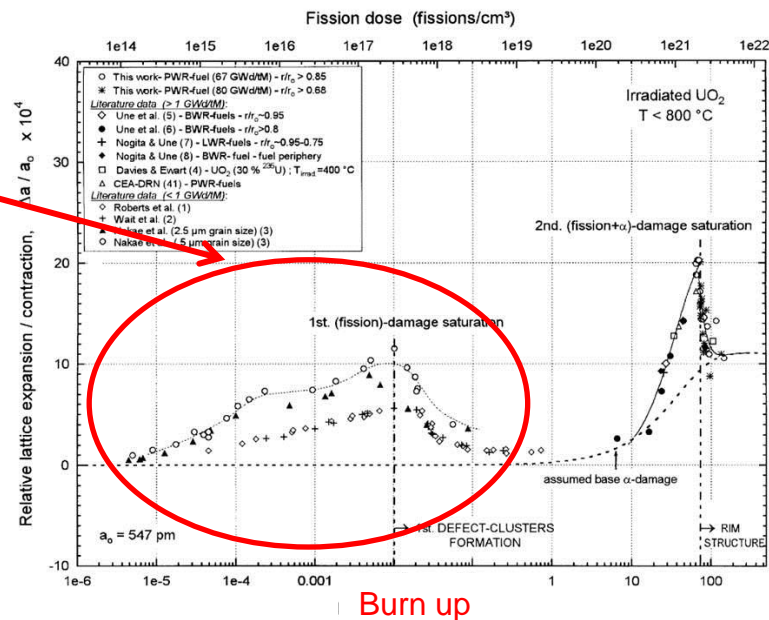
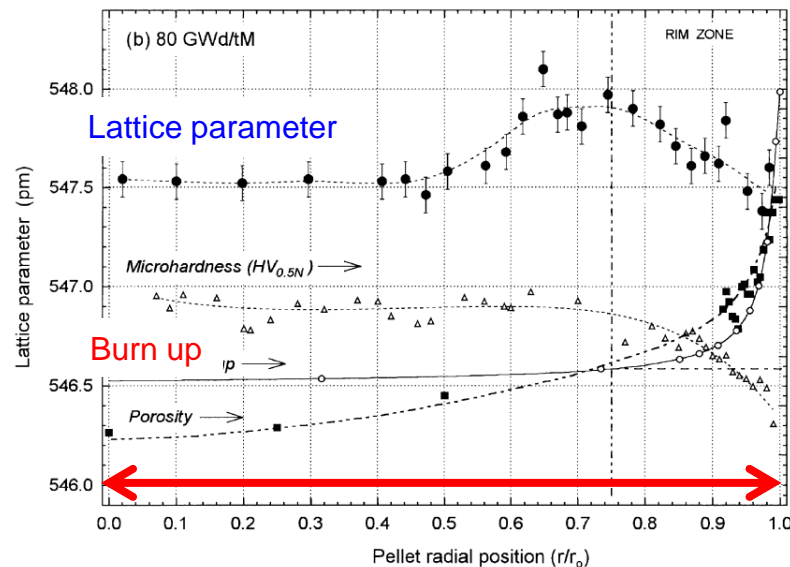
## First dilatation-contraction peak:

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



Relative lattice parameter

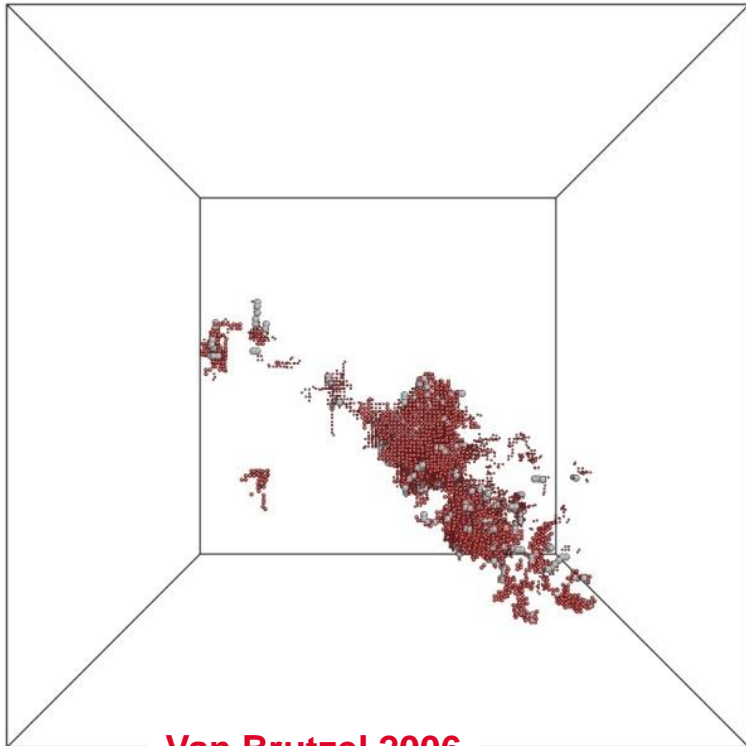
**Molecular dynamics**

Code: LAMMPS

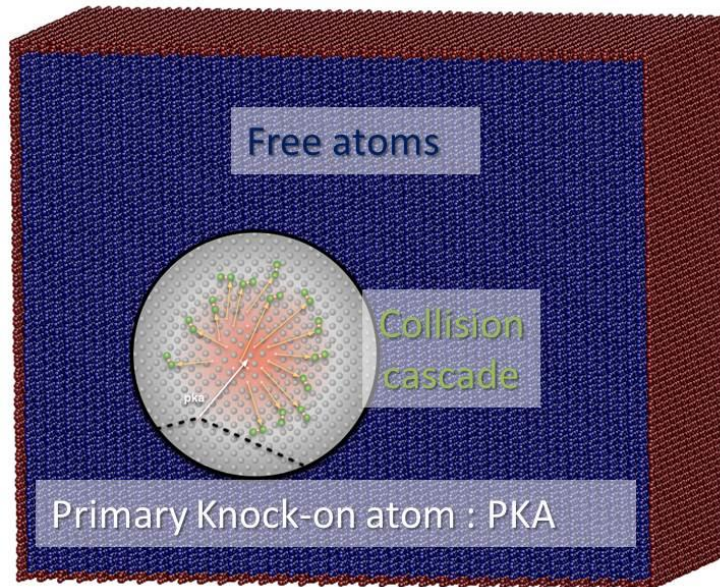
Empirical potentials: Morelon

**Displacement cascades:**

Primary Knock-on Atom: U

 $E_{\text{PKA}} = 80 \text{ keV}$ 

Thermostat : frozen atoms

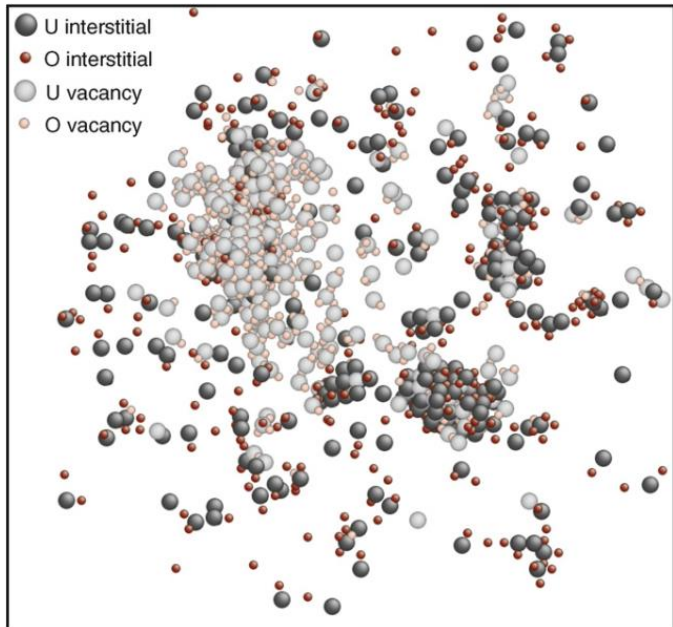


- no direct impact amorphisation
- small defects (interstitials / vacancies)

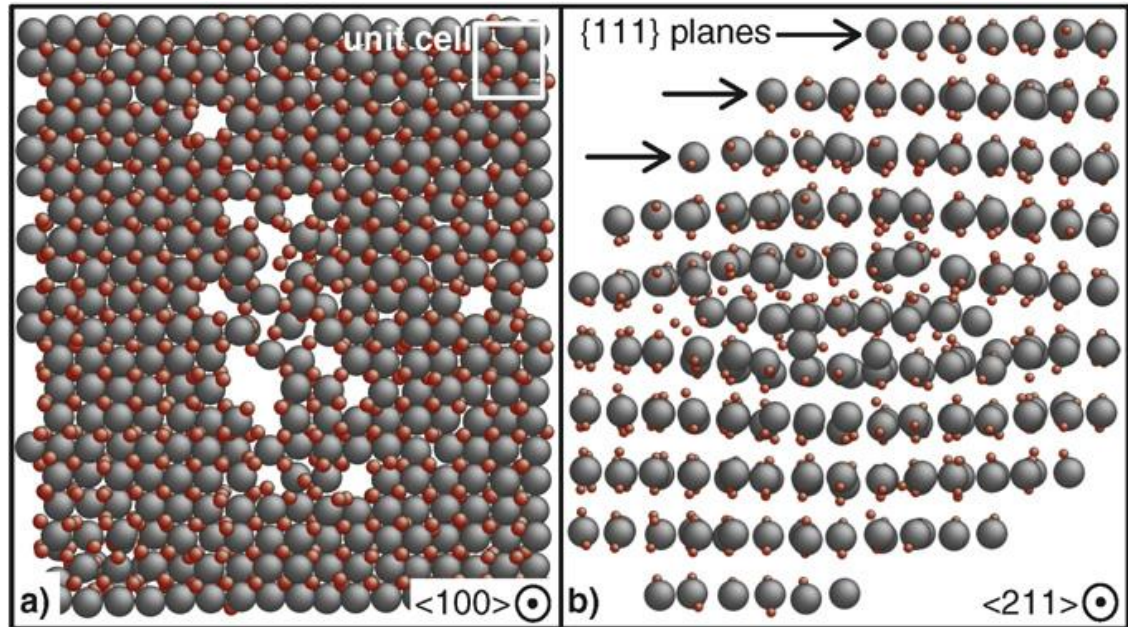
Primary damage in  $\text{UO}_2$ 

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

Martin 2010



Small defects



Cluster of vacancies

Dislocations

**Dose effect:** defects accumulation

Creation of Frenkel pairs every 2 ps

Dose rate:  $1.6 \cdot 10^9$  dpa/s

Dose:  $\rightarrow 10$  dpa

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

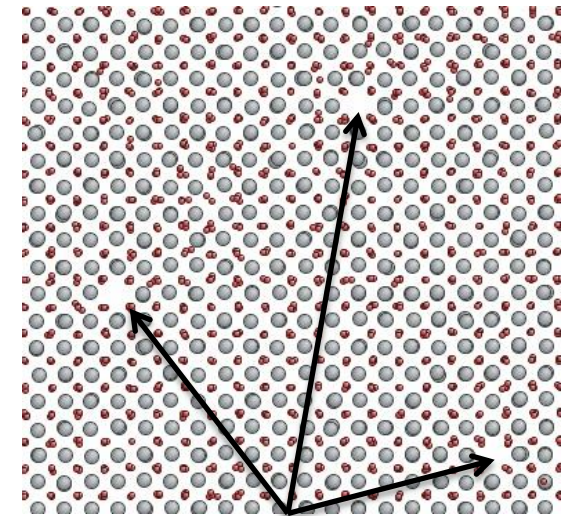
**Analysis:**

Dislocations with DXA algorithm

Defects with Voronoï

Lattice expansion - contraction from XRD

HRTEM and SAED



Frenkel pairs

**Experiments versus calculations:**

Dose rate :  $10^{-3}$  dpa/s  $\rightarrow 34 \text{ \AA}$  between two displaced Uranium after 1 second

$D(\text{U at } 900\text{K}) = 10^{-31} \text{ m}^2/\text{s} \rightarrow 1.18 \cdot 10^{14} \text{ s}$  for travelling  $34 \text{ \AA}$

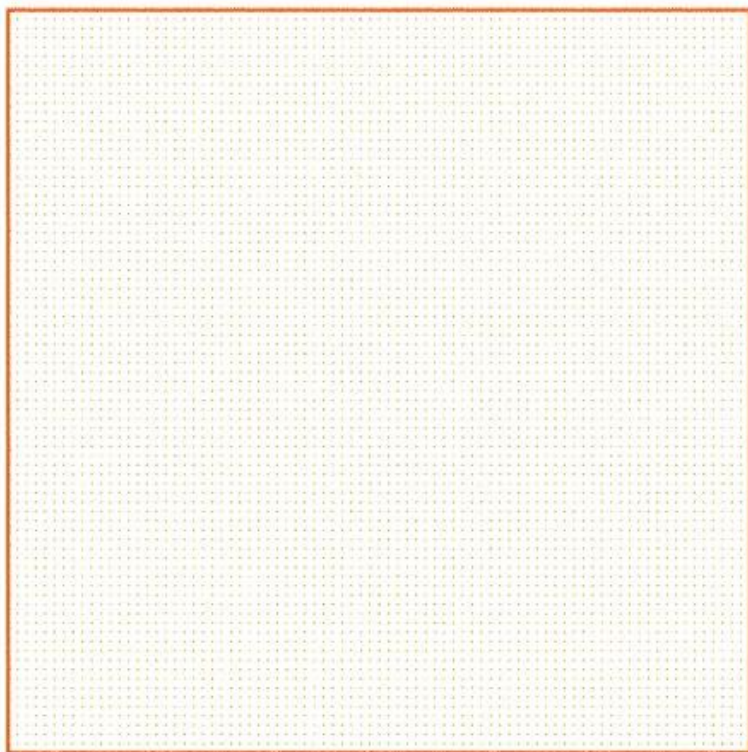
$\rightarrow$  **recombination regime !**

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

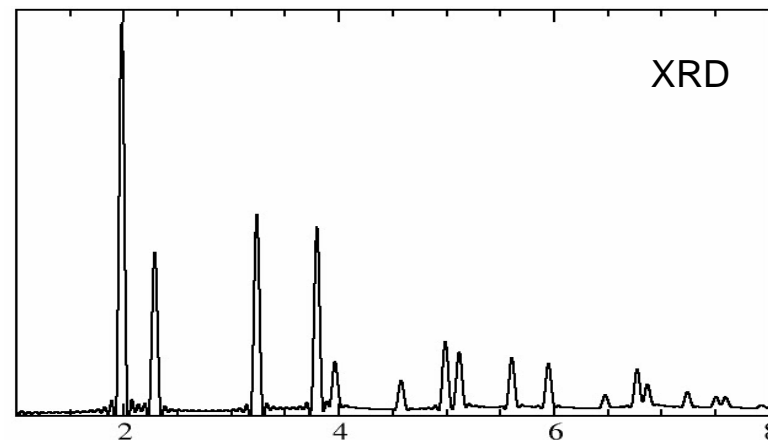
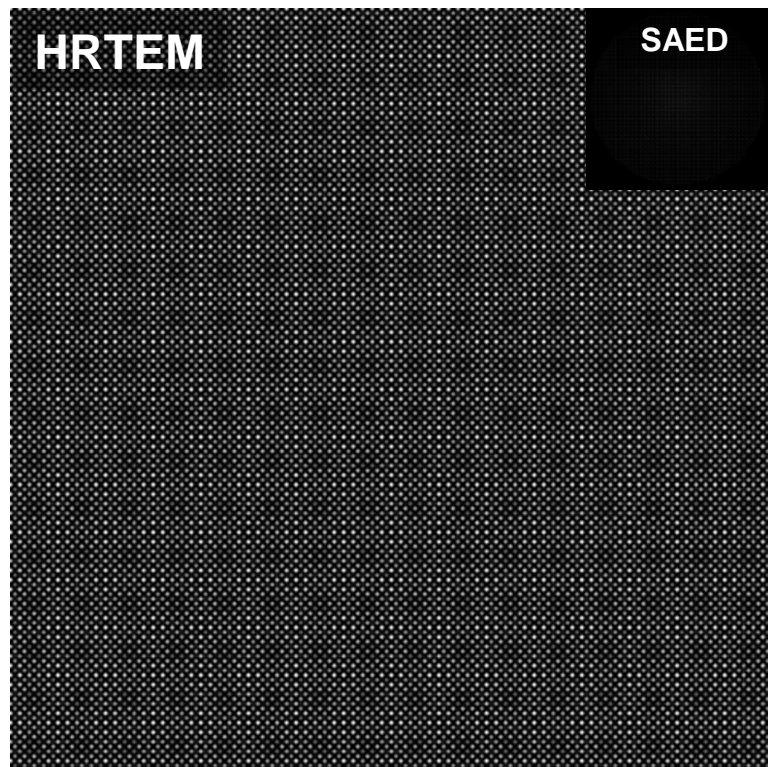
# Dose effect from MD - Analysis from curvature criterion, HRTEM, SAED and XRD calculations

Dose = 0.0000 dpU

- Perfect dislocation
- Frank dislocation
- Shockley dislocation
- Undefined

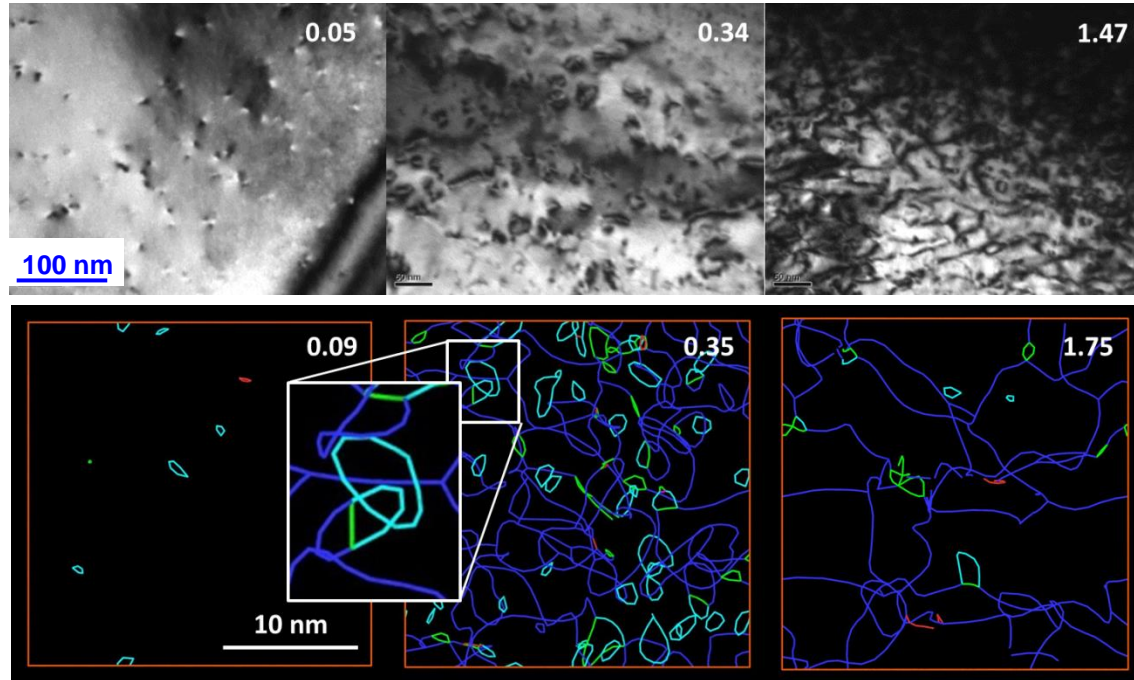
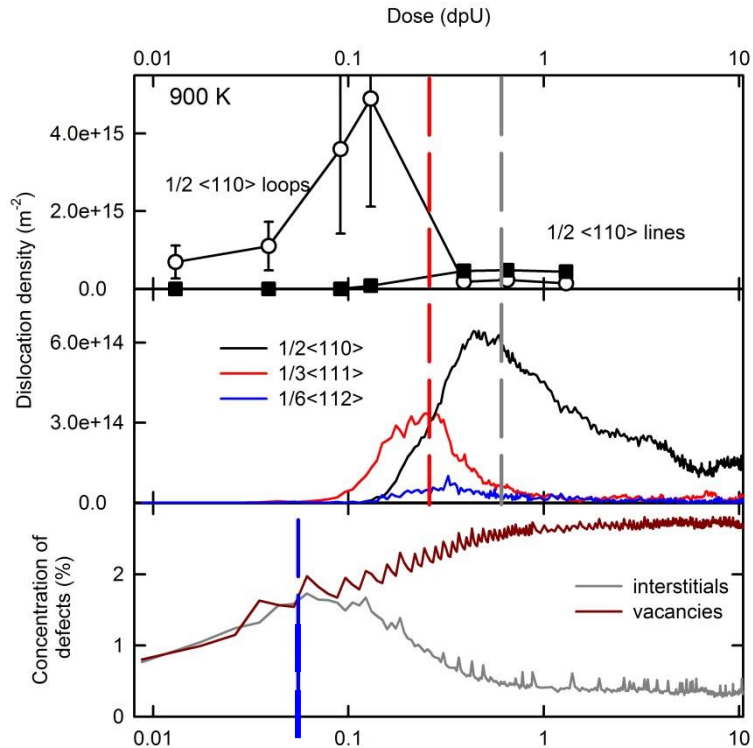


A. Chartier - CEA Saclay



- Visualization 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<sub>2</sub>



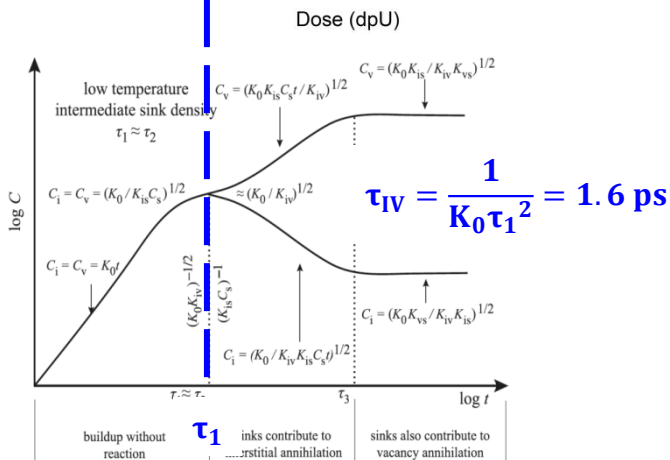
Exp: *not seen* ↔ *smaller than 2 nm*      1/2 <110> loops → 1/2 <110> lines

MD: point defects → 1/3 <111> loops → 1/2 <110> loops → 1/2 <110> lines  
 Frank loops      perfect loops

dislocation density: MD = 4 10<sup>14</sup> m<sup>-2</sup> and Exp. = 6 10<sup>14</sup> m<sup>-2</sup> (rescaled)

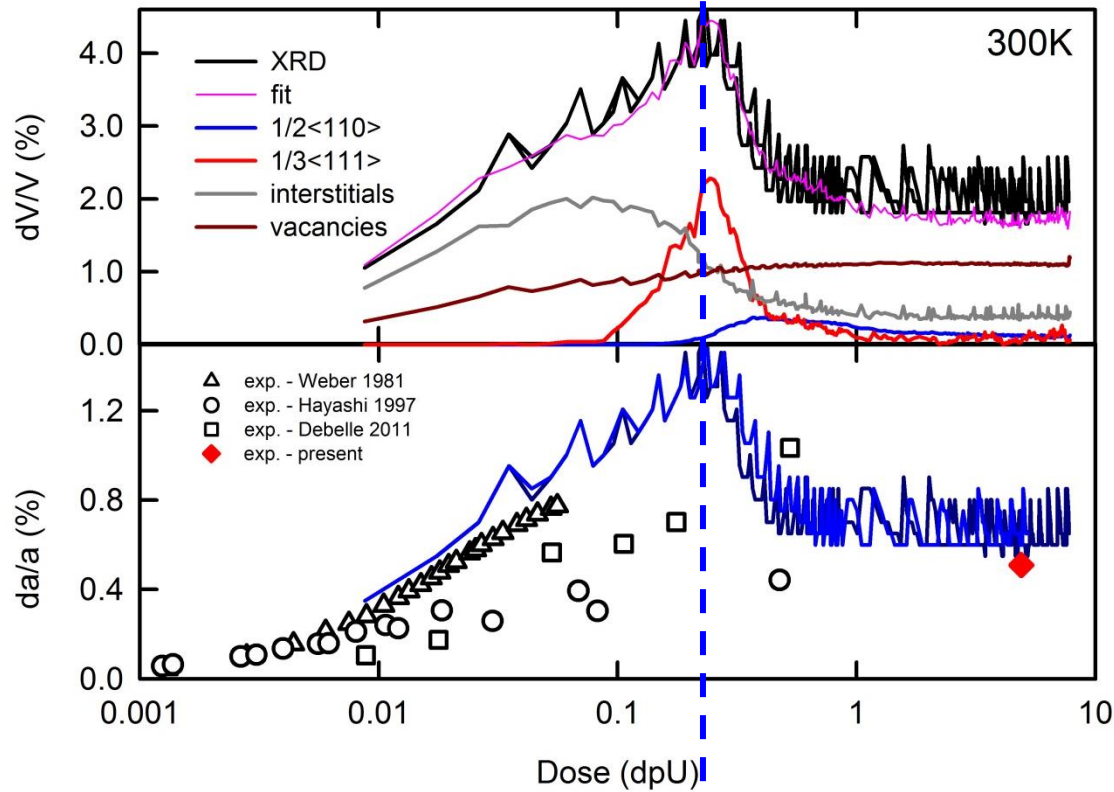
Nota:

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





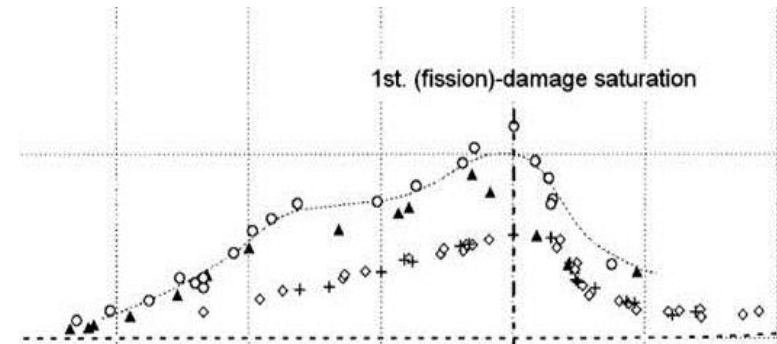
# Lattice evolution with irradiation dose



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

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

Lattice expansion - contraction comparable to measurements on nuclear fuel



## Expansion:

- interstitials and vacancies
- $1/3\langle 111 \rangle$  Frank loops

## Contraction:

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

### Mechanism of irradiation induced dislocations in $\text{UO}_2$ :

- Molecular dynamics: point defects  $\rightarrow \frac{1}{3}\langle 111 \rangle$  loops  $\rightarrow \frac{1}{2}\langle 110 \rangle$  loops  $\rightarrow \frac{1}{2}\langle 110 \rangle$  lines
- Experimental observations: *not seen*  $\leftrightarrow$  *smaller than 2 nm*  $\frac{1}{2}\langle 110 \rangle$  loops  $\rightarrow \frac{1}{2}\langle 110 \rangle$  lines

### Expansion - contraction peak:

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

$\rightarrow$  re-interpretation of accepted picture

### Recombination regime arguments:

- Comparison between dose rates and characteristic time for diffusion  $\Rightarrow$  recombination regime
- Link between MD and exp. dislocation density with renormalisation formula  $\Rightarrow$  recombination regime

# Thank you for listening

## Questions ?

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