Colloque GANIL 2023

Irradiation-induced Effects in Nuclear Materials

Case Study of Nuclear Fuels

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EXPERIMENTAL SIMULATION USING ION BEAMS

Irradiation effects
- Fission fragments
- α particles
- Recoil nucleus
- Radiolysis

Irradiation

Low energy ions (100 KeV)

High energy ions (100 MeV)

Nuclear Materials

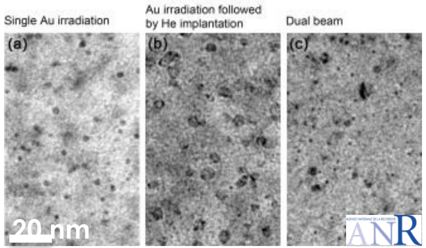
Doping

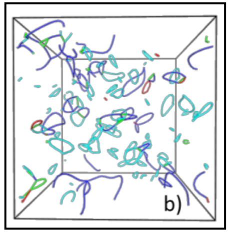
Stables elements

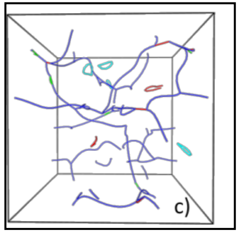
Radioactive elements

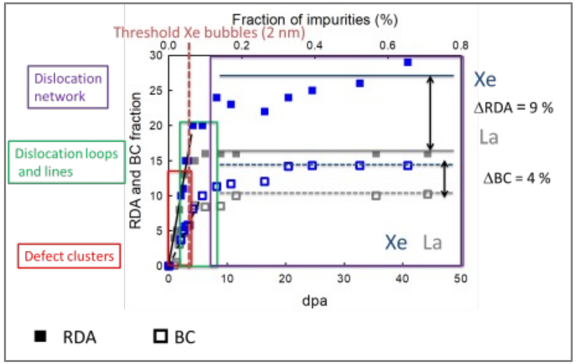
Radionuclides retention
- Actinides
- Fission products, He, H

Nano-precipitates synthesis



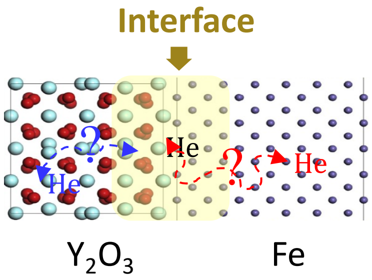


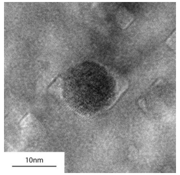




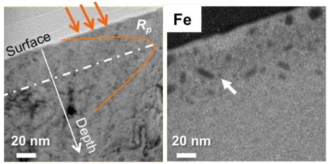
Characterization (*ex* & *in situ*)

RBS, ion channelling, NRA, diffractions, TEM, Spectroscopies, …





Irradiation-induced defects, atomic diffusion, physical and chemical properties,… using ion-beams platforms (GANIL, MOSAIC, …)



SCIENTIFIC GOALS

Better understanding of the behaviour of nuclear materials using energetic ion beams

- Irradiation-induced effects: radiation defects, (micro)-structural transformations
- Role played by embedded impurities: fission products, gases (He, Kr, Xe) – lattice location and behaviour (T, irradiation, solubility, chemical interaction)

Ultra simplified system: single crystals

Parametric approach

Coupling of experiments and computational tools

Better understanding of the nuclear materials using energetic ion beams: towards the *Modelling of Radiation-Induced Effects in Nuclear Materials*

EXPERIMENTAL SIMULATION OF IRRADIATION INDUCED EFFECTS

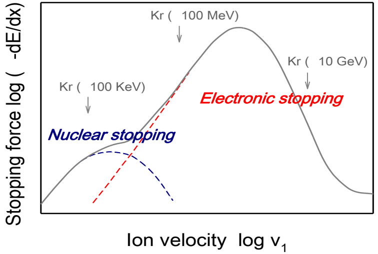
Nuclear fuel exhibits the fluorite-type structure

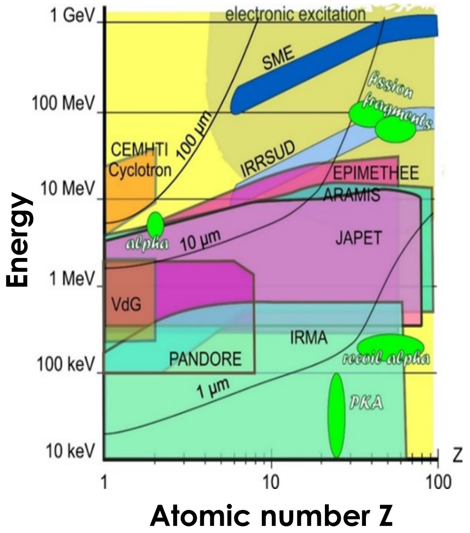
- Structure of UO_2, PuO_2 (MOX) and in reactor transmutation matrices: (Zr, *An*)O_2

Single crystals as a simplified model of nuclear fuel or transmutation matrix

Simulation of radiation-induced damage

- Atomic collisions (MOSAIC platform IJCLab); electronic excitations (GANIL)
- Doping – chemical contribution – role played by soluble versus insoluble specie (fission products)





EXPERIMENTAL SIMULATION OF IRRADIATION INDUCED EFFECTS

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Investigating the High Burnup Structure of spent nuclear fuels

- Microstructural phase transformation occurring at the rim of fuel pellets: grain subdivision (100 nm) and high porosity
- Related to the local enrichment in ^{239}Pu (neutron capture cross section in the resonance region up to 1 eV)
- Atomic mechanisms not understood; possible parameters include low T, higher concentration of impurities, radiation damage (atomic and electronic)
- Parametric approach: burnup, T, chemistry of impurities, radiation defects

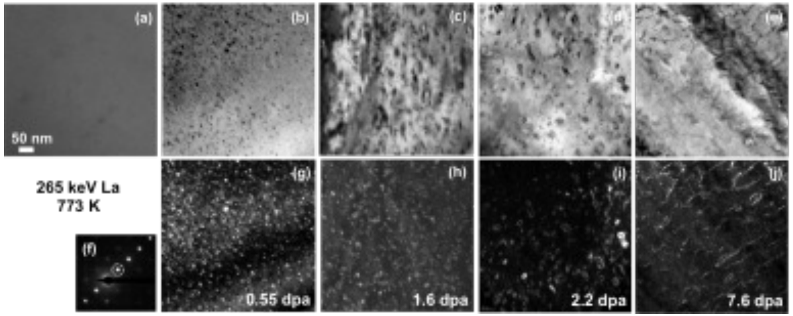
In situ channelling and TEM experiments coupled to implantation at 773 K – Role of insoluble versus soluble fission products

Swift ion irradiation coupled to channelling and TEM experiments – Role of electronic stopping

EXPERIMENTAL SIMULATION OF IRRADIATION INDUCED EFFECTS

In situ channelling and TEM experiments coupled to implantation at 773 K – Role of insoluble versus soluble fission products

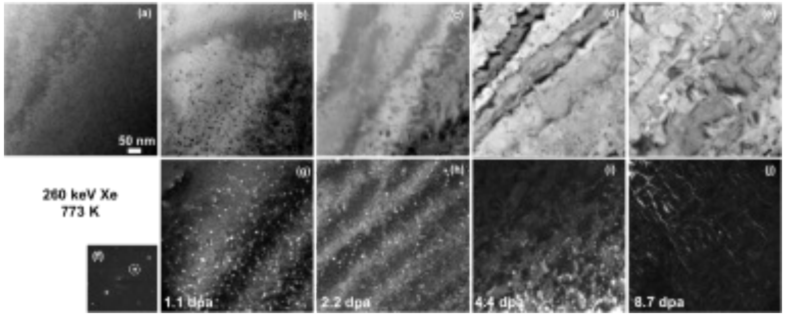
- Direct comparison between the fate of Xe and La (Z =54, insoluble in UO_2; Z = 57, fully soluble) - In situ TEM evolution (images recorded at 773 K)



Black dots
6-8 nm

Dislocation loop growth
form 13 to 34 nm

Merging of
dislocation lines



Black dots
6-8 nm

Black dots
Some loops

Dislocation
loop growth
Some
dislocation
lines

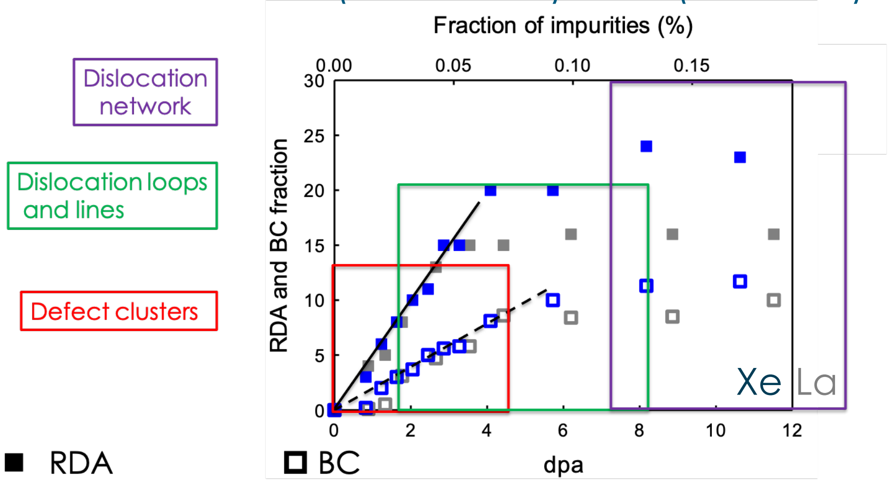
Merging of
dislocation lines

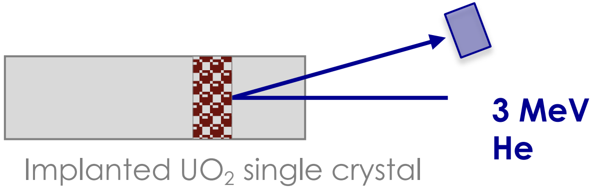
- Same evolution for both specie: sequential evolution from black dots to dislocation loops and lines

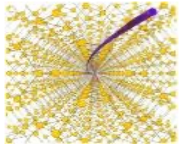
EXPERIMENTAL SIMULATION OF IRRADIATION INDUCED EFFECTS

In situ channelling and TEM experiments coupled to implantation at 773 K – Role of insoluble versus soluble fission products

- Direct comparison between the fate of Xe and La (Z =54, insoluble in UO_2; Z = 57, fully soluble) - In situ channelling evolution
- Defect model: RDA (obstruction) & BC (distortion)







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mosaic
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Energy & Environment

In situ channelling and TEM experiments coupled to implantation at 773 K – Role of insoluble versus soluble fission products

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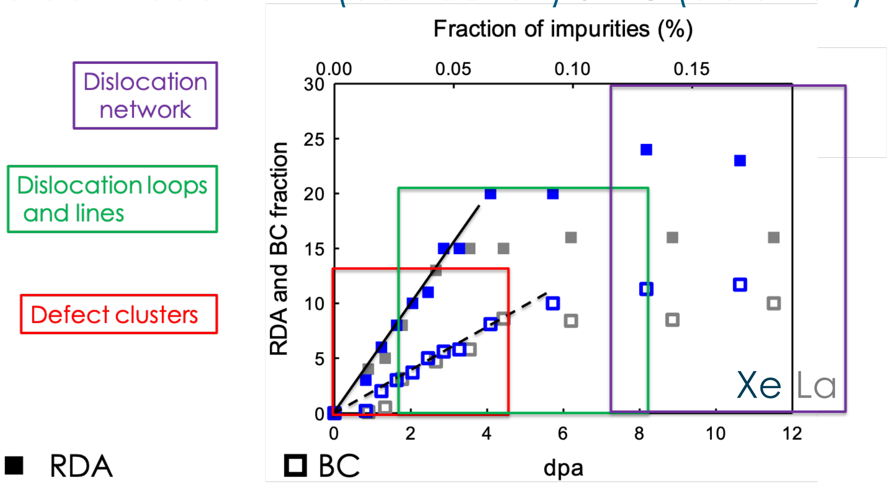

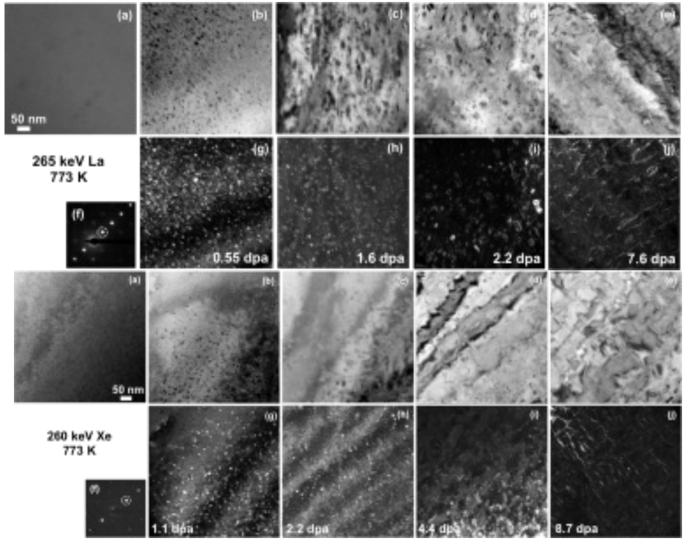

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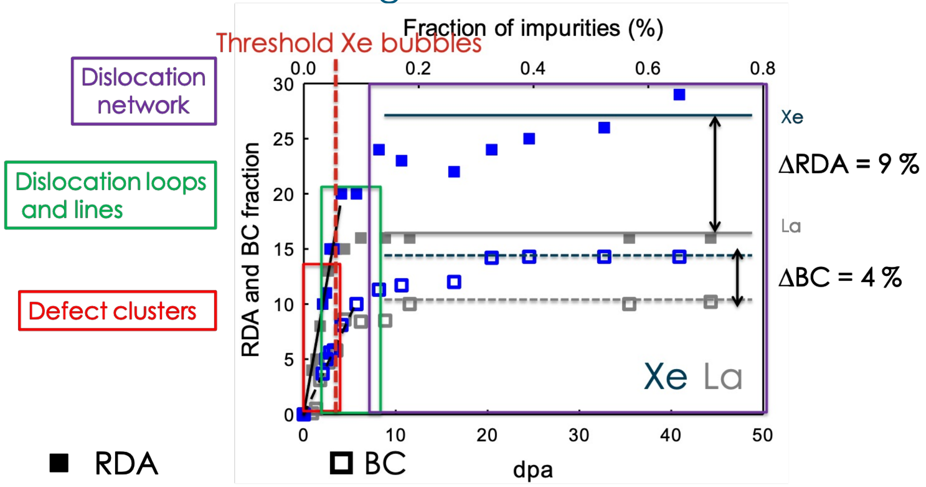
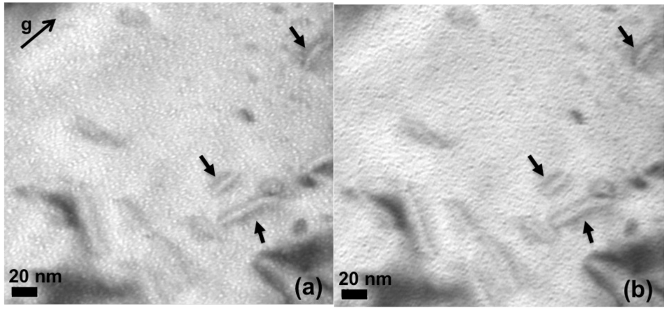
In situ channelling and TEM experiments coupled to implantation at 773 K – Role of insoluble versus soluble fission products

- Direct comparison between the fate of Xe and La (Z =54, insoluble in UO_2; Z = 57, fully soluble) - In situ evolution at 773 K
- Channelling



- TEM



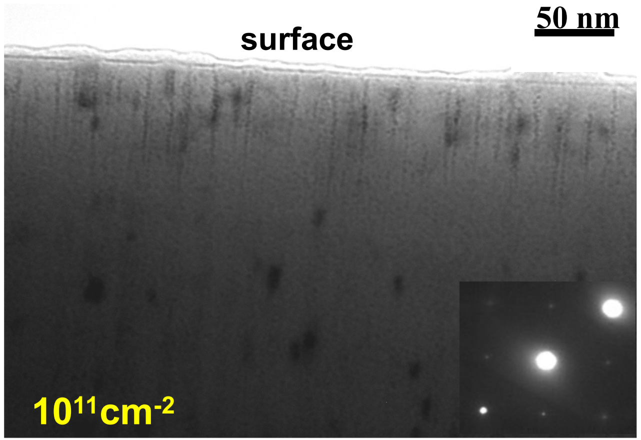
Images recorded in situ at 773 K at 5 dpa; mean bubble size is (2.0 ± 0.5) nm

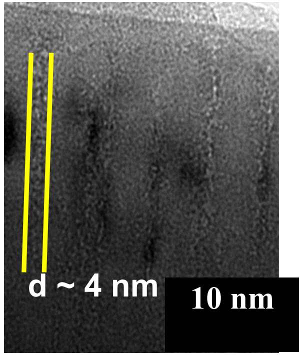
- Formation of Xe bubbles homogeneously distributed (no bubble or cavity for La)

EXPERIMENTAL SIMULATION OF IRRADIATION INDUCED EFFECTS

Swift ion irradiation coupled to channelling and TEM experiments – Investigating the role of electronic stopping

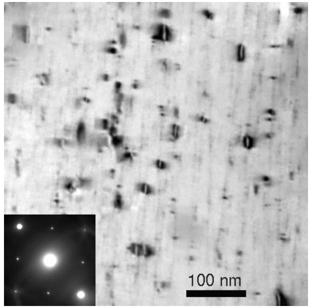
- Low fluence range – Track formation (1 GeV Pb ions SME beam line)





zoom (×5)

Hollowed tracks at surface



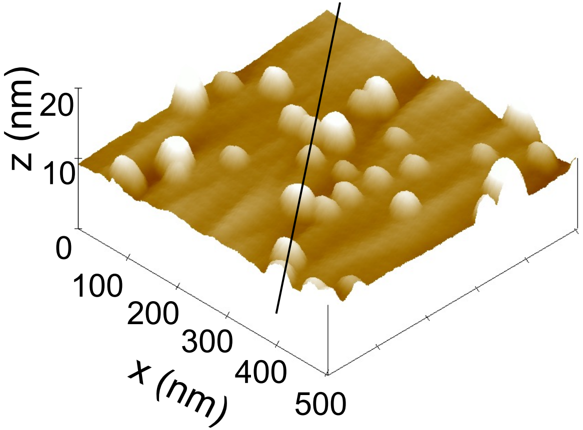
Filled tracks and dislocation loops along tracks

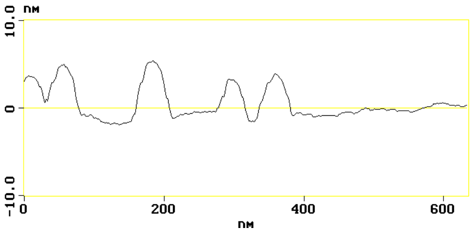
- Regular tracks in the crystal bulk (d ~ 4 nm)
- Strong strain fields in the vicinity of tracks with presence of dislocation loops
- Evidence of 'hollowed' tracks in the crystal *surface* region (spatial extension ~ 100 nm)

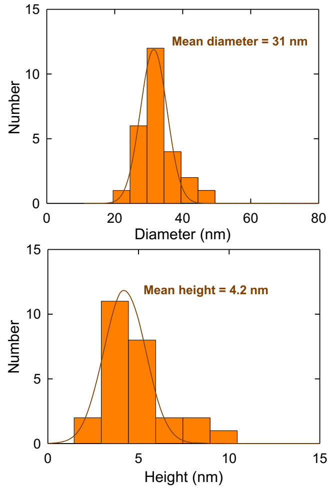
EXPERIMENTAL SIMULATION OF IRRADIATION INDUCED EFFECTS

Swift ion irradiation coupled to channelling and TEM experiments – Investigating the role of electronic stopping

- Low fluence range – Track formation (1 GeV Pb ions)





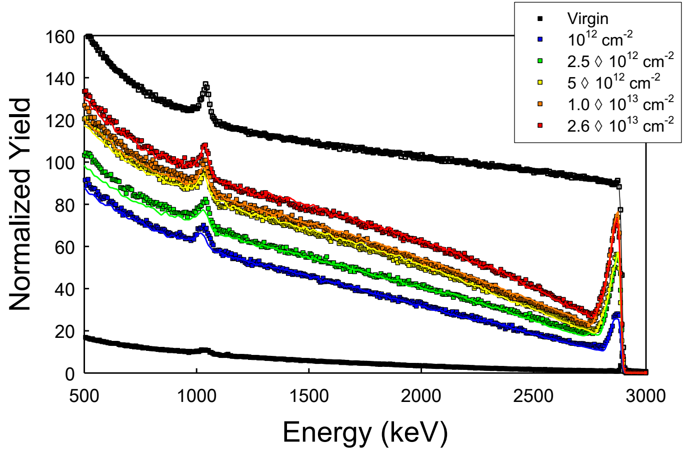


- Large hillocks at the surface (d ~ 30 nm)
- Discrepancy may be explained assuming a thermal spike model: ‘eruption’ of matter from the inner track at the crystal surface (missing volume of matter in hollowed tracks compatible with the volume of ejected matter)

EXPERIMENTAL SIMULATION OF IRRADIATION INDUCED EFFECTS

Swift ion irradiation coupled to channelling and TEM experiments – Investigating the role of electronic stopping

- Increasing the ion fluence range – Microstructural evolution – Nature of defect



- Axial yield at a depth z in a defective single crystal

$$\chi_D(z) = \chi_R(z) + (1 - \chi_R(z))\frac{f n_D(z)}{n}$$

- Dechannelling parameter at depth z

$$\gamma(z) = \ln\left(\frac{1-\chi_V(z)}{1-\chi_R(z)}\right) = \ln\left(\frac{1-\chi_V(z)}{1-\chi_D(z)}\right) + \ln\left(1 - \frac{f n_D(z)}{n}\right) = \int_0^z \sigma_D n_D(z')dz'$$

- Present case: defects characterised by $f \neq 0$ but they are *uniformly* distributed in z (distribution is depth independent), $n_D(z) = n_D$

$$\gamma(z) = \gamma_{f=0}(z) + \ln\left(1 - \frac{f n_D}{n}\right)$$

- The energy dependence of the channelling parameter is given by the $\gamma_{f=0}$ (z) contribution, which is accessible to the experiment

GANIL spiral2 laboratoire commun CEA/DSM CNRS/IN2P3

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Swift ion irradiation coupled to channelling and TEM experiments – Investigating the role of electronic stopping

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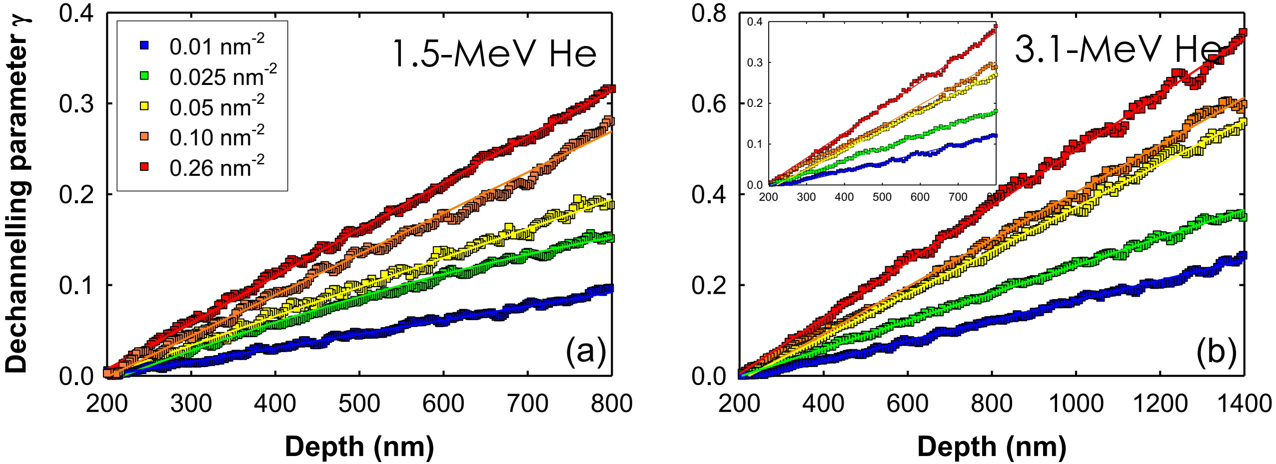
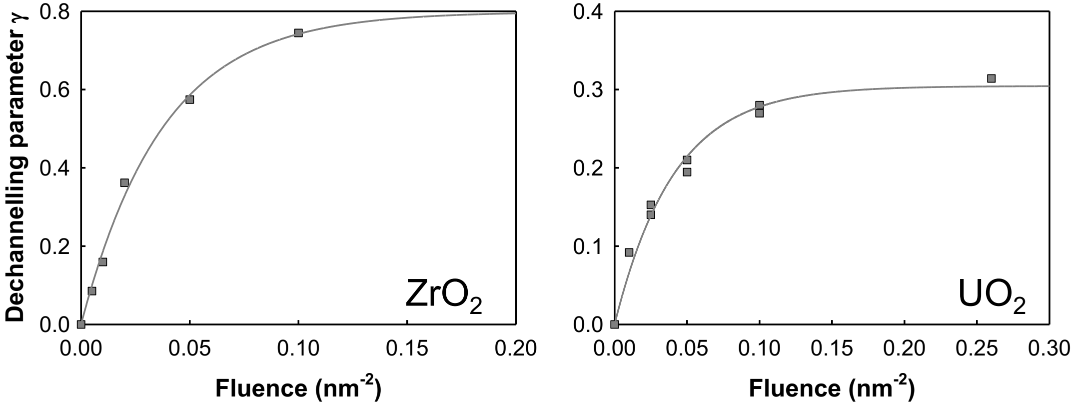


- Linear increase of γ with z: constant density of defects (in accordance with S_e)
- At a given depth γ increases with increasing the beam energy : presence of strained regions

EXPERIMENTAL SIMULATION OF IRRADIATION INDUCED EFFECTS

Swift ion irradiation coupled to channelling and TEM experiments – Investigating the role of electronic stopping

- Increasing the ion fluence range – Microstructural evolution – Nature of defects



- Direct impact model : $\gamma(\Phi) = \gamma^S \, [1 - \exp(-\sigma\Phi)]$
- ZrO_2 σ = 26 nm² (R = 2.9 nm) ; UO_2 σ = 24 nm² (R = 2.8 nm)

EXPERIMENTAL SIMULATION OF IRRADIATION INDUCED EFFECTS

Swift ion irradiation coupled to channelling and TEM experiments
Investigating the role of electronic stopping

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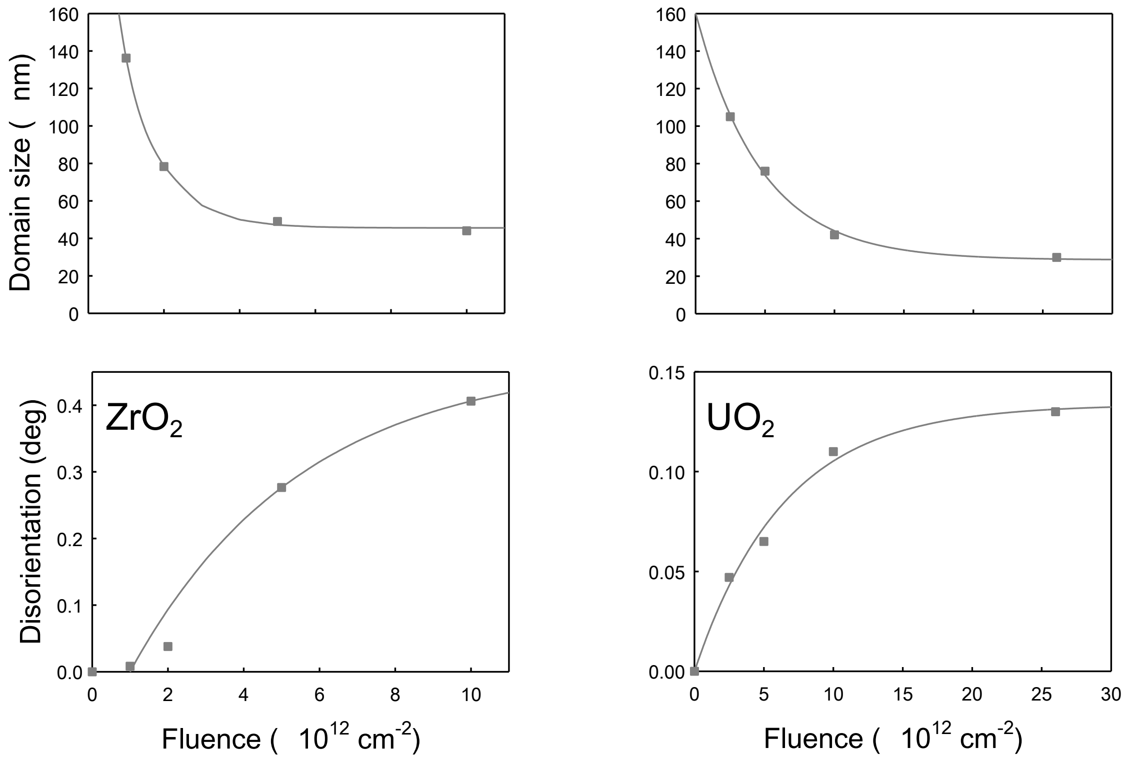
- Increasing the ion fluence range – Microstructural evolution – Nature of defects

- XRD analysis – θ–2θ/rocking curves size of coherent diffraction domains – Disorientation of coherent diffraction domains ($\Lambda_\perp$, δ_{tilt})



GANIL Spiral2

EXPERIMENTAL SIMULATION OF IRRADIATION INDUCED EFFECTS

Swift ion irradiation coupled to channelling and TEM experiments
Investigating the role of electronic stopping

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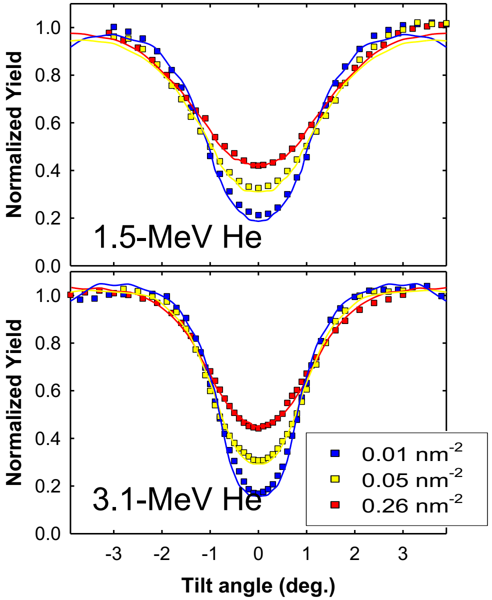
- Increasing the ion fluence range – Microstructural evolution – Nature of defect
- Analysis of angular scans – Monte Carlo simulation assuming the formation of domains ($\Lambda_{\perp}$, δ_{tilt}) over the entire thickness probed by RBS/C



Fluence (nm^{-2})	Domain size (nm)		Mosaicity δ_{tilt} (deg.)	
	XRD	RBS-C	XRD	RBS-C
0.025	105	110	0.047	0.07
0.05	76	60	0.065	0.15
0.10	42	40	0.11	0.19
0.26	30	30	0.13	0.21

GANIL spiral2

EXPERIMENTAL SIMULATION OF IRRADIATION INDUCED EFFECTS

Investigating the High Burnup Structure of spent nuclear fuels

Irradiation experiments In situ channelling and TEM experiments coupled to implantation at 773 K – Role of insoluble versus soluble fission products

- First damage step is progressive: occurs for the same dpa for Xe and La and is due to atomic collisions (same evolution for both RDA and BC); it corresponds to the sequence formation of black dots defects, dislocation loops and lines
- Clear difference between soluble and insoluble specie: insoluble specie results in a higher fraction of RDA and BC associated to the formation of nanometre-sized Xe bubbles (difference in the saturation plateaus)
- Very important contribution at high fluence leading to polygonization of the structure induced by the presence of highly pressurised gas bubbles

Investigating the High Burnup Structure of spent nuclear fuels

Swift ion irradiation coupled to channelling and TEM experiments – Investigating the role of electronic stopping

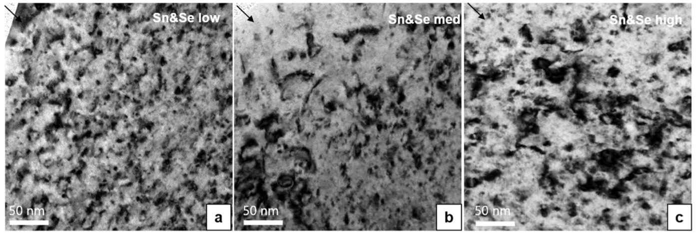
- Similar behaviour for the two investigated fluorite-structured oxides Urania and Zirconia
- Track formation at low fluence: hillocks at the surface/hollowed tracks up to 100 nm
- Electronic stopping induced micrometre-size domains: in UO_2 $(\Lambda_{\perp}, \delta_{tilt}) = (30$ nm; 0.15°); in ZrO_2 $(\Lambda_{\perp}, \delta_{tilt}) = (45$ nm; 0.4°);
- Similar to the HBS in a genuine fuel but at higher electronic stopping : no chemical contribution required

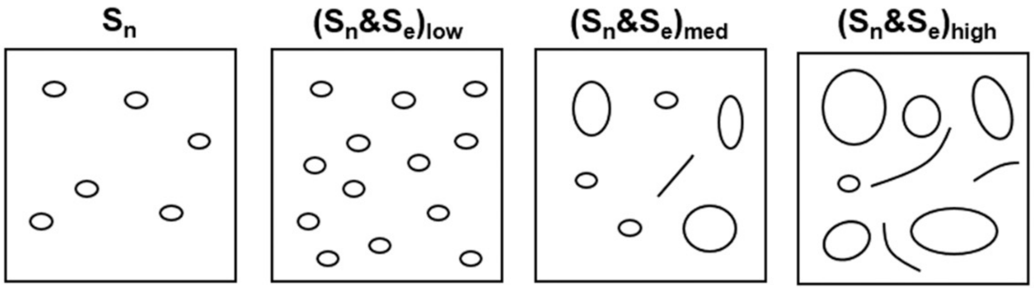
EXPERIMENTAL SIMULATION OF IRRADIATION INDUCED EFFECTS

Future experiments – Investigating the synergy between electronic and atomic contributions

Dual beam experiments performed at the JANNuS-Saclay facility

- UO_2 pellets bombarded simultaneously with low energy (atomic collisions) and medium energy (mostly electronic interactions) – TEM investigation





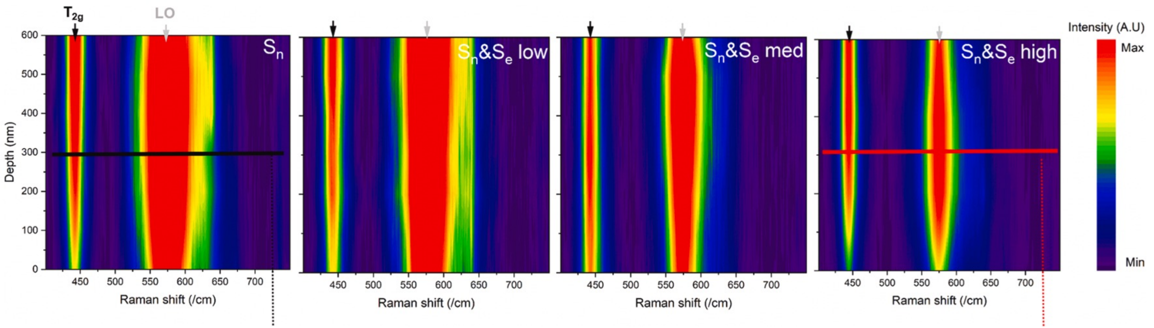
- Strong acceleration of formation and growth of dislocation loops while electronic energy deposition increases

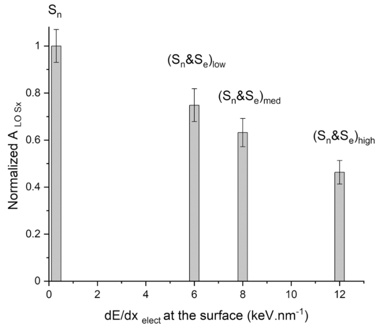
EXPERIMENTAL SIMULATION OF IRRADIATION INDUCED EFFECTS

Future experiments – Investigating the synergy between electronic and atomic contributions

Dual beam experiments performed at the JANNuS-Saclay facility

- UO_2 pellets bombarded simultaneously with low energy (atomic collisions) and medium energy (mostly electronic interactions) – Raman investigation





- Large decrease of the local disorder correlated with the increase in electronic energy deposition (decrease of uranium point defect concentration)

EXPERIMENTAL SIMULATION OF IRRADIATION INDUCED EFFECTS

Experiments at GANIL, IJCLab, JANNuS-Saclay are possible thanks to the skills and dedication of technicians, engineers and physicists

I am particularly indebted to my colleagues

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- CEA Saclay and Cadarache: Gaëlle Gutierrez, Claire Onofri
- CIMAP physicists: Clara Grygiel, Isabelle Monnet