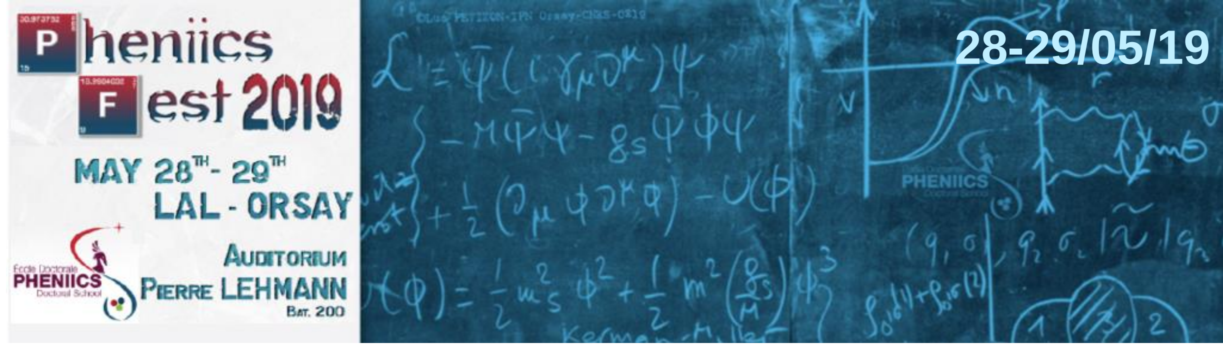

DE LA RECHERCHE À L'INDUSTRIE

cea den

BEHAVIOUR OF URANIUM DIOXIDE UNDER IRRADIATION: COMBINED EFFECT OF BALLISTIC AND ELECTRONIC DAMAGE

Marion Bricout

PhD supervisors : *G. Gutierrez*[1], *C. Onofri*[2]
PhD directors : *F. Garrido*[3], *R. Belin*[2]

JANNUS

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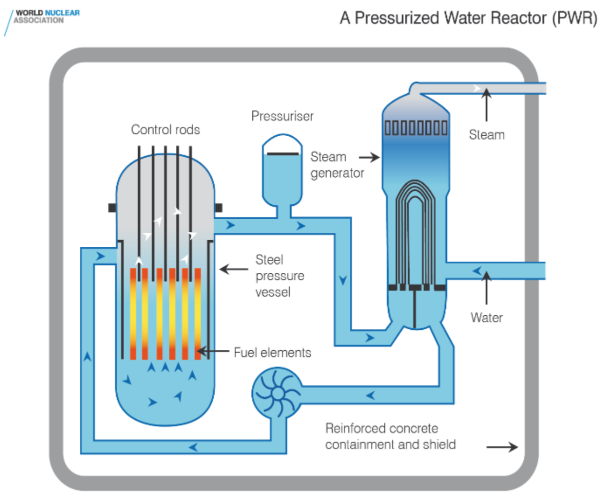
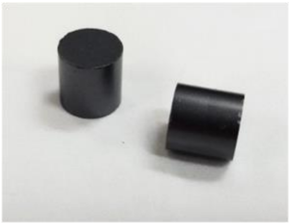
[2]*CEA, DEC/SA3E/LCPC - Cadarache, France*
[3]*CSNSM, CNRS/IN2P3 - Orsay, France*

INTRODUCTION

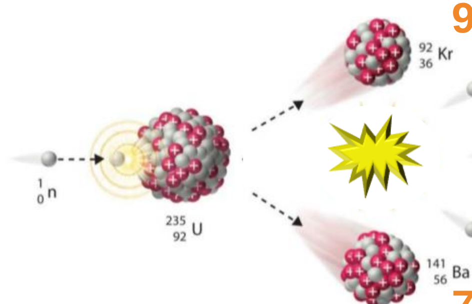


UO_2 = nuclear fuel

→ fission chain reaction



Example

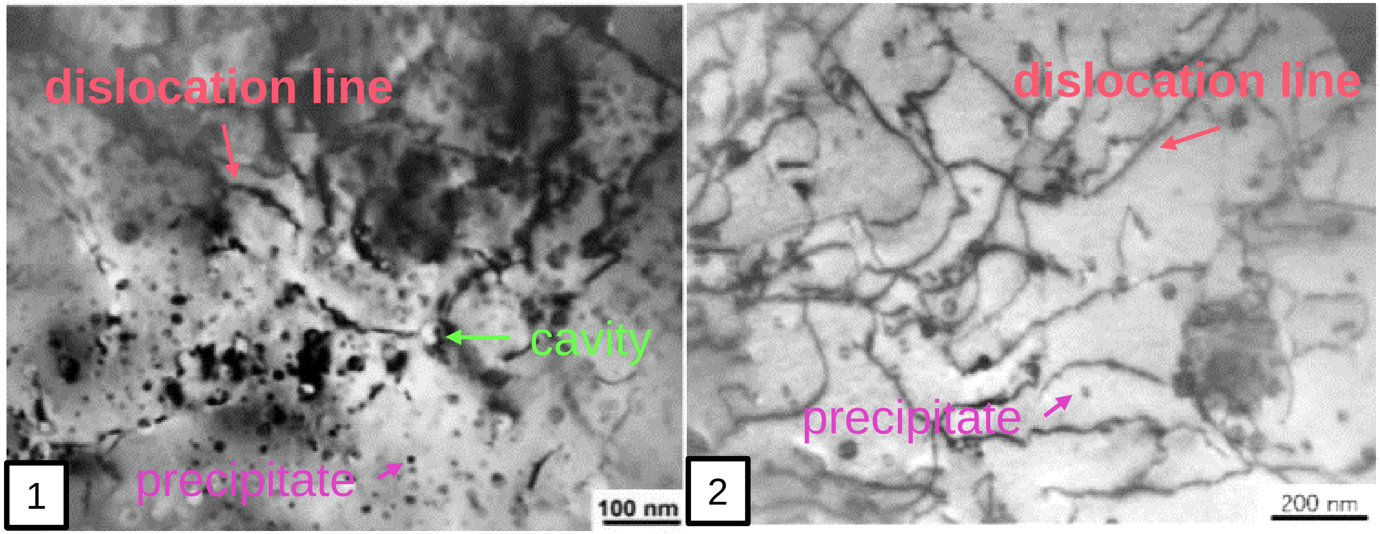


$$^{235}_{92}\mathrm{U} + ^{1}_{0}\mathrm{n} \rightarrow ^{92}_{36}\mathrm{Kr} + ^{141}_{56}\mathrm{Ba} + 3\,^{1}_{0}\mathrm{n}$$

α-decay, β-decay, neutrons, fission products…

Simultaneous radiation damages
↪ material modification (possible cracks, swelling…)
⇒ microstructural evolution

INTRODUCTION



TEM micrographs of UO_2 fuel of (1) 55 MWd/kgU, 450 °C and (2) 92 MWd/kgU, 1310 °C [1]

Associated mechanisms ?

Many contributions + simultaneous phenomena

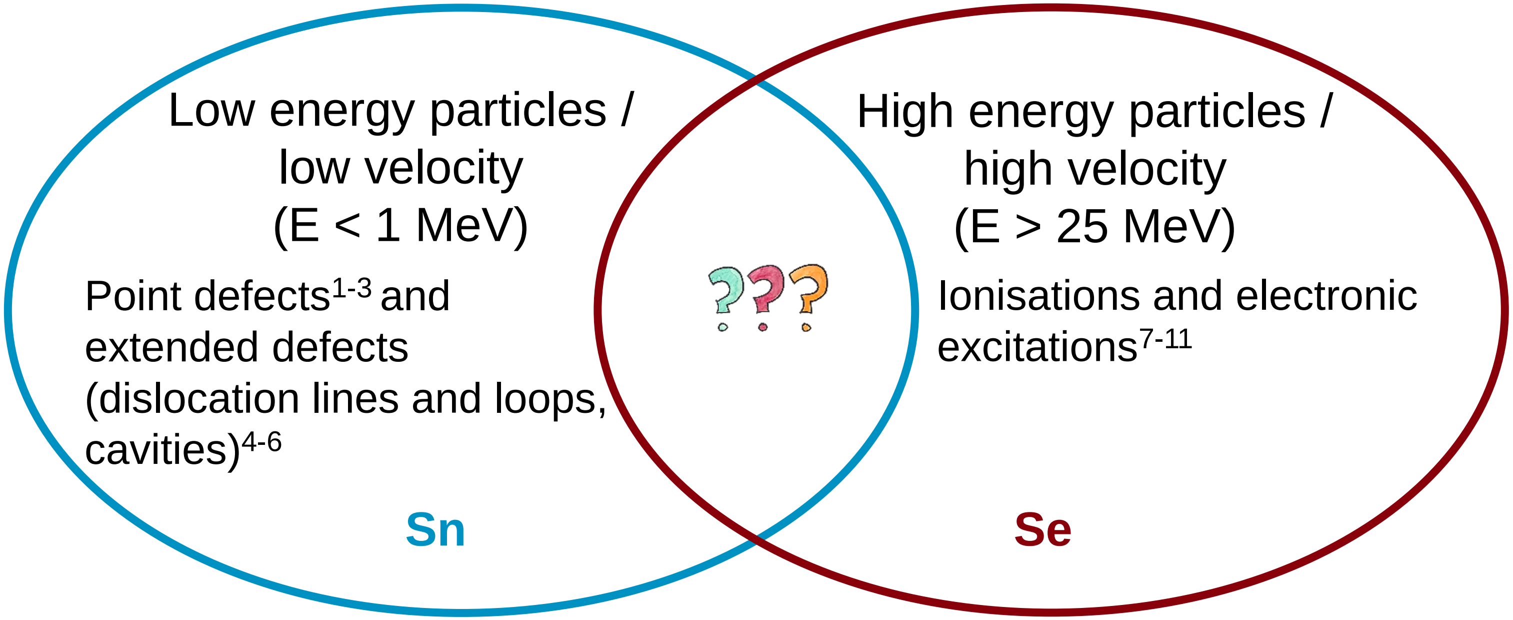
→ Necessity to decorrelate the effects

➔ Ion irradiations

Varying conditions : ion energy, flux, irradiation temperature…

[1] T. Sonoda *et al*., *Nucl. Instr. and Meth*. B 191 (2002) 622-628

CURRENT STATE OF KNOWLEDGE



Coupled effects → Need to be systematically explored

Aim of this study : physical understanding

[1] L. Desgranges et al., JOM 66, 12 (2014) 2546-2552
[2] G. Martin et al., NIMB 352 (2015) 135-139
[3] R. Mohun, PhD thesis, Université d'Aix Marseille (2017)
[4] C. Sabathier et al., NIMB 326 (2014) 247-250
[5] L.F. He et al., JNM 456 (2015) 125-132
[6] C. Onofri et al., JNM 494 (2017) 252-259
[7] F. Garrido et al., NIMB 127 (1997) 634-638
[8] Hj. Matzke et al., NIMB 167 (2000) 920-926
[9] T. Sonoda et al., NIMB 268 (2010) 3277-3281
[10] V.V. Pisarev et al., J. Phys.-Condens. Matter 26, 47 (2014) 475401
[11] L. Sarrasin et al., NIMB 435 (2018) 111-115

1. EXPERIMENTAL PROCEDURE
2. RESULTS
3. CONCLUSION & PERSPECTIVES

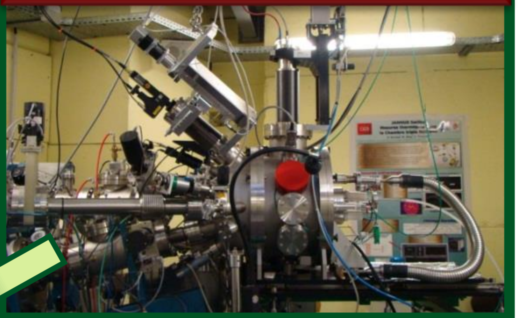
1. **EXPERIMENTAL PROCEDURE**
   - IRRADIATION CONDITIONS
   - CHARACTERIZATION TECHNIQUES

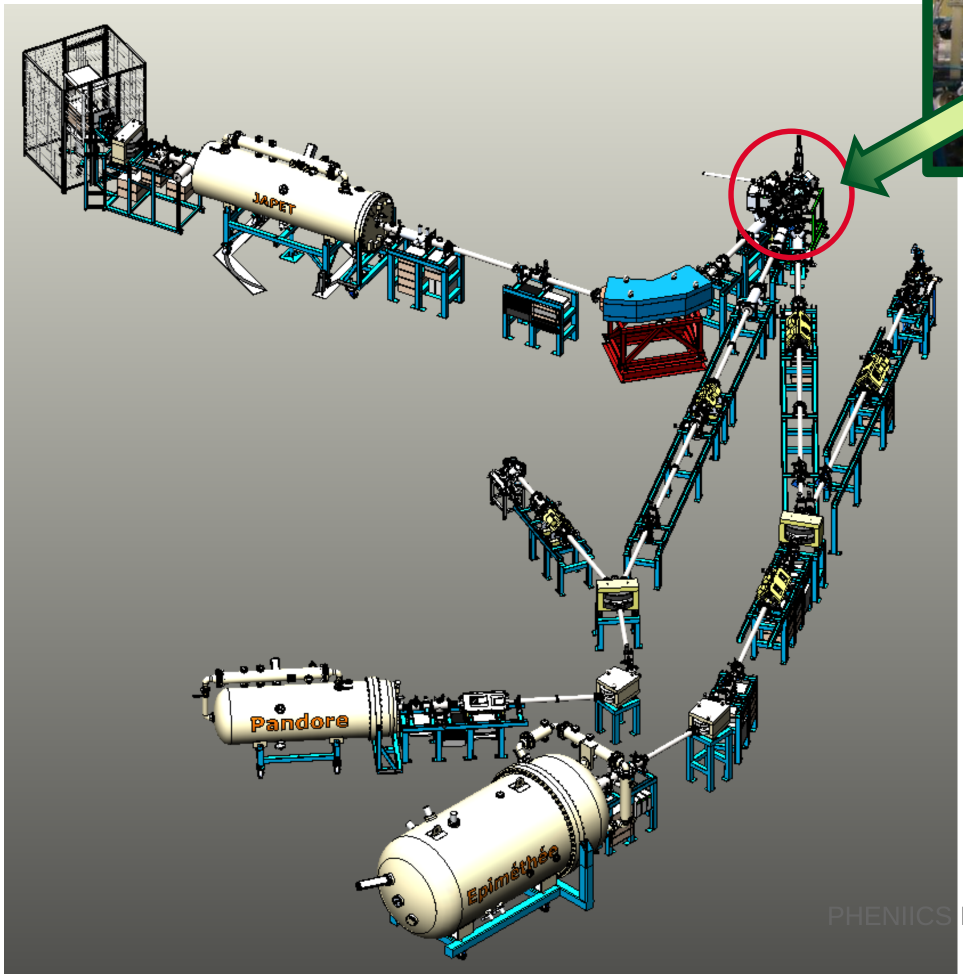
2. **RESULTS**

3. **CONCLUSION & PERSPECTIVES**

EXPERIMENTAL PROCEDURES
IRRADIATION CONDITIONS

Triple ion beam chamber





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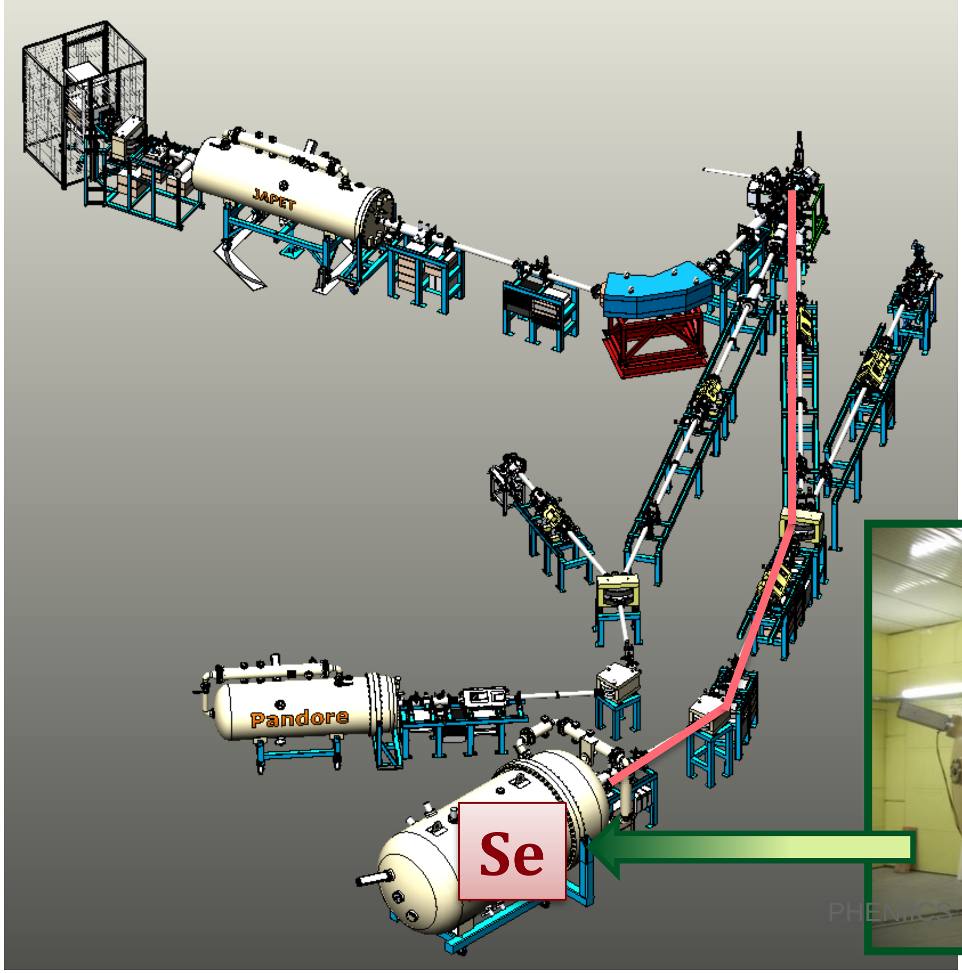
High energy irradiation (high velocity particles) :

- High electronic energy losses
- Low nuclear energy losses
- Large flux

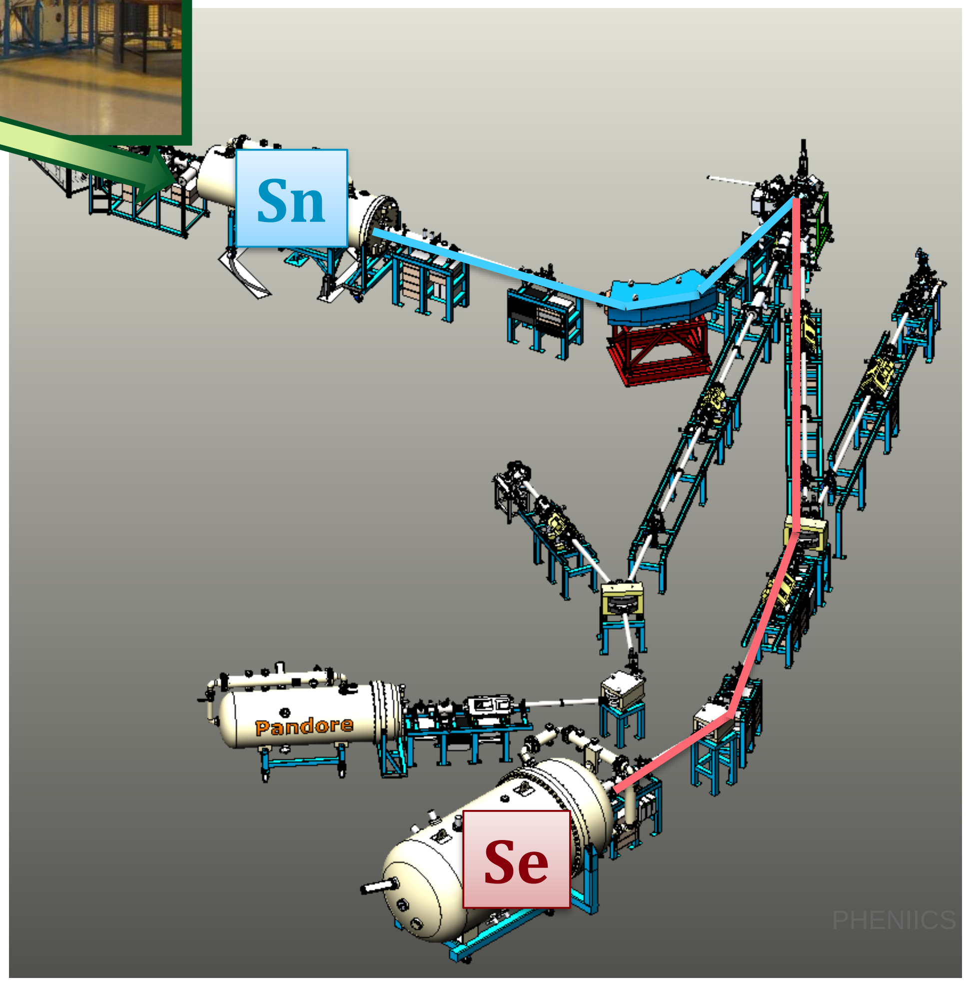


EXPERIMENTAL PROCEDURES

IRRADIATION CONDITIONS

Low energy irradiation (low velocity particles) :

- nuclear energy losses

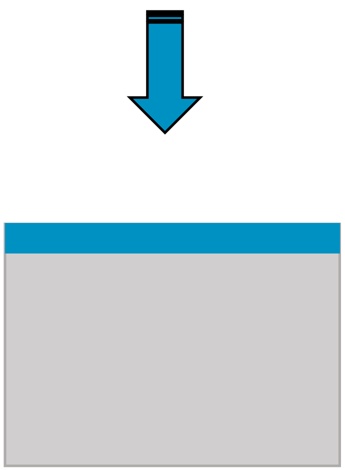


EXPERIMENTAL PROCEDURES

IRRADIATION CONDITIONS

Single ion beam (Japet)

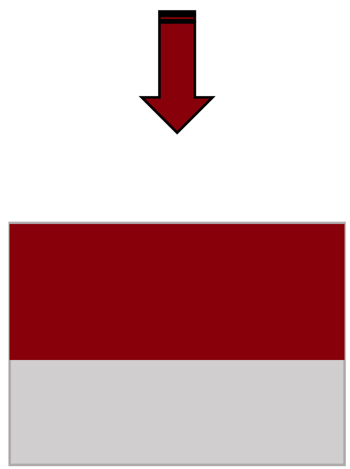
Sn



Single

Single ion beam (Épiméthée)

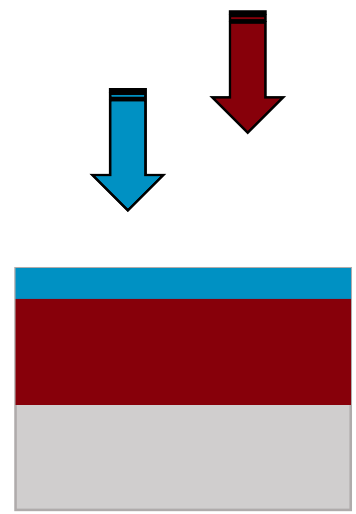
Se



Single

Two single ion beams

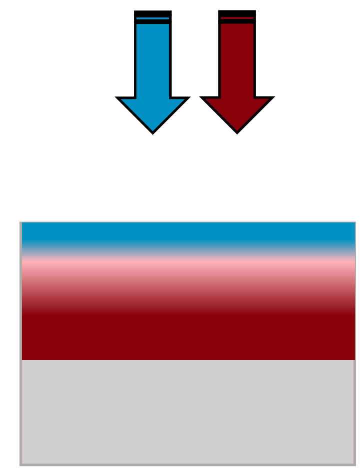
First Sn and then Se



Sequential

Dual ion beam

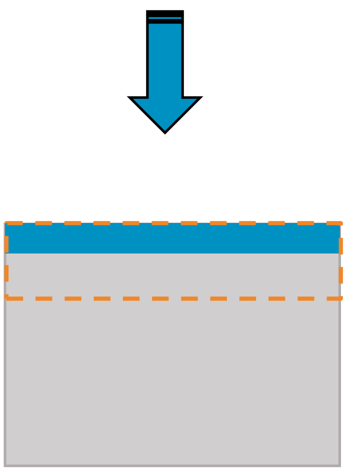
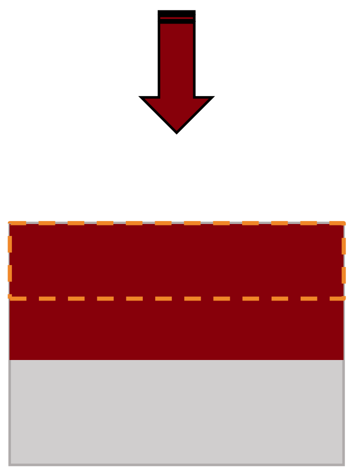
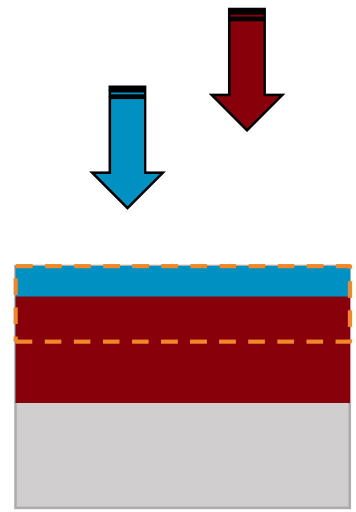
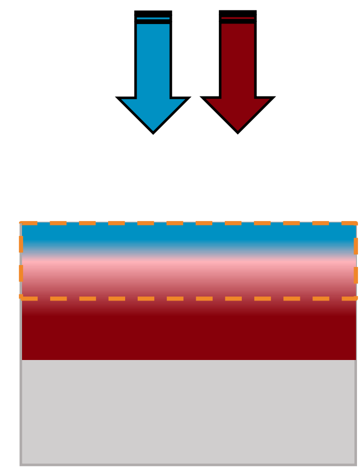
Sn and Se



Simultaneous

EXPERIMENTAL PROCEDURES

IRRADIATION CONDITIONS

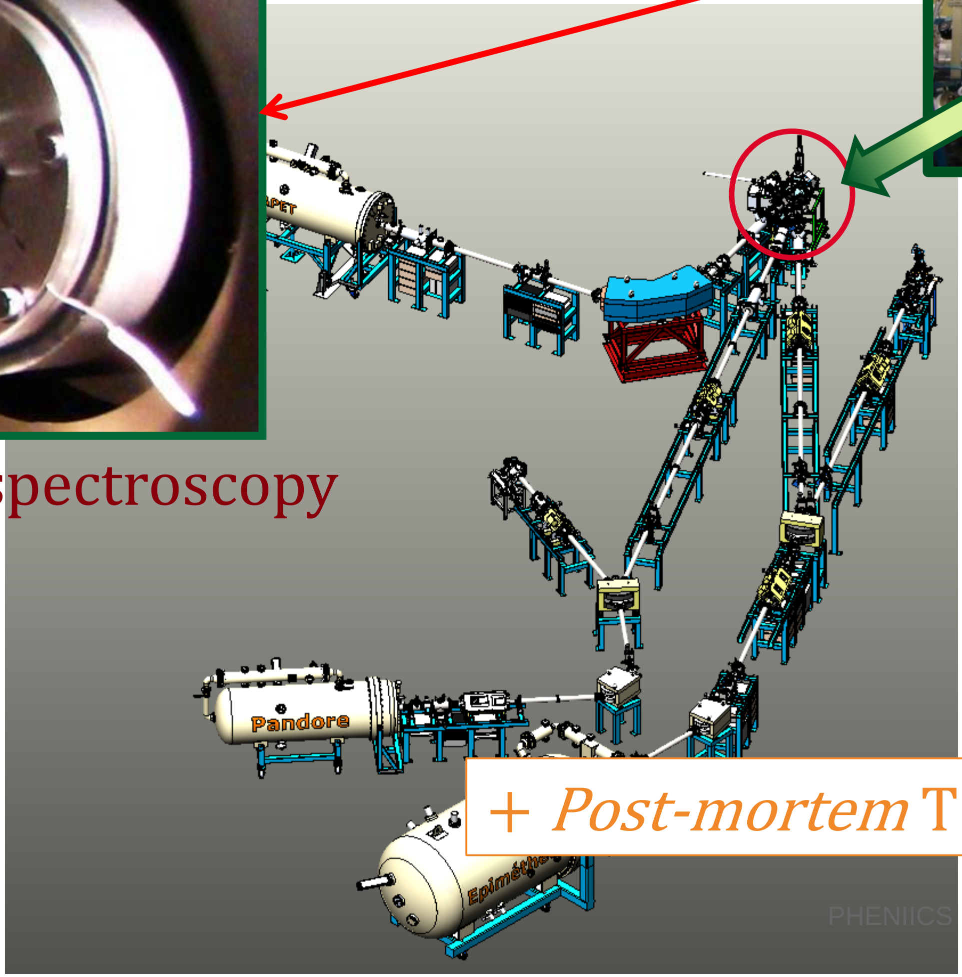
Single ion beam (Japet)	Single ion beam (Épiméthée)	Two single ion beams	Dual ion beam
Sn	Se	First Sn and then Se	Sn and Se
			
Single	Single	Sequential	Simultaneous

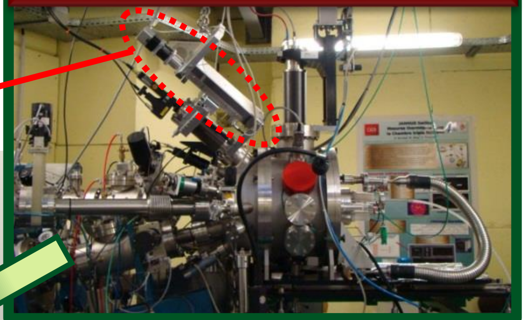
probed area by characterization techniques

EXPERIMENTAL PROCEDURES

IRRADIATION CONDITIONS

In situ Raman spectroscopy





Triple ion beam chamber

\+ *Post-mortem* TEM observations

1. EXPERIMENTAL PROCEDURE

2. RESULTS

- RAMAN SPECTROSCOPY
- TRANSMISSION ELECTRON MICROSCOPY

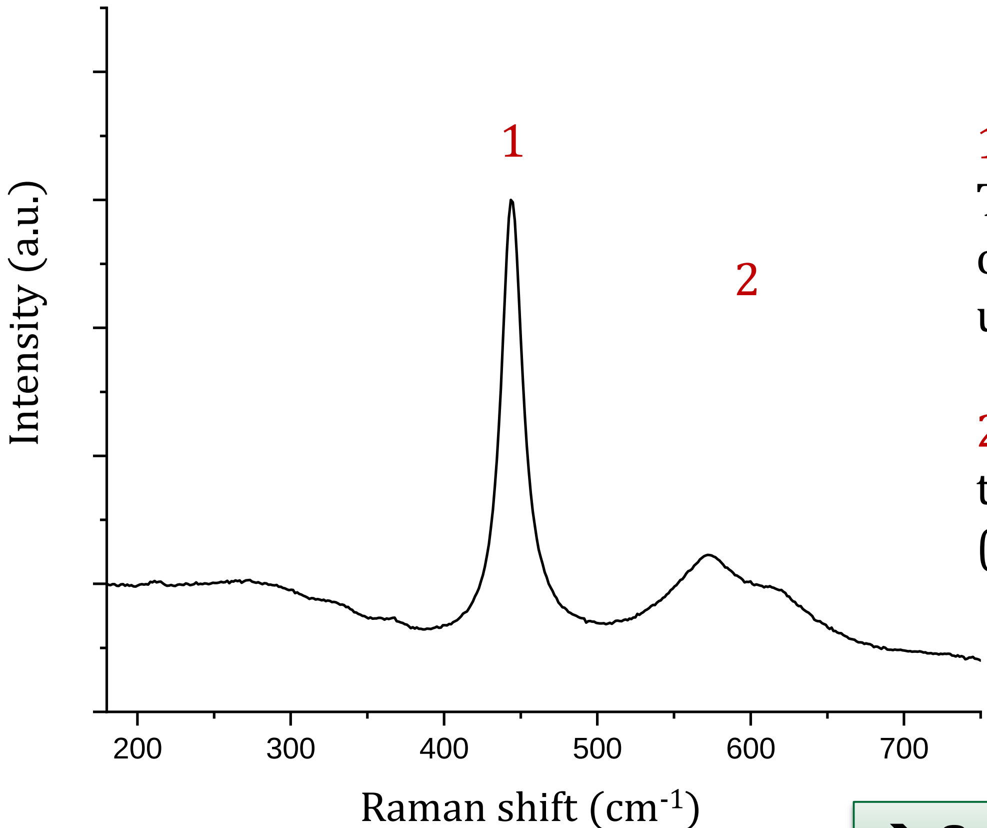
3. CONCLUSION & PERSPECTIVES

RESULTS

RAMAN SPECTROSCOPY

Raman : Technique based on inelastic scattering of monochromatic light

Green laser (λ = 532 nm) → analyzed depth ≈ 1 µm
Power < 1 mW



Virgin UO_2 spectrum

1 : **445 cm^{-1}**
T_{2G} structural mode → vibration of oxygen atoms around the uranium atom

2 : **between 500 and 700 cm^{-1}**
three new Raman modes (U_1[1], LO[2], U_3[3])

➔ Spectrum progress under irradiation ?

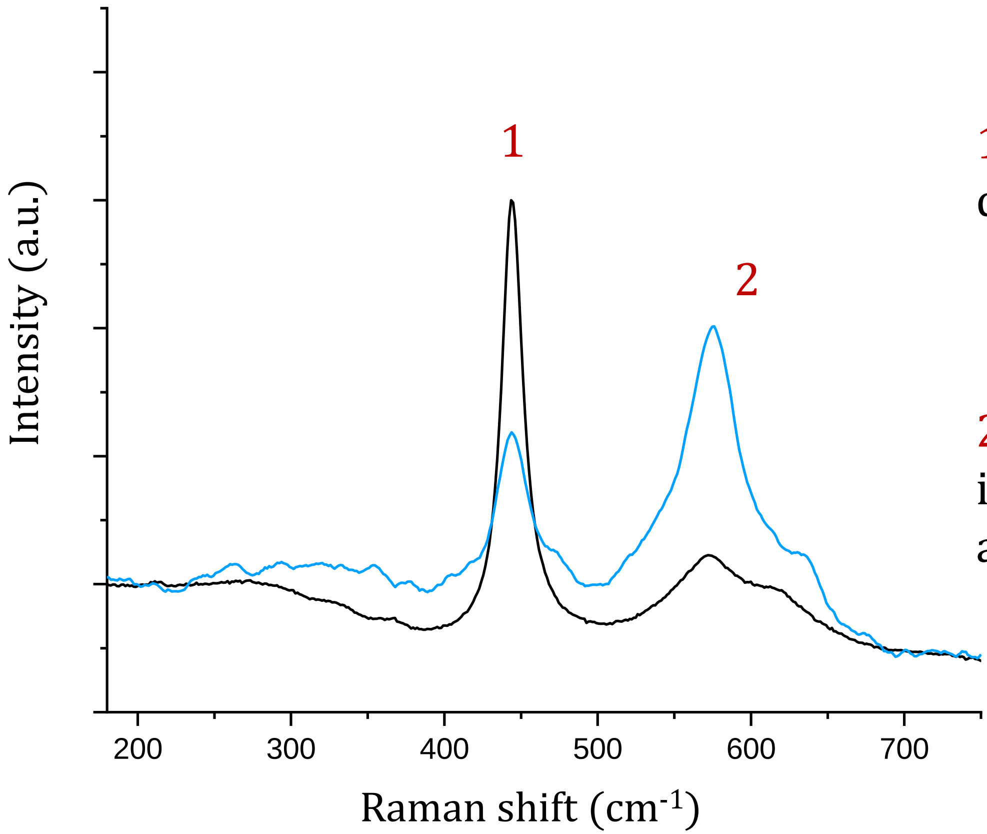
[1] L. Desgranges et.al., *JOM*. 66,12 (2014)
[2] J.D. Axe et.al., *Phys. Rev.* 151, 676 (1966)
[3] G. Guimbretière et.al., *App. Phys.* 100, 251914 (2012)

RESULTS

RAMAN SPECTROSCOPY

Evolution of the triplet defect band area

Sn



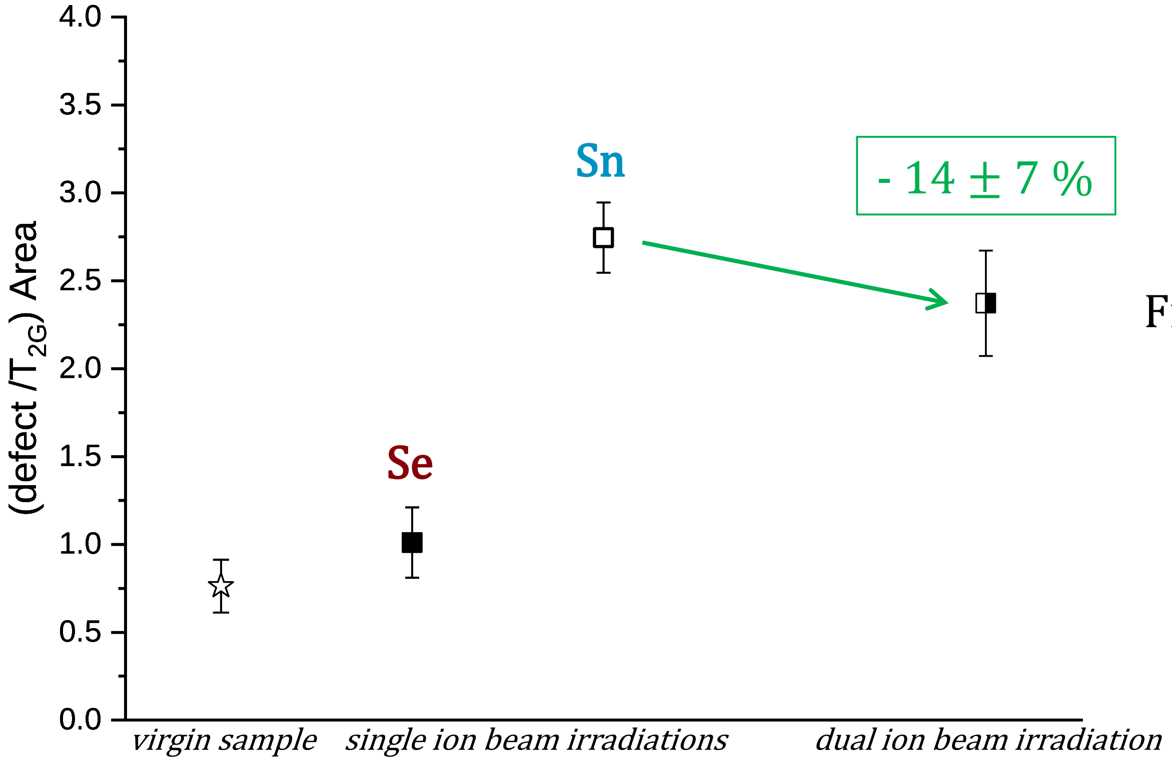
1 : **445 cm^{-1}**
decrease of T_{2G} band with a broadening

2 : **between 500 and 700 cm^{-1}**
increase of the triplet defect bands area [1, 2, 3]

[1] P.R. Graves, *Applied Spectroscopy*. 44, 10 (1990)
[2] L. Desgranges et.al., *JOM*. 66, 12 (2014)
[3] L. Desgranges et.al., *NIMB.* 327 (2014)

RESULTS

RAMAN SPECTROSCOPY

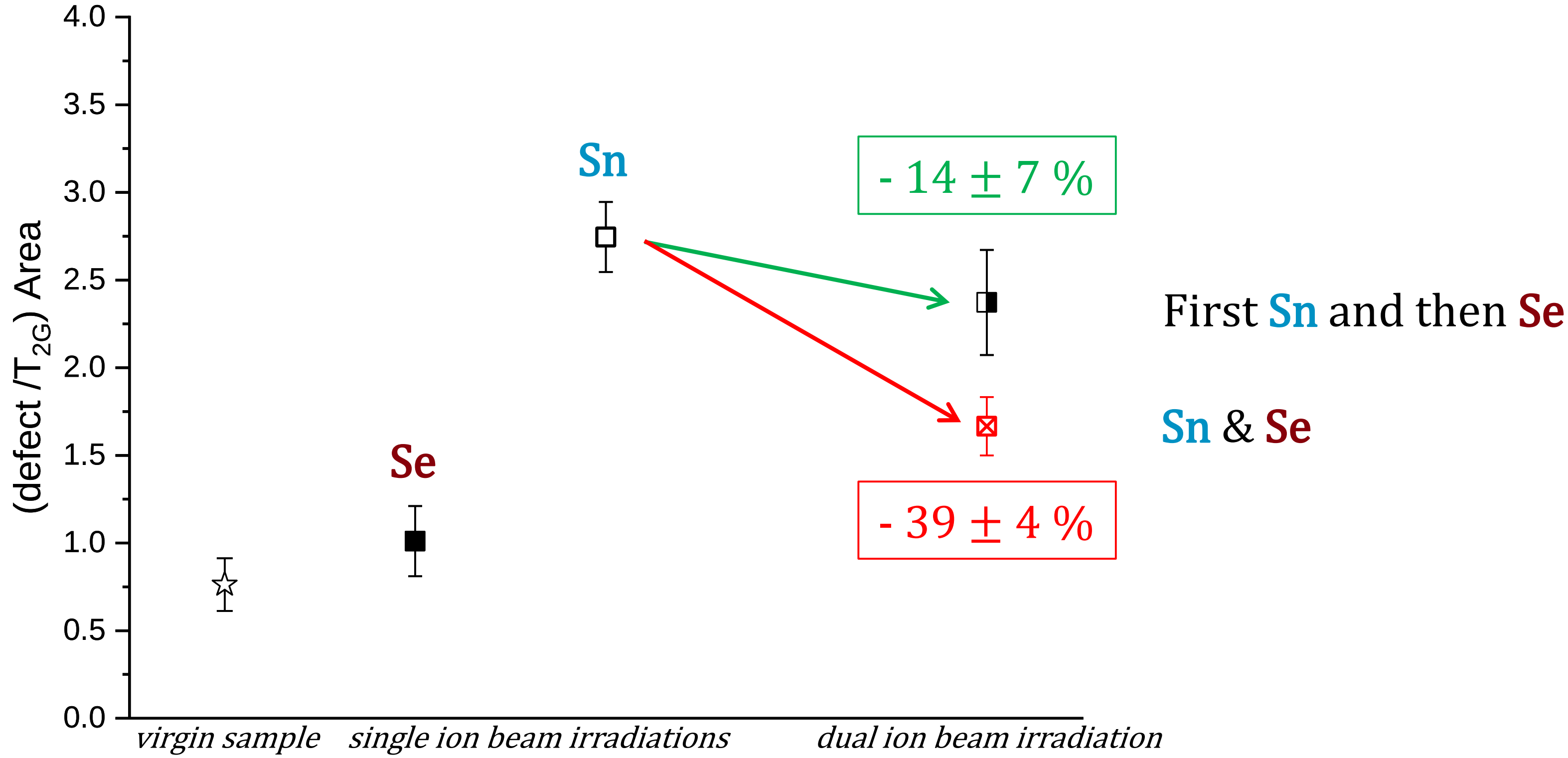


First Sn and then Se

Slight effect of electronic energy loss on the pre-damaged sample

RESULTS

RAMAN SPECTROSCOPY

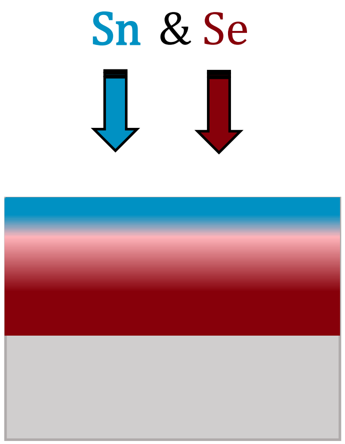


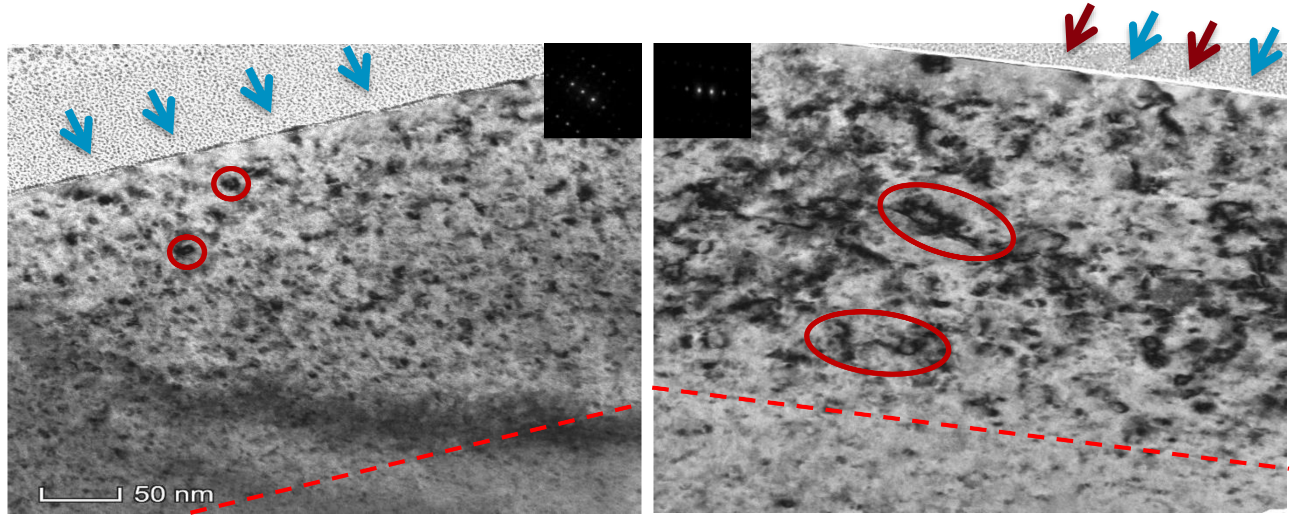
More significant effect on simultaneous irradiation than sequential irradiation

Microstructural evolution ?

RESULTS

TEM





Different microstructure

→ Transformation of dislocation loops into dislocation lines

Irradiation conditions	Dislocation loops density (10^{16} loops.cm^{-3})	Dislocation lines density (10^{9} cm^{-2})
Sn	13.4 ± 5.4	–
Sn & Se	7.6 ± 3.0	14.2 ± 5.7

MET FEI Talos, SA3E/LCPC, Cadarache

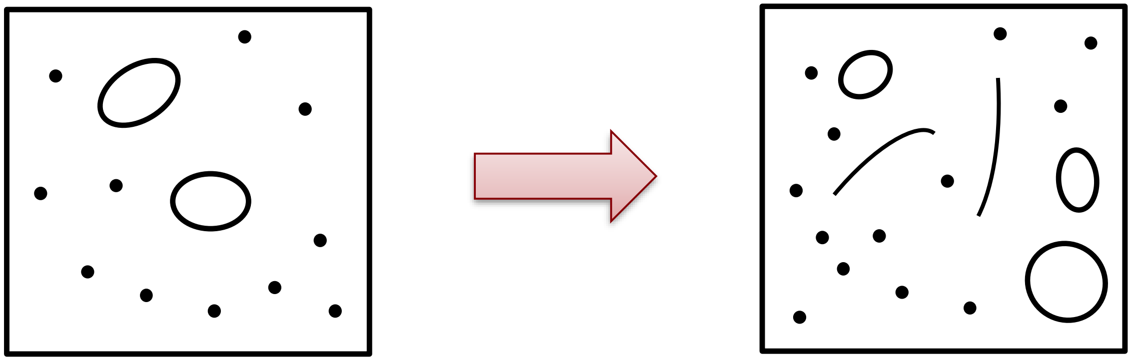
1. EXPERIMENTAL PROCEDURE

2. RESULTS

3. CONCLUSION & PERSPECTIVES

CONCLUSION

TEM and Raman spectroscopy = complementary techniques



Defect recombination ⇨ Stress/strain relaxation

Macroscopic effect ?

PERSPECTIVES

- **Evaluation of the local temperature, closed to the ion track** (thermal spike model)

- **Change of ion beam conditions**
  lower velocity (JANNUS Saclay)
  higher velocity (CIMAP - GANIL)

Thank you for your attention

Commissariat à l’énergie atomique et aux énergies alternatives
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EXPERIMENTAL PROCEDURE

Irradiation conditions and paramaters using the SRIM code

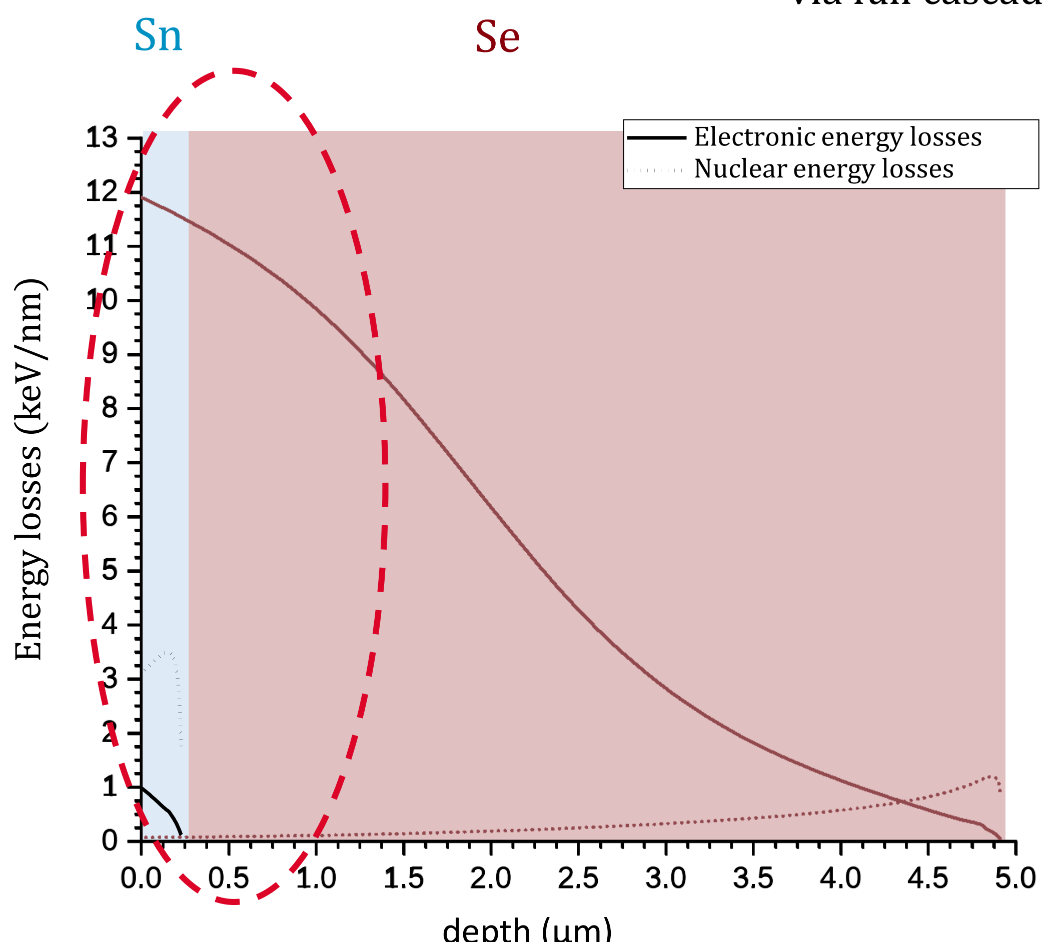
Accelerator	Irradiation conditions	Dpa max on 1 µm depth at 10^{14} ions/cm²	Se* (keV/nm)	Sn* (keV/nm)	Se/Sn
Japet	*0,9 MeV I^{2+}*	0.76	0.75	3.4	0.22
Epiméthée	*27 MeV Fe^{9+}*	0.03	10.4	0.11	94

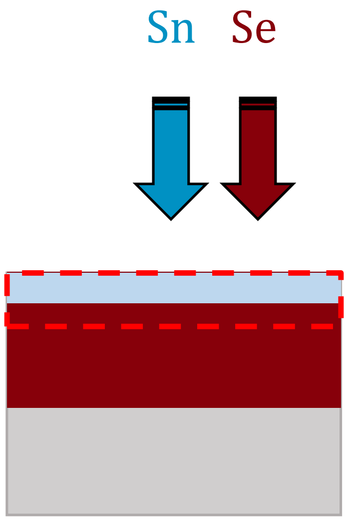
*on the depth analyzed by Raman spectroscopy on the maximum rate of damage on 1 µm depth

EXPERIMENTAL PROCEDURES

IRRADIATION CONDITIONS

Parameters calculated using the SRIM code[1] via full cascade calculations





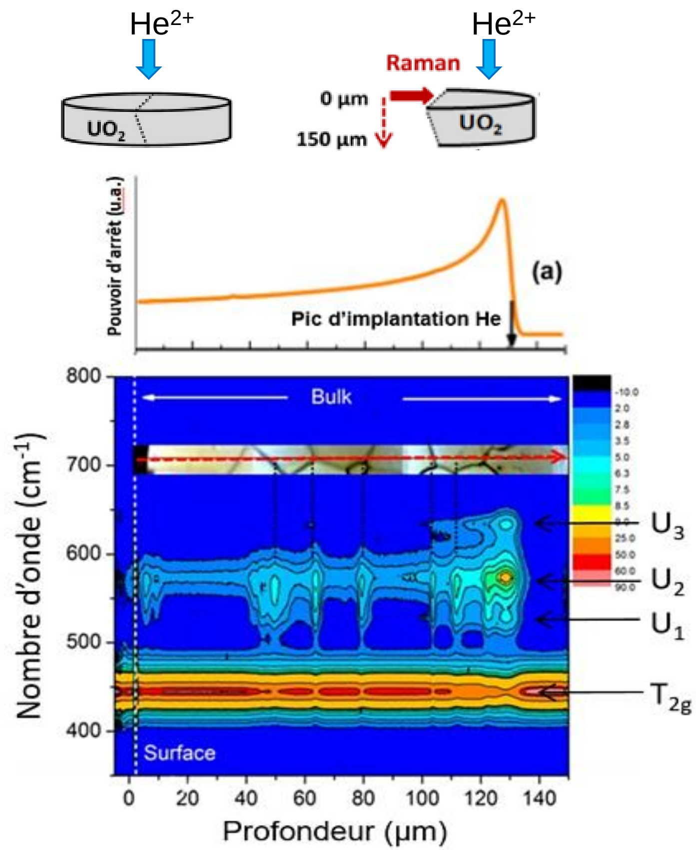
Probed area by all characterization techniques

[1] J.F.Ziegler *et al*, NIMB 268 (2010) 1818-1823

RAMAN SPECTROSCOPY

SRIM code[3]
$E_d(U)$ = 40 eV
$E_d(O)$ = 20 eV

- <u>Defects band</u>



Raman spectrum as a function of the implantation depth in 25 MeV He irradiated UO_2[1,2]

→ Maximum defects band signal on the depth of the most damage created by irradiation

Probed depth with 532 nm laser ≈ 1µm

[1] G.Guimbretière *et.al.*, *Appl.Phys.Lett.* 100, 251914 (2012)
[2] L.Desgranges *et.al.*, *NIMB*, 315 (2013) 169-172
[3] J.F.Ziegler *et al*, Nuc. Instr. Met. In Phy. Research, B 268 (2010) 1818-1823