Understanding of radiation effects and damage in nuclear reactors materials using JANNuS ion beams

C. Cabet, A. Gentils

DE LA RECHERCHE À L'INDUSTRIE

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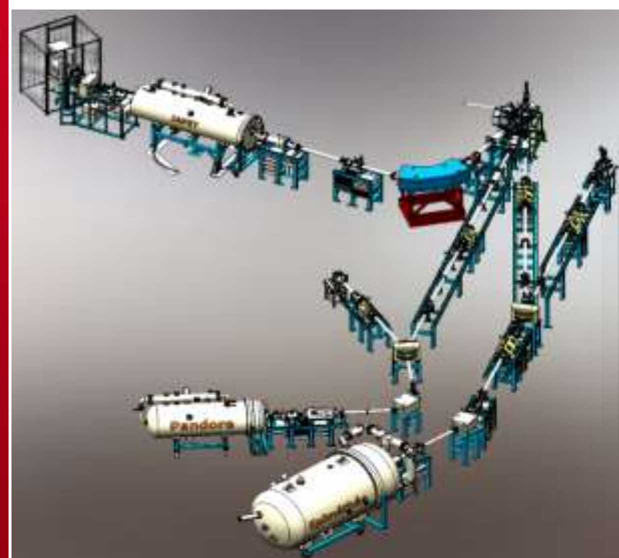
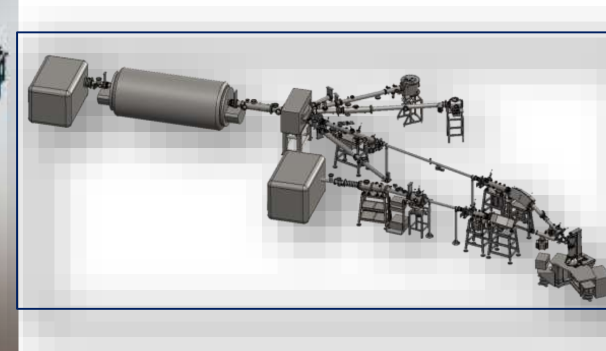




Céline Cabet, Aurélie Gentils,

Université Paris-Saclay, France

JANNUS

http://jannus.in2p3.fr

JANNuS: 1 platform, 2 facilities

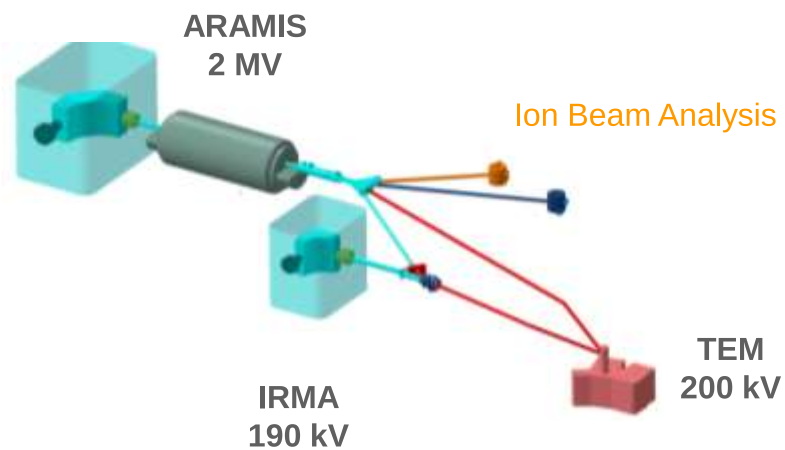
Joint Accelerators for Nanoscience and Nuclear Simulation

GIS JANNuS (scientific interest grouping since 2004)

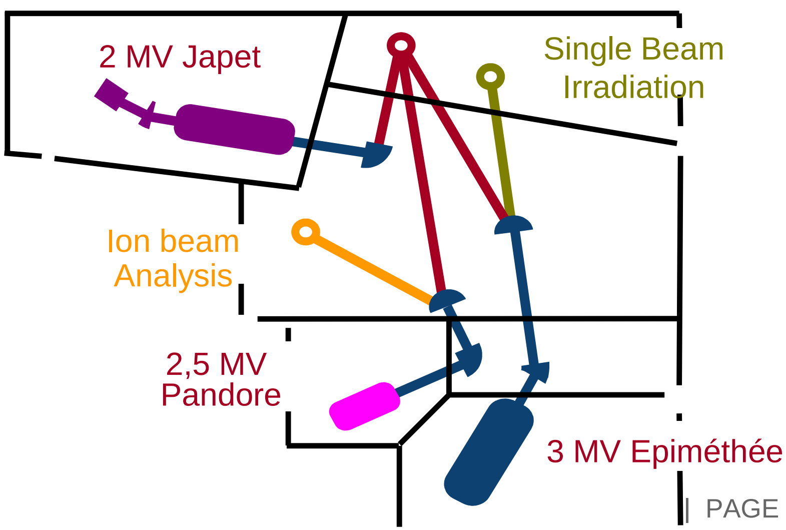
- ➔ **Founding partner of the French accelerator network** emir
  http://emir.in2p3.fr

- ➔ **Simulation of damage accumulation induced by neutrons in nuclear materials**

JANNuS Orsay
on line TEM coupled to one or two beams



JANNuS Saclay
triple beam irradiation facility with high damage rate



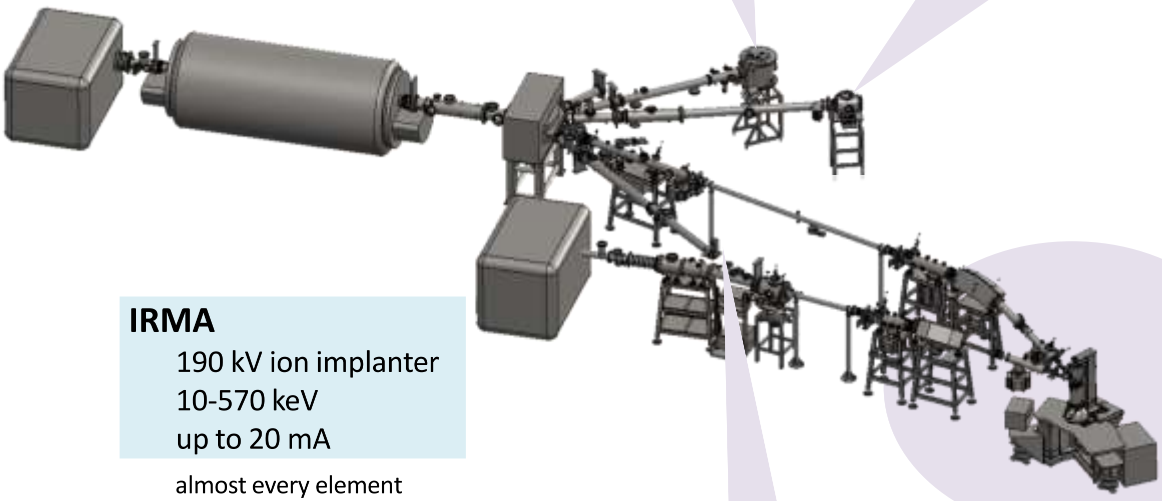
The *in situ* dual ion beam TEM at CSNSM : JANNuS-Orsay facility

ARAMIS
- 2MV Tandem - VdG
- 0.5 – 11 MeV *
- 10 nA – 10µA

> 40 elements
* limited to 1 MeV per charge state inside the TEM

Ion Beam Analysis
RBS, RBS/C, ERDA,
PIXE, *µPIXE, PIGE*

implantation / irradiation
LN_2 –> 1000°C

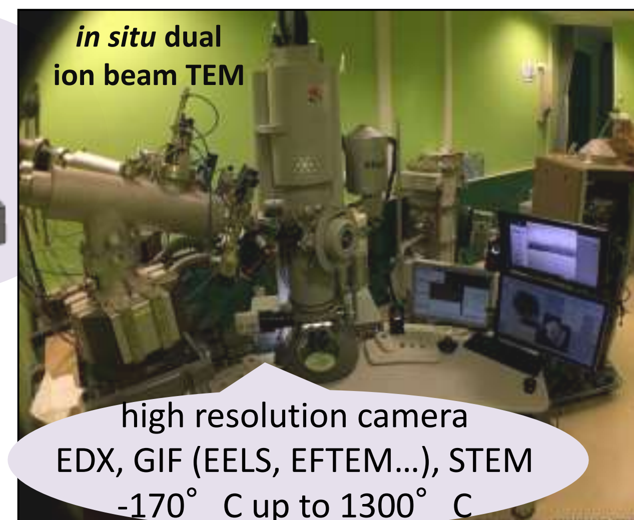


TRANSMISSION ELECTRON MICROSCOPE
- 200 kV *FEI Tecnai G2 F20 Twin*
- Resolution: 0.25 nm
- Magnification range: 70-700 000

IRMA
- 190 kV ion implanter
- 10-570 keV
- up to 20 mA

almost every element

in situ RBS/C
and implantation
LN_2 –> 600°C

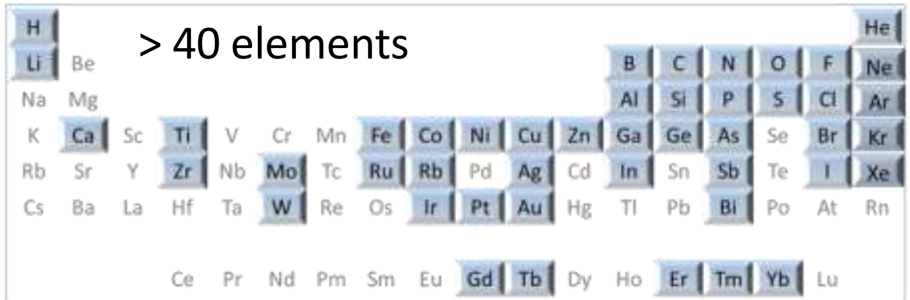


high resolution camera
EDX, GIF (EELS, EFTEM…), STEM
-170° C up to 1300° C

ARAMIS ion accelerator

2MV Tandem – VdG

HOME BUILT IN 1987



VAN DE GRAAFF MODE

Mainly H, He (also noble gaz)

200 keV – 1.8 MeV (H), 3.6 MeV (He)

Positive Penning ion source

> 20 µA

Ion beam analysis

(RBS, RBS/C, ERDA, PIXE, *µPIXE, PIGE*)

Ion irr./impl.

Connection with TEM

Connection with IRMA ion implantor

TANDEM MODE

0.5-11* MeV, 10 nA – 10µA

SNICS source (Source of Negative Ions by Cesium Sputtering)

* limited to 1 MeV per charge state inside the TEM

Typical irradiation conditions:

- Rastered beam

(80 Hz vertical and 400 Hz horizontal)

- Average flux range

$1.10^{10} – 5.10^{11}$ $cm^{-2}.s^{-1}$

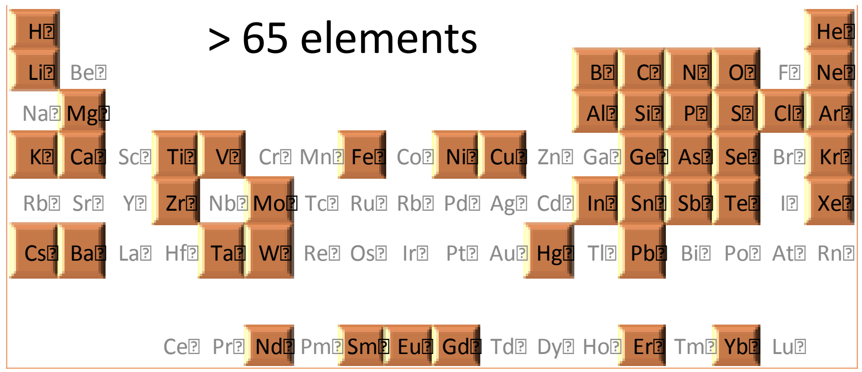
(depending on ions and energies)

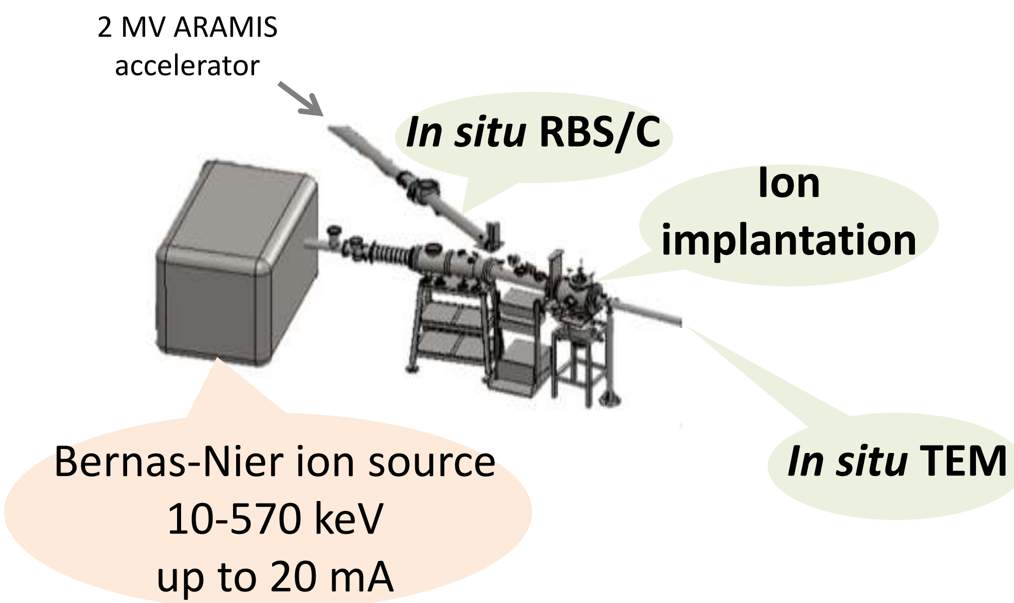
- Temperature LN_2 ->1000°C

(thermocouple control)

IRMA 190 kV ion implanter

HOME BUILT IN 1979



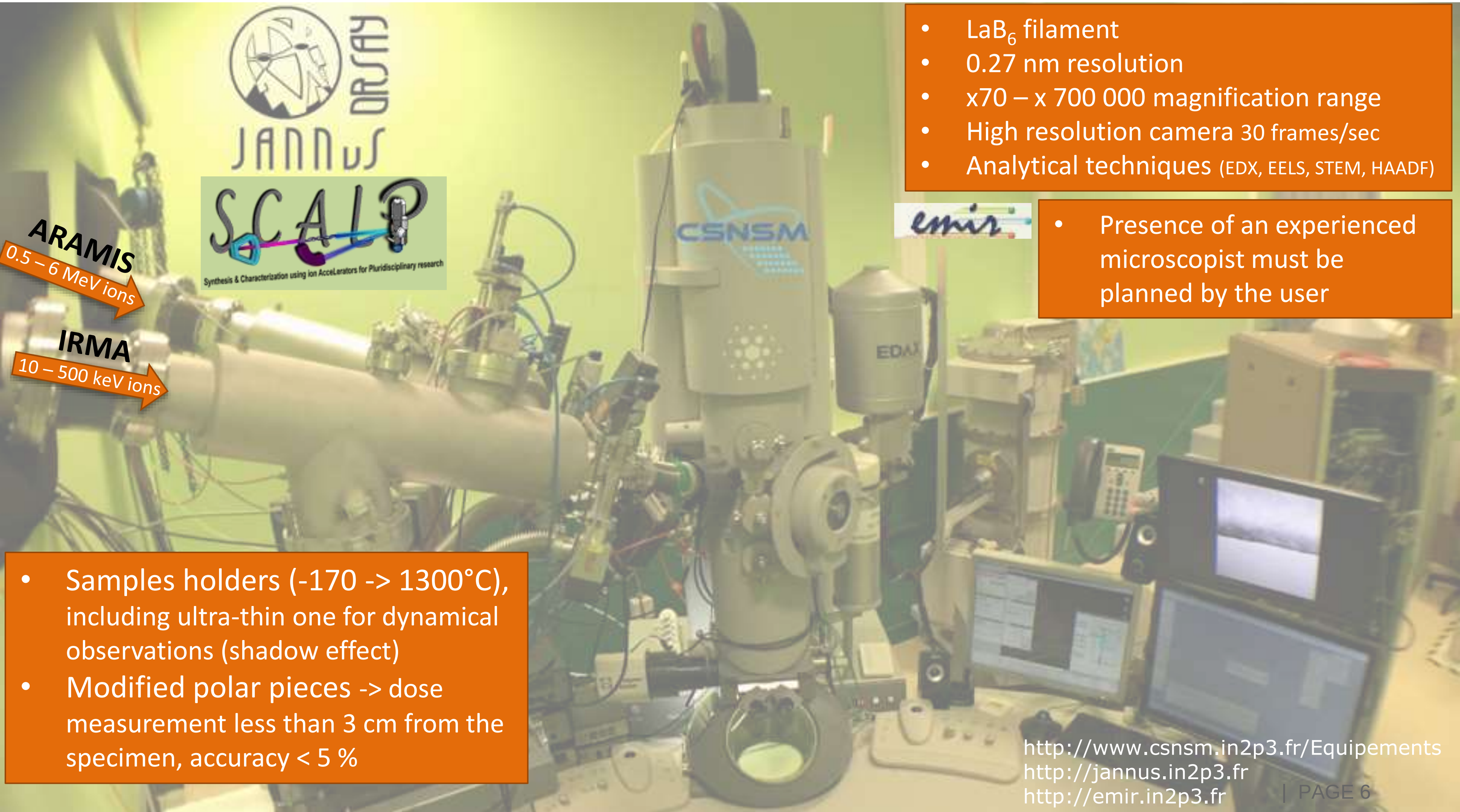


Typical irradiation conditions:

- Rastered beam
  (400 Hz vertical and 80 Hz horizontal)
- Average flux range
  $1.10^{10} – 5.10^{12}$ $cm^{-2}.s^{-1}$
  (depending on ions and energies)
- Temperature LN_2 ->1000°C
  (thermocouple control)

In situ dual ion beam Transmission Electron Microscope

1[st] TEM (120 kV EM400) connected to IRMA ion implanter in 1980
3[rd] TEM (200 kV FEI Tecnai G^2 F20 Twin, shown below) since 2009 for external users



- LaB_6 filament
- 0.27 nm resolution
- x70 – x 700 000 magnification range
- High resolution camera 30 frames/sec
- Analytical techniques (EDX, EELS, STEM, HAADF)

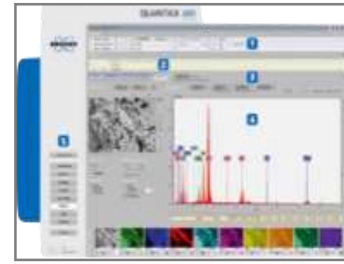
- Presence of an experienced microscopist must be planned by the user

- Samples holders (-170 -> 1300°C), including ultra-thin one for dynamical observations (shadow effect)
- Modified polar pieces -> dose measurement less than 3 cm from the specimen, accuracy < 5 %

http://www.csnsm.in2p3.fr/Equipements
http://jannus.in2p3.fr
http://emir.in2p3.fr

The *in situ* dual ion beam TEM at CSNSM : JANNuS-Orsay facility

New and future developments



- **EDX update**

Chemical analysis => Brucker Flash 6 detector
and Esprit software -> hyper-maps

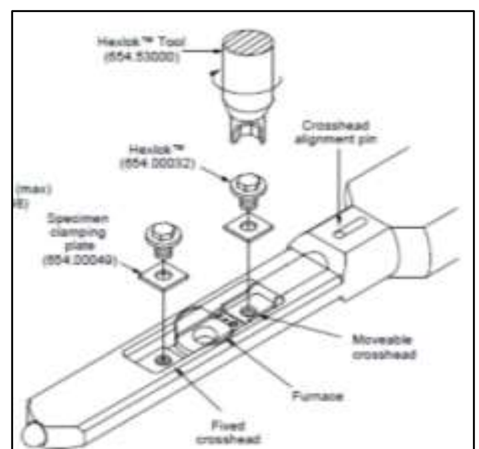
- **New ultra-fast high resolution camera**

High quality image (4K)
+ high-speed video capture (25 fps at 4K up to 300 fps at 512x512)
+ ultra fast shutter = **great tool for *in situ* imaging !**

- **Environmental sample holder**

Liquid and/or gas under irradiation
with heating control (+ options)

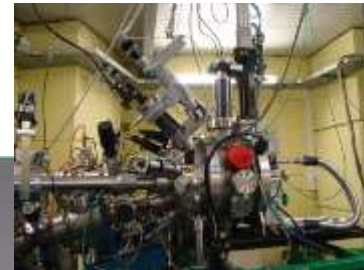
- **Straining heating holder** (1000°C)

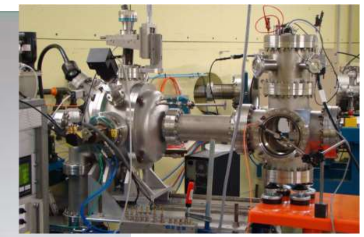


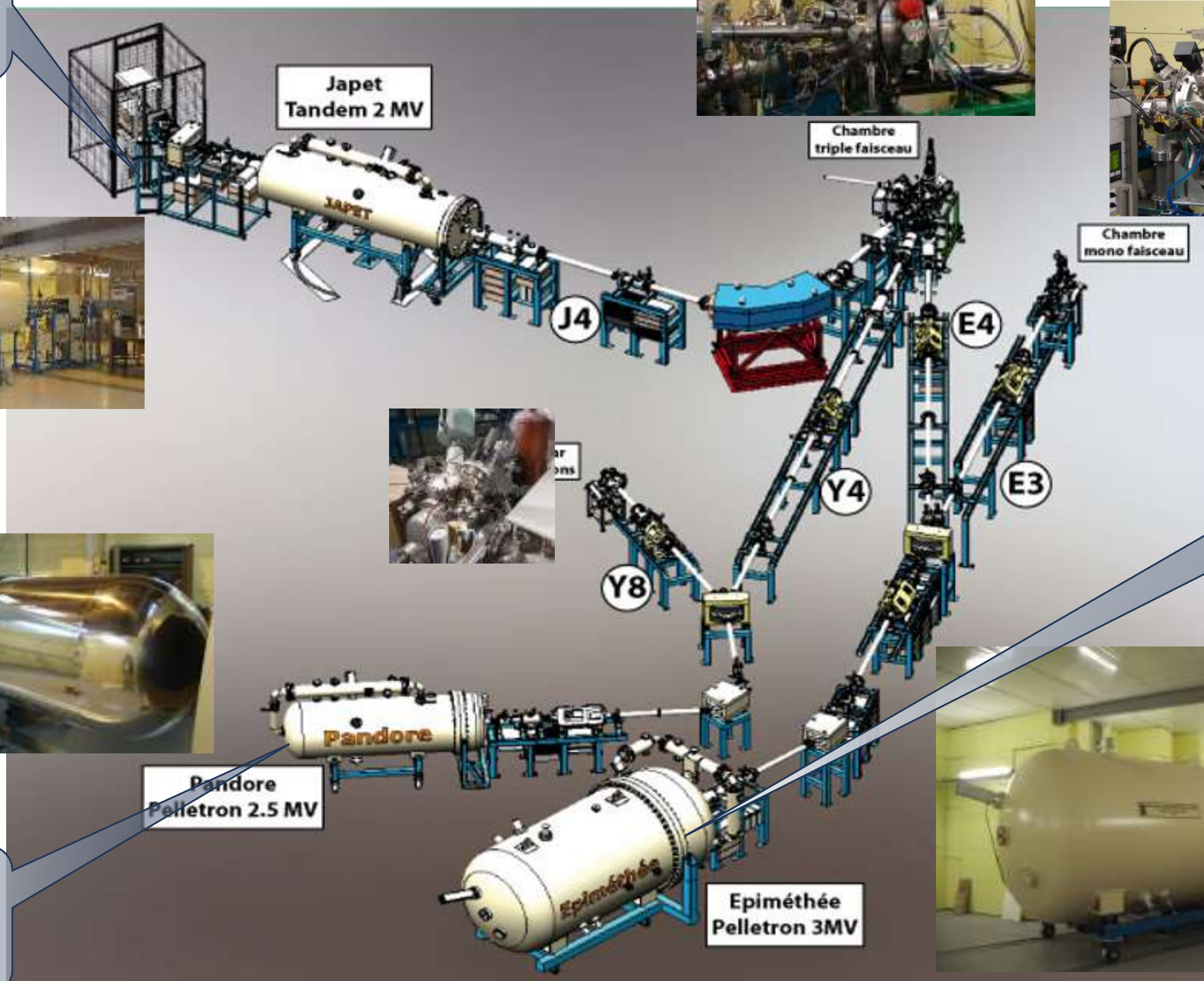
JANNuS-Saclay: 3 complementary accelerators

Heavy ion damage
Hydrogen implantation

First triple beam irradiation in March 2010





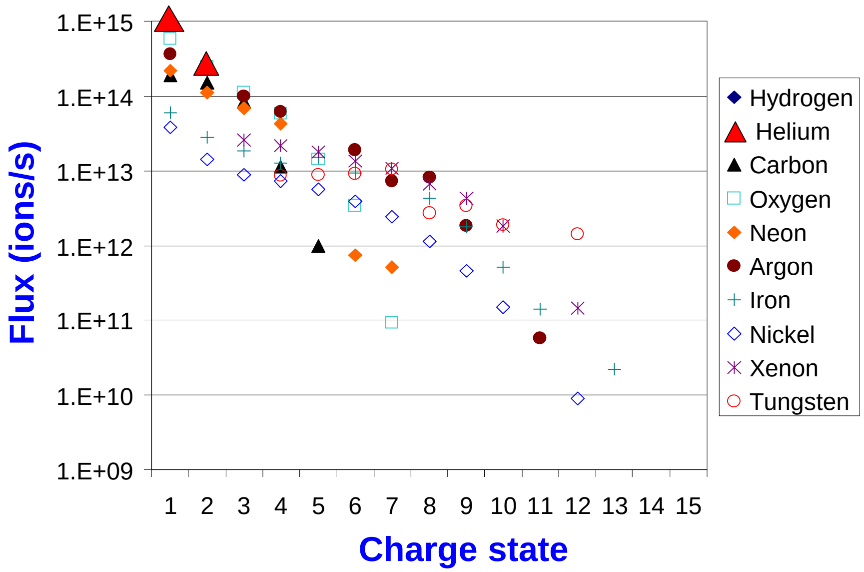


Heavy ion damage
Helium and hydrogen implantation

Helium and hydrogen implantation

JANNuS-Saclay: 3 MV Pelletron Épiméthée

➢ **Accelerator: 3 MV Pelletron NEC (National Electrostatics Corporation) with a ECR (Electron Cyclotron Resonance) source from Pantechnik**



⇨ **Positive multi-charged ions**
1 < m < 209
400 keV < E < 40 MeV

Ballistic damage with Fe (~10 dpa/h)

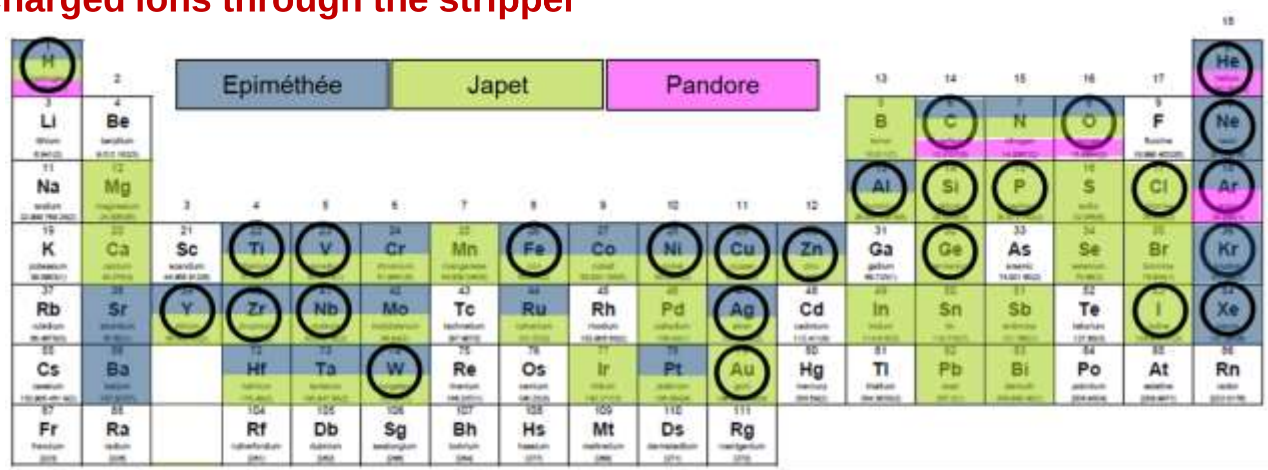
JANNuS-SACLAY: 2 MV Tandem Japet and 2.5 MV Pelletron Pandore

JAPET 2 MV Tandem with Source of Negative Ions by Cesium Sputtering (SNICS II)

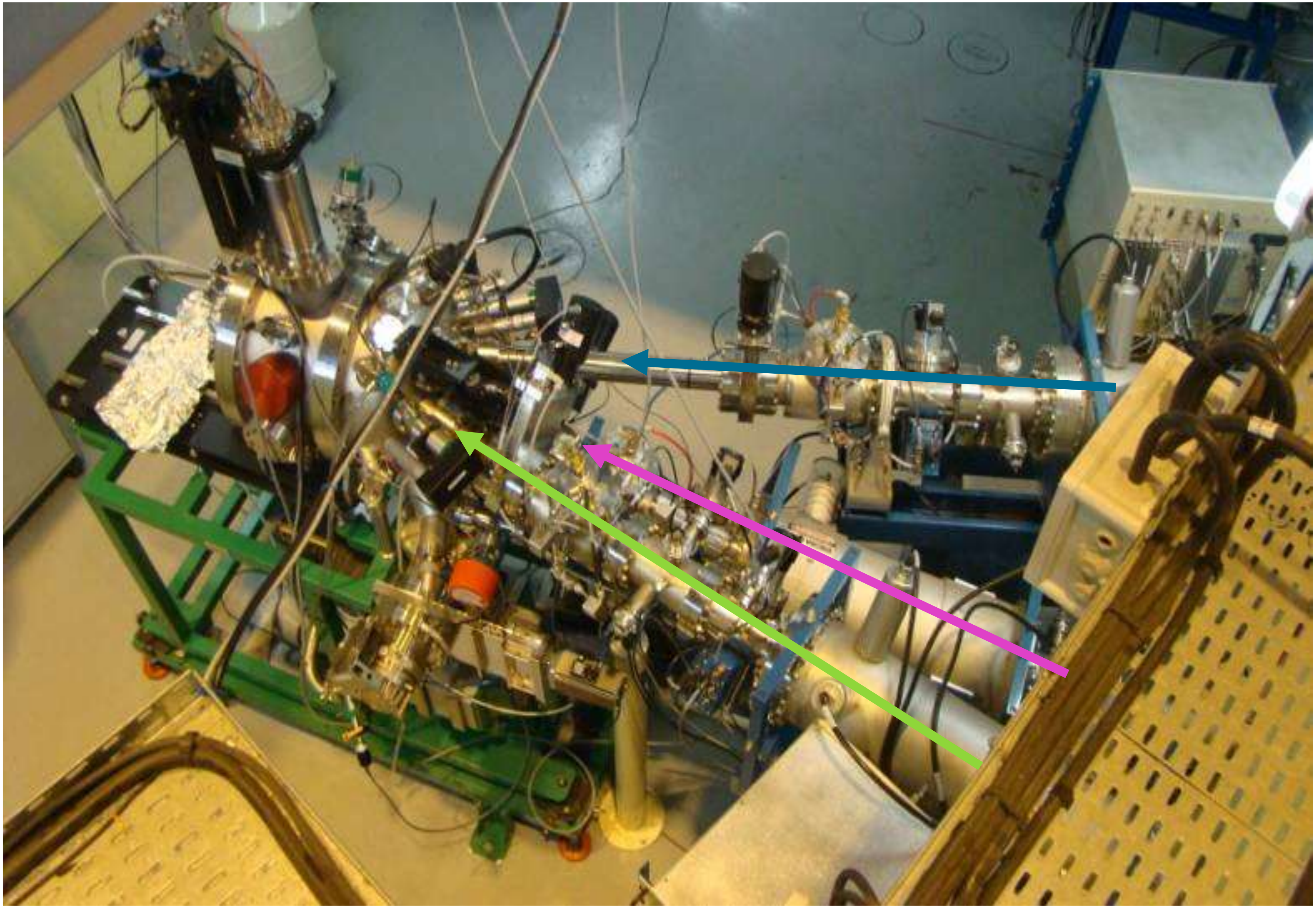
⇨ **Negative single-charged ions are converted into positive multi-charged ions through the stripper**

PANDORE 2.5 MV Pelletron with Radio Frequence source

⇨ **Single-charged gasses: H, He, D, N, Ar**



JANNuS-SACLAY: Irradiation chambers



Complementary laser in the Raman spectrometer

- new laser with 633 nm in addition to the 532 nm

Retrofit of the IBA chamber

- New detectors
- Specimen deck with 2 to 6 degrees of freedom allowing for canalization

High temperature chamber

- Extension of the single beam chamber for irradiation at homogenous temperature up to 1200°C

Cryogenic chamber with resistivity recovery measurement

- Cryogenic irradiation at 30K
- Controlled step by step annealing
- On line measurement of sample resistivity (defect population and mobility)

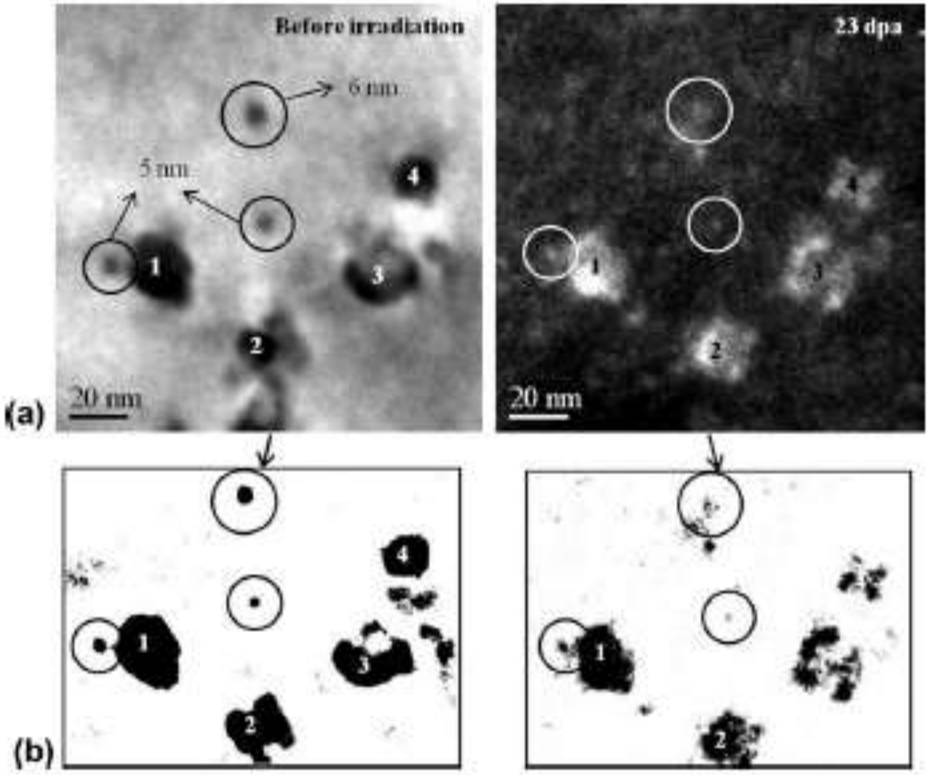
Radiation stability of nano-oxides in ODS steels

Example 1: M.-L. Lescoat, J. Ribis, Y. de Carlan et al.

In situ TEM at JANNuS-Orsay

- Nano particle evolution

~23 dpa



Fe^+ 150 keV, 500°C
φ 3.7 10^{12} ion.cm^{-2}.s^{-1}
Φ_{max} 1.6 10^{12} ion.cm^{-2}.s^{-1}
(~45 dpa)

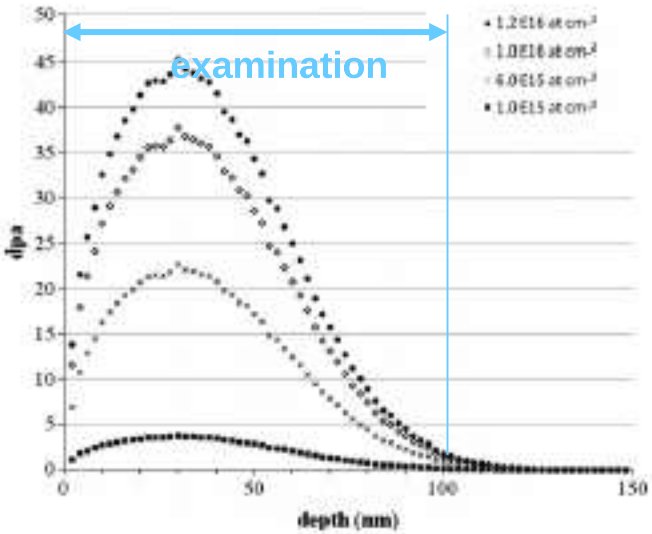


Fig. 1. Depth profiles of displacement damages in Fe calculated by SRIM.

M-L. Lescoat, J. Ribis, A. Gentils, O. Kaïtasov, Y. de Carlan, A. Legris, JNM 428 (2012) 176

Radiation stability of nano-oxides in ODS steels

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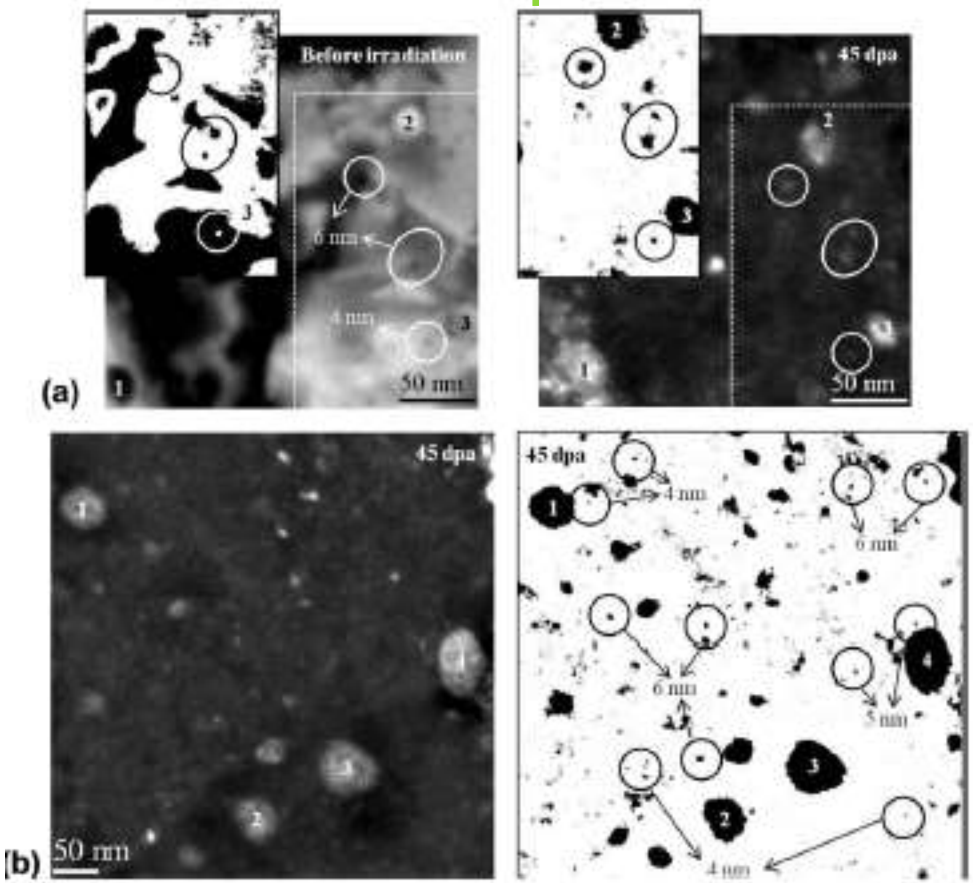


Fe^{+} 150 keV, 500°C
φ 3.7 10^{12} ion.cm^{-2}.s^{-1}
Φ_{max} 1.6 10^{16} ion.cm^{-2}.s^{-1}
(~45 dpa)

Nanoparticles are still visible after irradiation at ~45 dpa

M-L. Lescoat, J. Ribis, A. Gentils, O. Kaïtasov, Y. de Carlan, A. Legris, JNM 428 (2012) 176

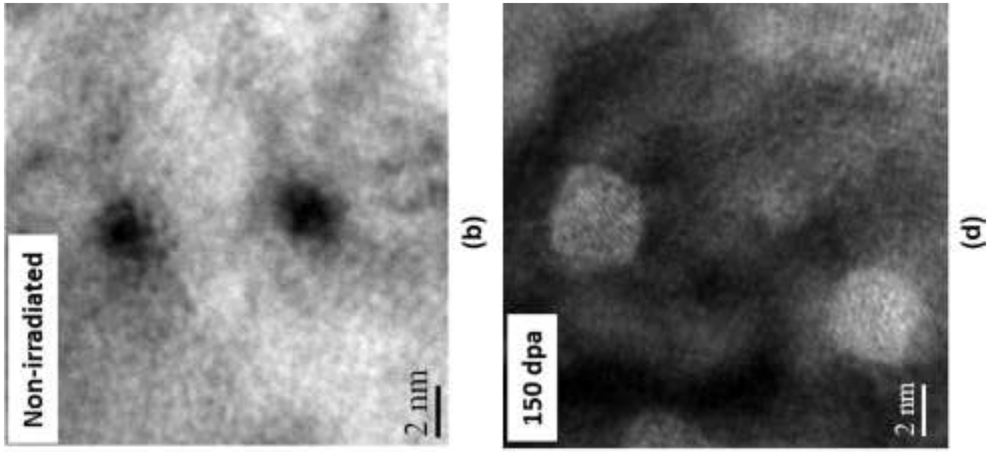
Radiation stability of nano-oxides in ODS steels

Example 1: M.-L. Lescoat, J. Ribis, Y. de Carlan et al.

High dose irradiation at JANNuS-Saclay

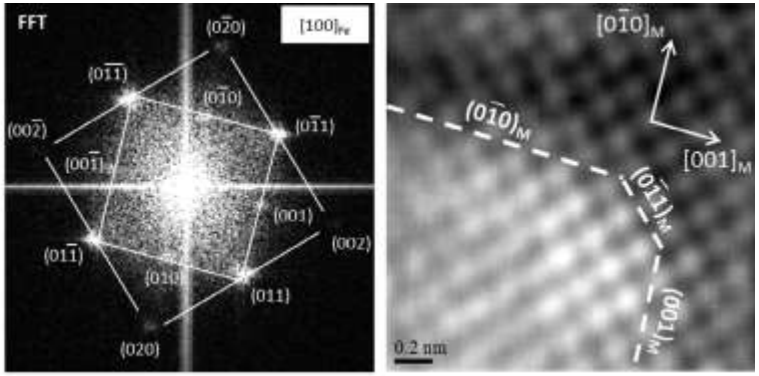
- Nanoparticle study

Fe^{+} 0.5 MeV, 500°C
φ 2.7 10^{12} ion.cm^{-2}.s^{-1}
Φ_{max} 8.9 10^{16} ion.cm^{-2}.s^{-1}
(~220 dpa)

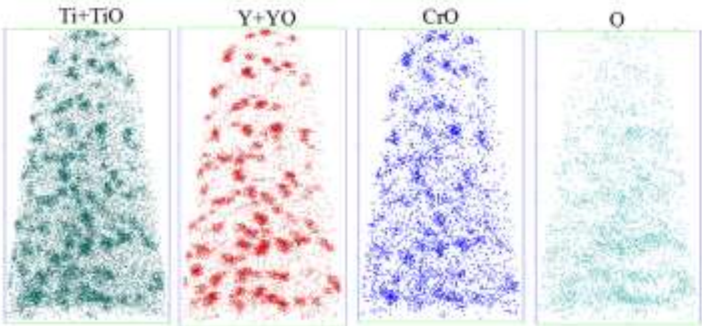


Nanoparticles are still dispersed after irradiation at high dose

FFT and HRTEM



3-D APT chemical reconstruction



M-L. Lescoat, J. Ribis, Y. de Carlan et al., Acta Materialia 78 (2014) 328

Radiation stability of nano-oxides in ODS steels

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High dose irradiation at JANNuS-Saclay

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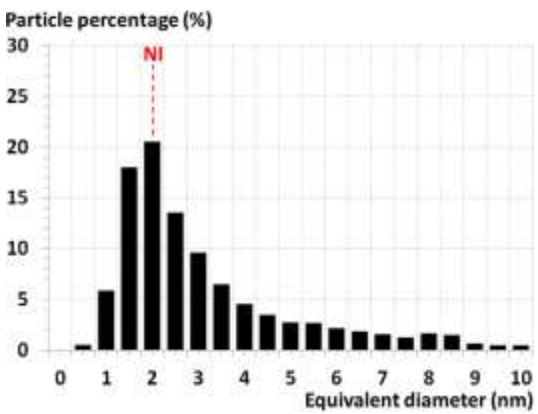
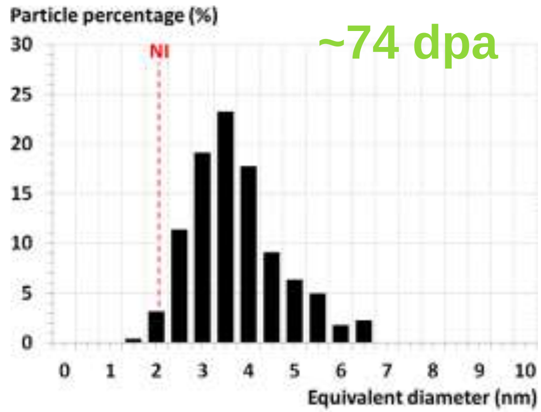
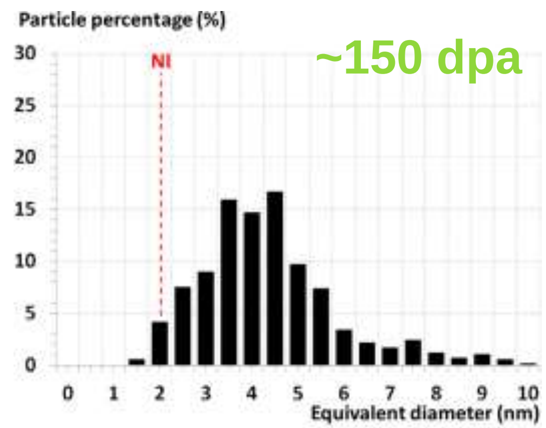
φ 2.7 10^{12} ion.cm^{-2}.s^{-1}
Φ_{max} 8.9 10^{16} ion.cm^{-2}.s^{-1}
(~220 dpa)

- Nanoparticle study

Statistics on nanoparticles distribution and size







Size increases and density decreases with increasing dose consistent with a Oswald ripening process

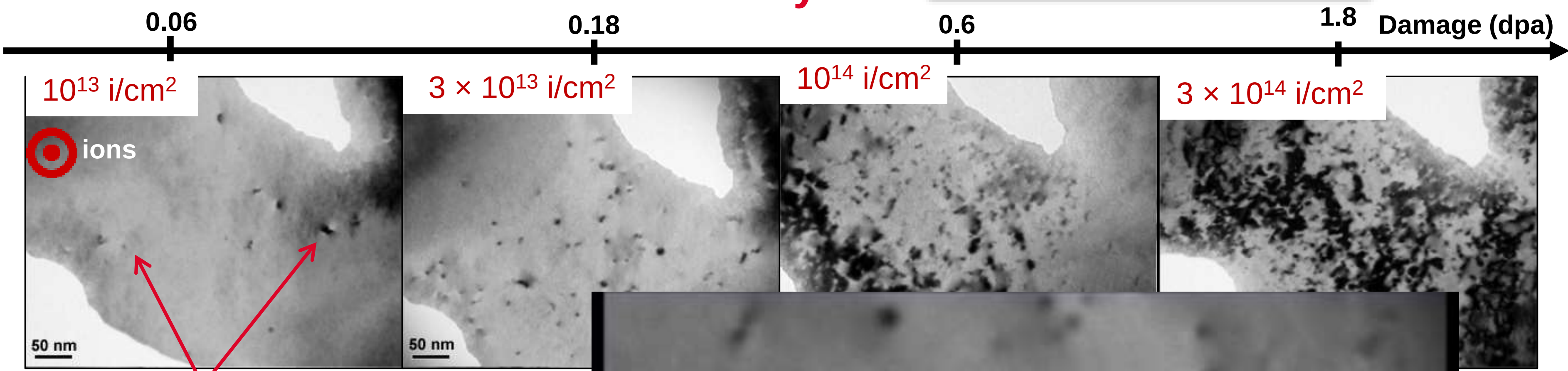
M-L. Lescoat, J. Ribis, Y. de Carlan et al., Acta Materialia 78 (2014) 328

Evolution of extended defects in UO_2 under irradiation

Example 2: C. Onofri, C. Sabathier, M. Legros et al.

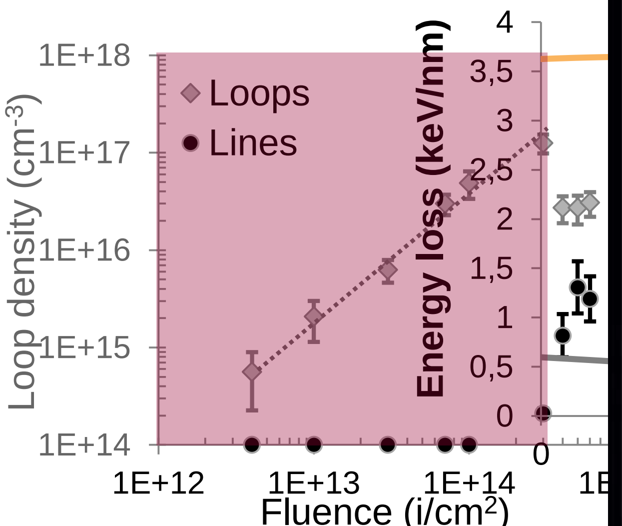
390 keV Xe at -180 °C

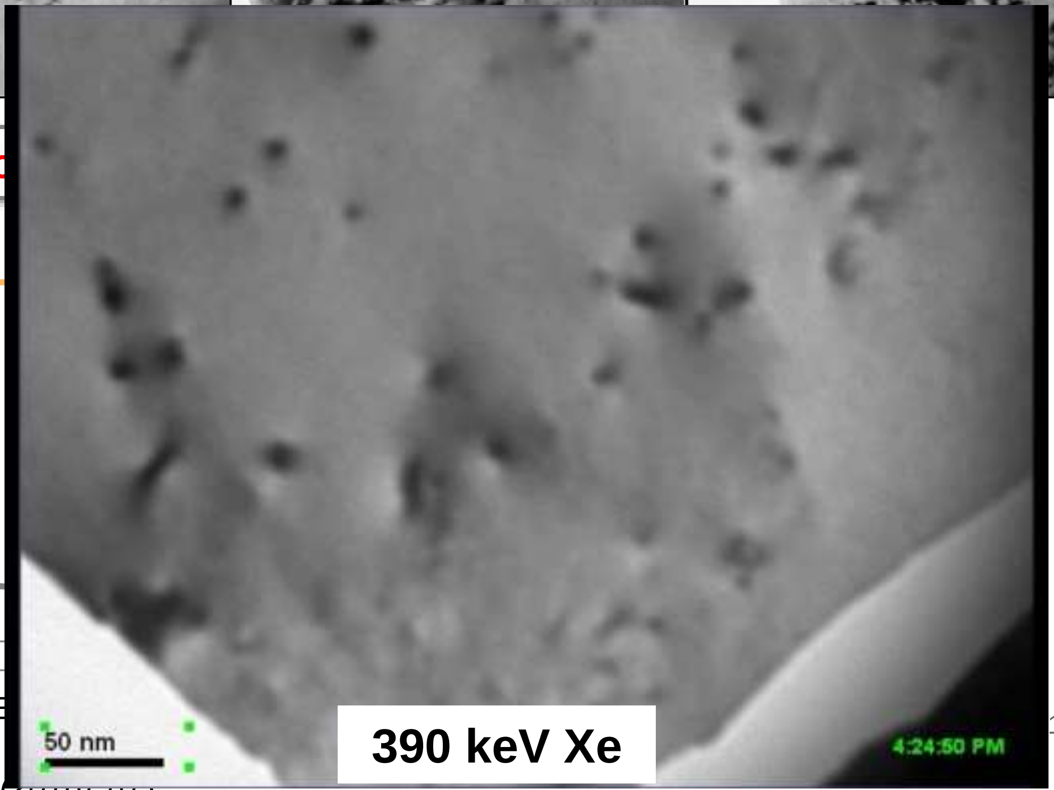
In situ TEM at JANNuS-Orsay



Defect clusters → **Future dislocation loops**

Small lo





C. Onofri, C. Sabathier, C. Baumier et al., JNM 482 (2016) 105

Evolution of extended defects in UO_2 under irradiation

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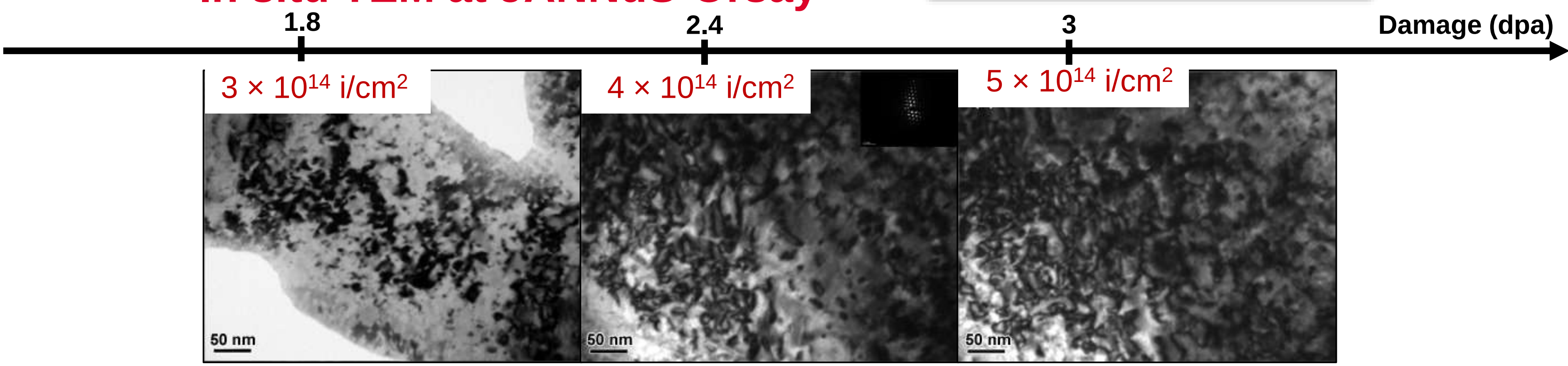
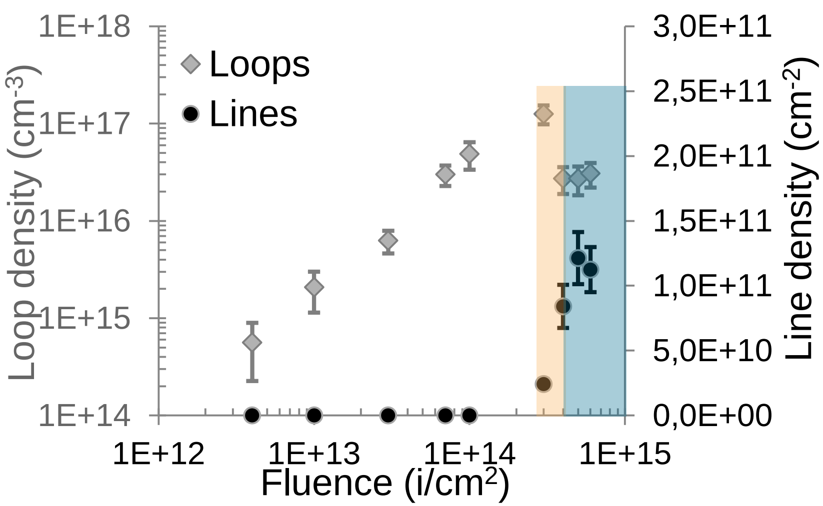
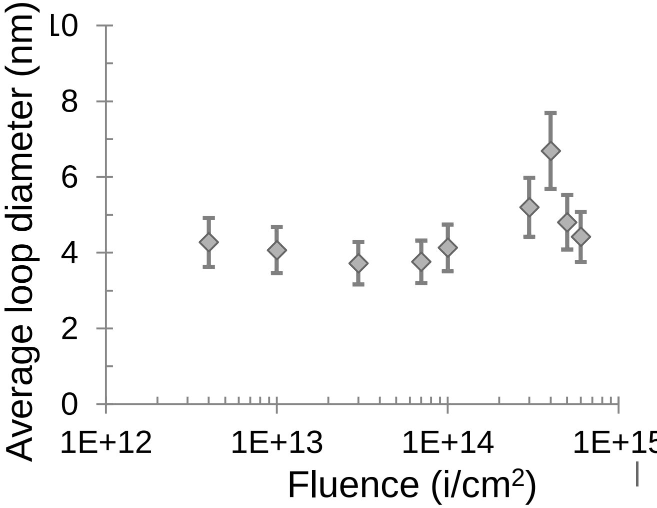


Loops overlapping for geometric reasons
→ Transformation into lines

Steady state equilibrium
→ Lines and small loops (< 10 nm)





C. Onofri, C. Sabathier, C. Baumier et al., JNM 482 (2016) 105

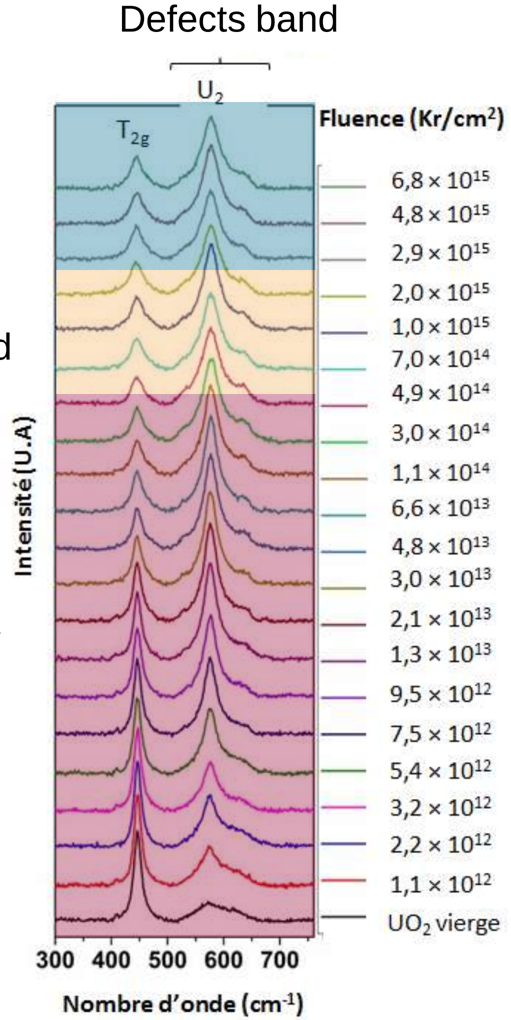
Evolution of extended defects in UO_2 under irradiation

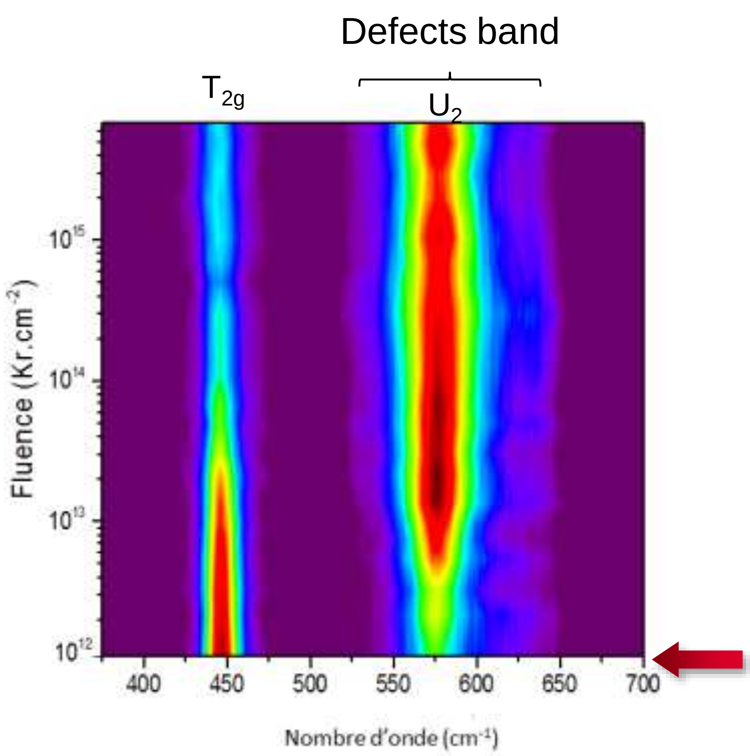
Example 2: C. Onofri, C. Sabathier, M. Legros et al.

4 MeV Kr at -160 °C

In situ Raman at JANNuS-Saclay

- No evolution
- Shrinking of defects band
- Decrease of T_{2g} intensity – *Loss of symmetry*
- Increase of defects band intensity – *Production of irradiation defects*





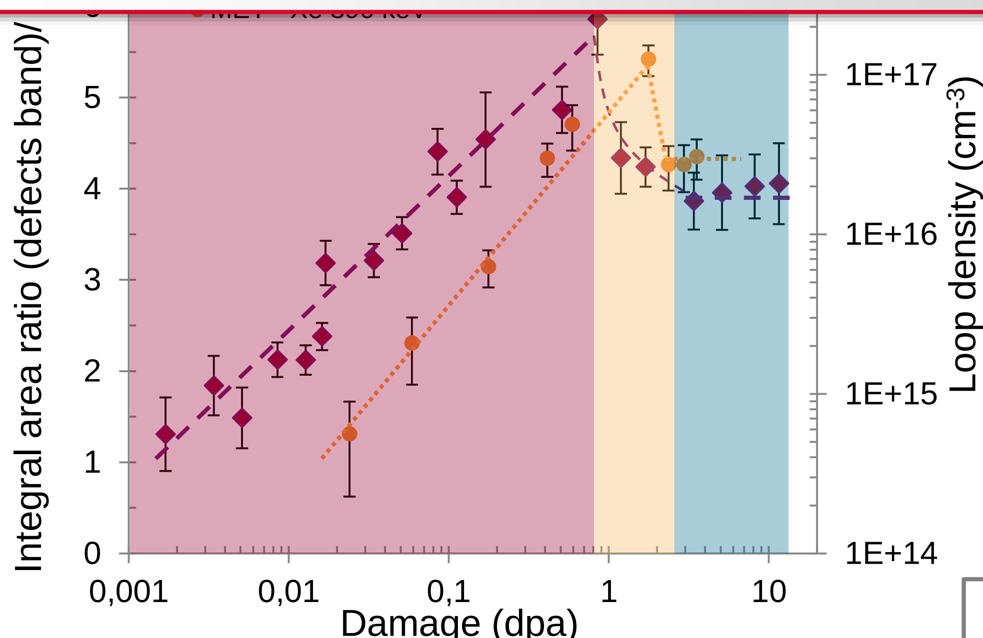
C. Onofri, C. Sabathier, H. Palancher et al., NIMB 374 (2016) 51

Evolution of extended defects in UO_2 under irradiation

Example 2: C. Onofri, C. Sabathier, M. Legros et al.

Good correlation between Raman and TEM characterizations

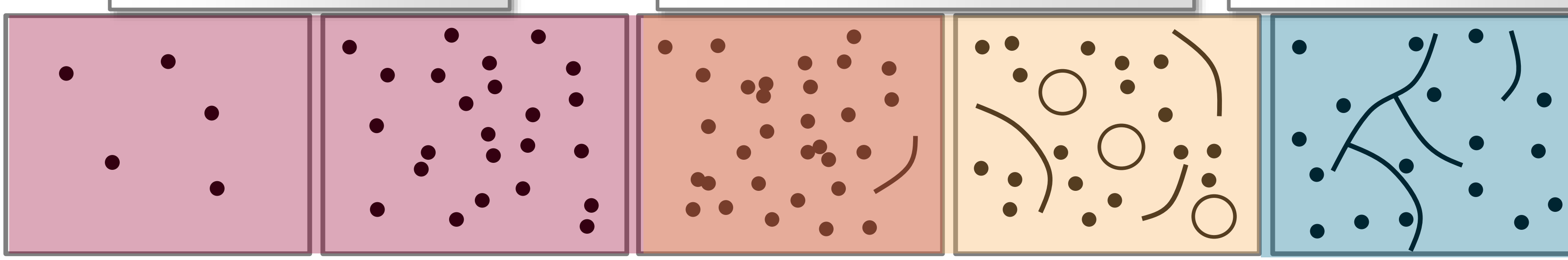
Extended defects could contribute to the defects band



Small loops nucleation stage (< 10 nm)

Loops transformation into lines by overlapping

Steady state equilibrium : Lines and continuous nucleation of small loops (< 10 nm)



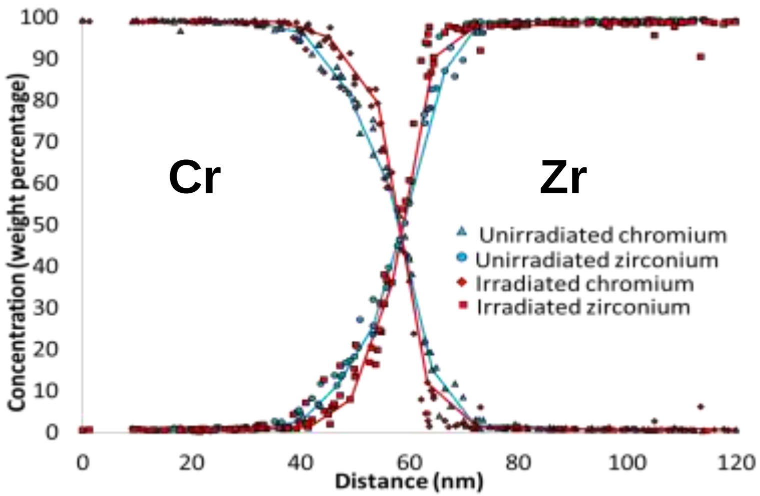
C. Onofri et al., EMIRUM2016, 20-21/10/16, Saclay, France

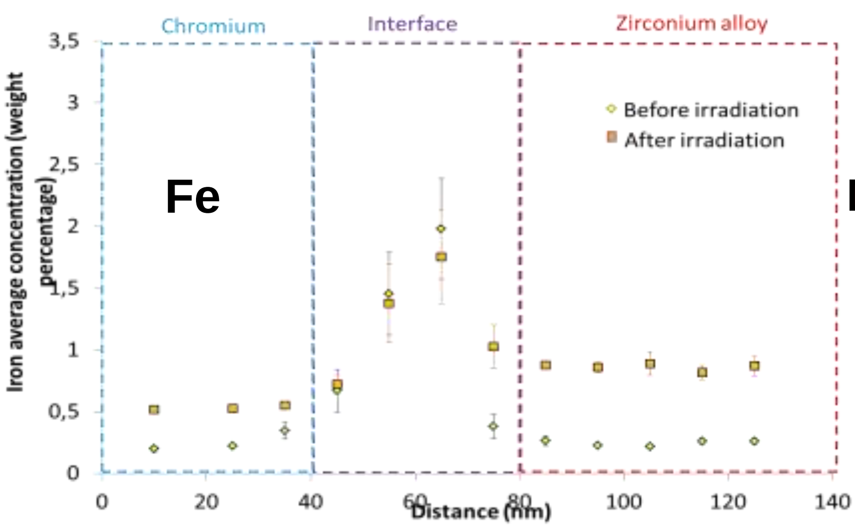
Radiation stability of a Cr-coating/Zr-alloy interface

Example 3: A. Wu, J. Ribis, J.C. Brachet et al.

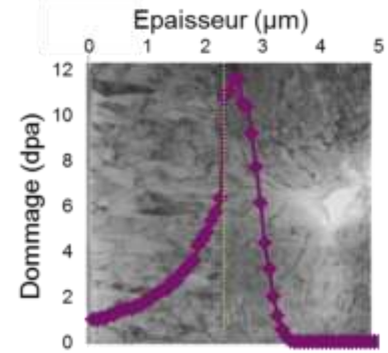
High energy irradiation at JANNuS-Saclay

- Chemical evolution at the interface (EDS)





Kr^{8+} 20 MeV, 400°C
φ 2.8 10^{11} ion.cm^{-2}.s^{-1}
Φ_{max} 6 10^{15} ion.cm^{-2}.s^{-1}
(~10-12 dpa at interface)



✓ **No Zr and Cr interdiffusion at ~10 dpa**

✓ **Slight Fe diffusion toward the interface**

A. Wu et al., EMIRUM2016, 20-21/10/16, Saclay, France

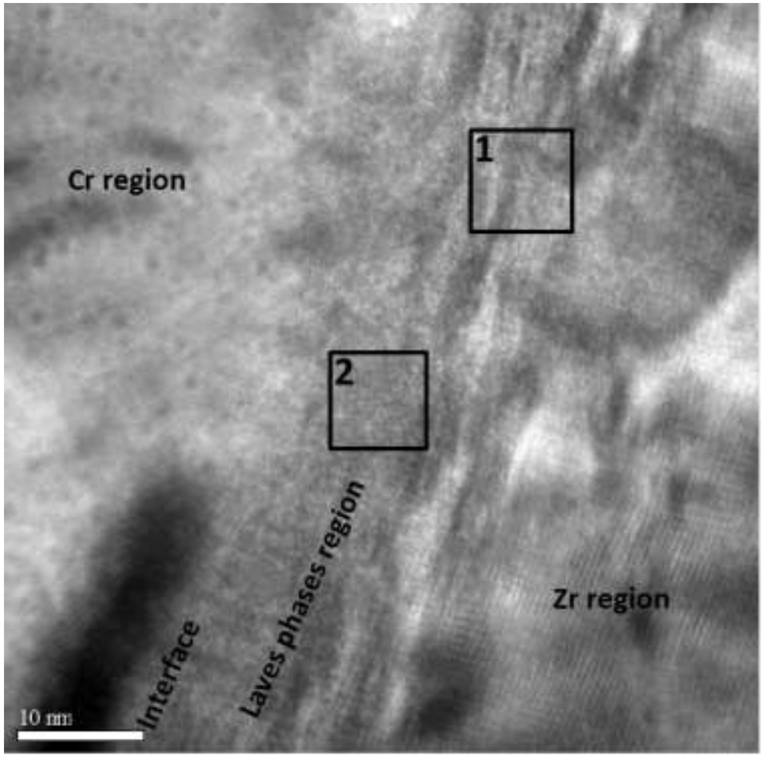
Radiation stability of a Cr-coating/Zr-alloy interface

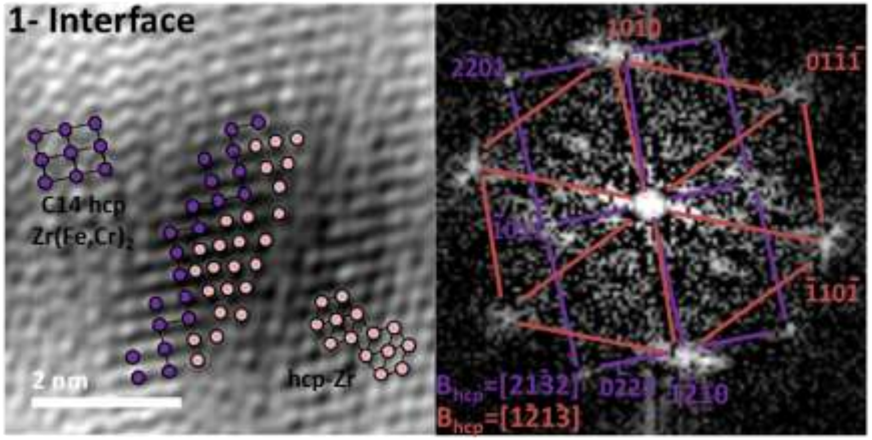
Example 3: A. Wu, J. Ribis, J.C. Brachet et al.

High energy irradiation at JANNuS-Saclay

- **Structural evolution at the interface (HR-TEM)**

Kr^{8+} 20 MeV, 400°C
φ 2.8 10^{11} ion.cm^{-2}.s^{-1}
Φ_{max} 6 10^{15} ion.cm^{-2}.s^{-1}
(~10-12 dpa at interface)





- ✓ **interface still crystalline and coherent at ~10 dpa**
- ✓ **Only the Laves phase C14 remains (no more the C15)**

A. Wu et al., EMIRUM2016, 20-21/10/16, Saclay, France

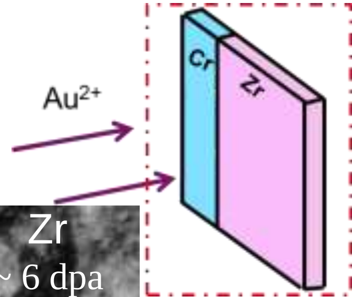
Radiation stability of a Cr-coating/Zr-alloy interface

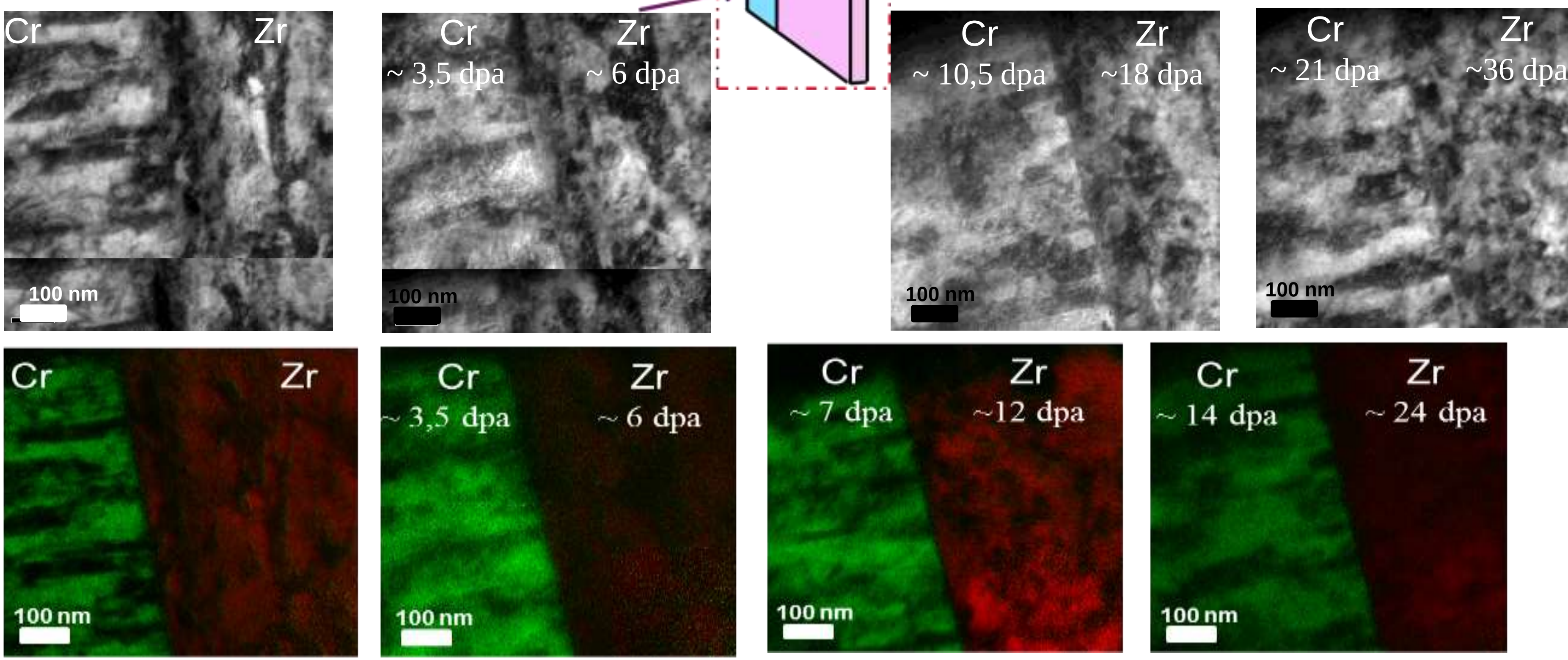
Example 3: A. Wu, J. Ribis, J.C. Brachet et al.

Interface evolution with in situ TEM

- EDS and EF-TEM

Au^{2+} 4 MeV, 400°C
φ 2.8 10^{11} ion.cm^{-2}.s^{-1}
Φ_{max} 4,8 10^{15} ion.cm^{-2}.s^{-1}
(~21dpa in Cr, ~36dpa in Zr)





A. Wu et al., EMIRUM2016, 20-21/10/16, Saclay, France

- ✓ **Defects density increases but the interface remains well visible**
- ✓ **Chemical stability with increasing irradiation dose**

Radiation stability of a Cr-coating/Zr-alloy interface

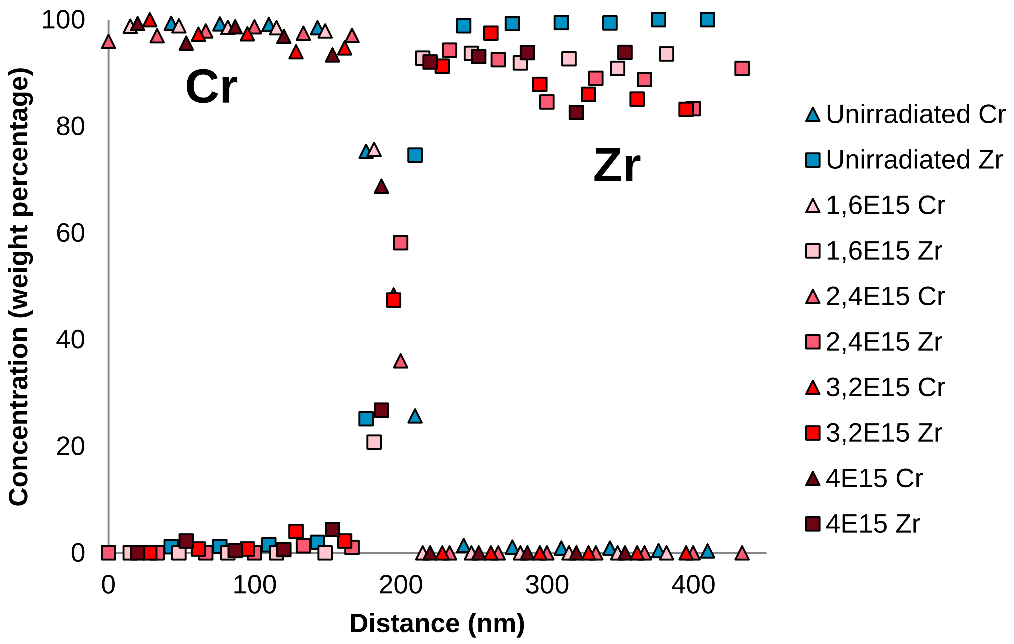
Example 3: A. Wu, J. Ribis, J.C. Brachet et al.

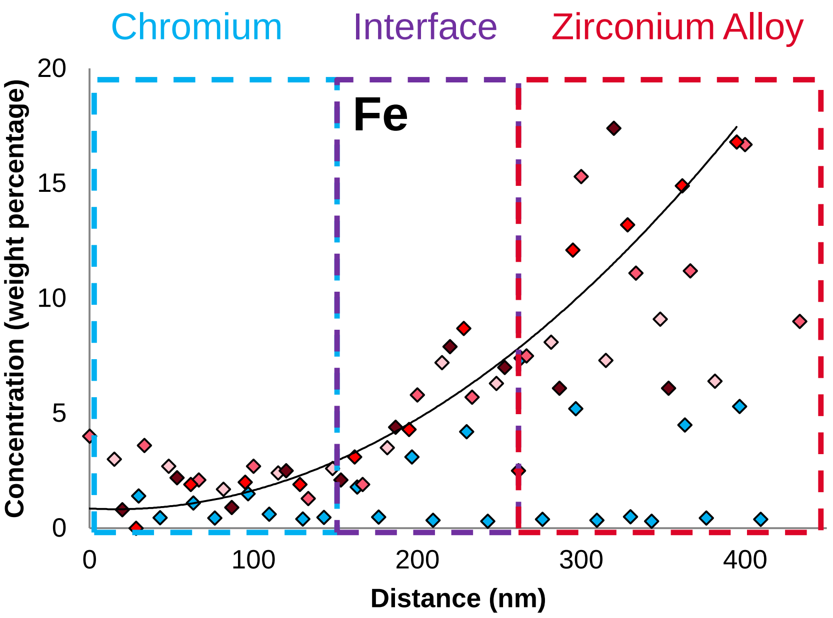
JANNUS

In situ examination of the interface

Au^{2+} 4 MeV, 400°C
φ 2.8 10^{11} ion.cm^{-2}.s^{-1}
Φ_{max} 4,8 10^{15} ion.cm^{-2}.s^{-1}
(~21dpa in Cr, ~36dpa in Zr)

- EDS up to 24dpa





✓ **Chemical profile Zr/Cr remains stable**
✓ **Fe diffusion toward interface increases with dose**

A. Wu et al., EMIRUM2016, 20-21/10/16, Saclay, France

JANNuS: two complementary facilities

- JANNuS = 5 coupled accelerators for cutting-edge researches on irradiation resistance of innovative materials
  - JANNuS-Orsay = dual ion beam with in situ TEM
  - JANNuS-Saclay = triple ion beam and high dose rate

  under the auspices of the new Université Paris-Saclay

- Among many studies, examples detailed in this presentation address
  - Radiation stability of nano-oxides in ODS steels for advanced reactors
  - Extended defects build-up in fuel under irradiation
  - Interface stability of a protective coating under irradiation

- National and international beam time access
  - EMIR: French accelerator network for Materials Study under Irradiation
    call for proposal 2018 has been issued with deadline on October 21st, visit http://emir.in2p3.fr/
    projects are evaluated by a Scientific Committee
    a financial contribution with reduced fee for EMIR accepted proposals is requested
  - European projects in the frame of H2020 or Eurofusion
  - With charges upon request