Behavior of metals under irradiation

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Specificity of material science under irradiation

System (the material) maintained permanently far from equilibrium

Permanente injection of energy (primary damage)

Permanente energy and matter fluxes in various directions in the system

Stationary state far from equilibrium

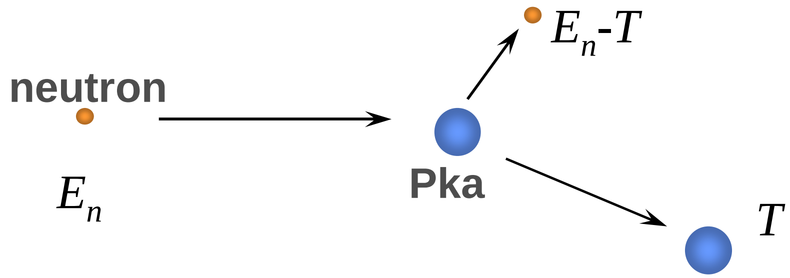
- Original microstructures
- Original micro-chemistry
- Shape changes
- Specific creep mechanism

Tendency to come back towards equilibrium (restoration)

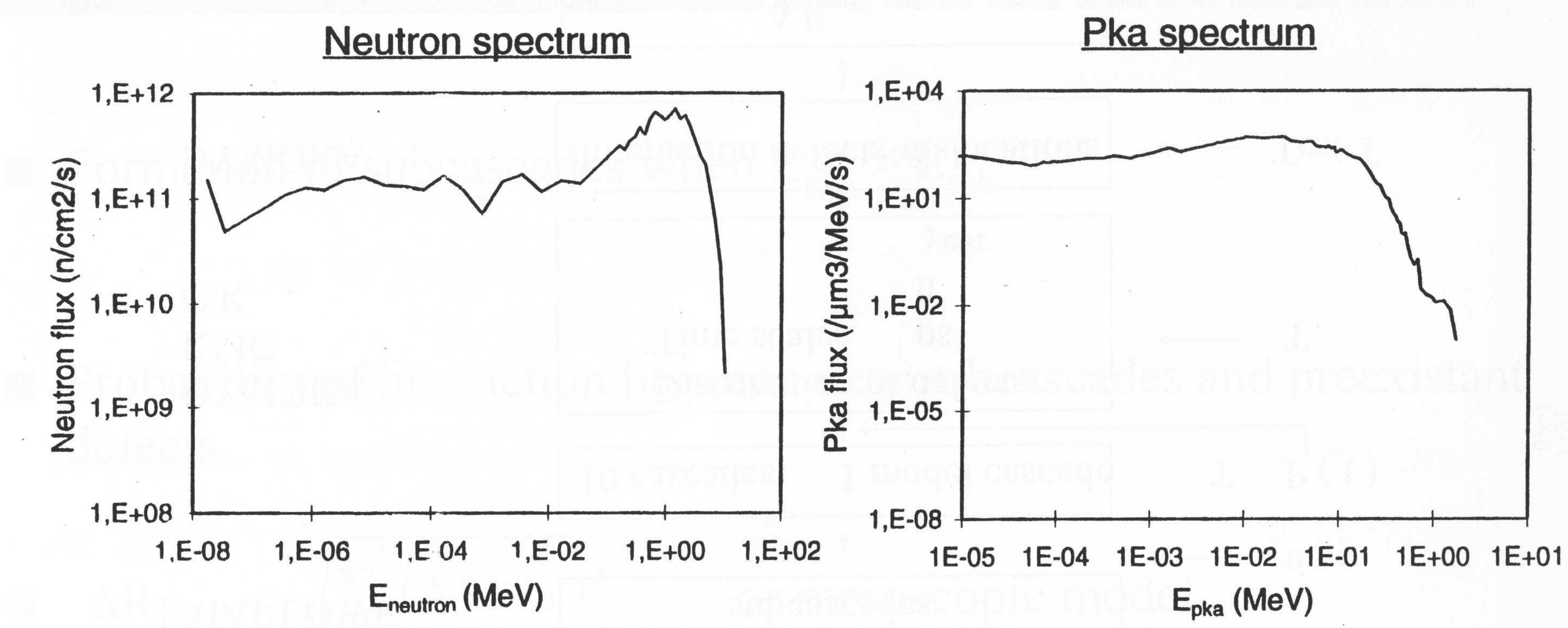
Metallurgy under irradiation

- Ballistic damage (Primary damage)
  - Neutron-atom collisions
  - Displacement cascades
  - Disorder induced by the ballistic damage
    - Chemical disorder
      - Ordered alloys
      - Precipitate resolution
    - Amorphization
- Properties of point defects and point defect clusters
  - Structure
  - Mobility
- Slow evolution (secondary damage)
  - Pont defect population evolution
  - Consequences of the point defect supersaturation
    - Point defects agglomeration
    - Phase transformation acceleration
    - Out of equilibrium segregation and precipitation
- Macroscopic consequences
  - Hardening
  - Void swelling
  - Irradiation creep
  - Irradiation growth

Ballistic damage : collisions neutrons – atoms



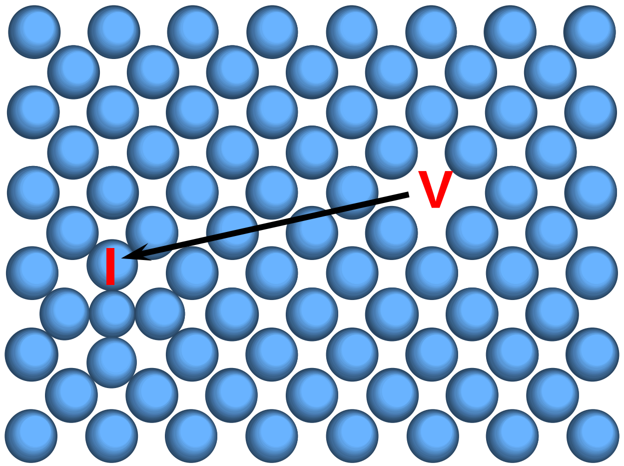
Pka = primary knock-on atom
T = kinetic energy transferred to the Pka

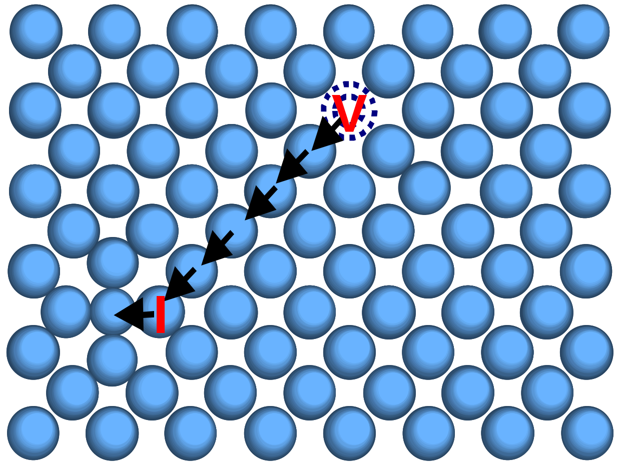


Ballistic damage : displacement threshold

For a low energy T transmitted, a only one Frenkel paire (vacancy + self interstitial atom) is create

T > E_d displacement threshold

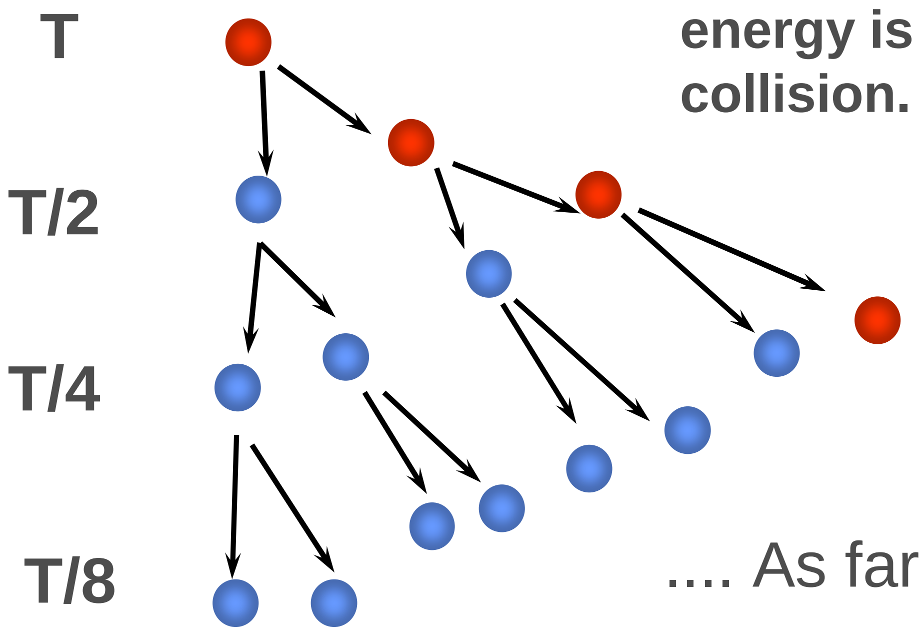




In compact crystallographic structures as metal

Ballistic damage: displacement cascades

If $T \gg E_d$, displacement cascade

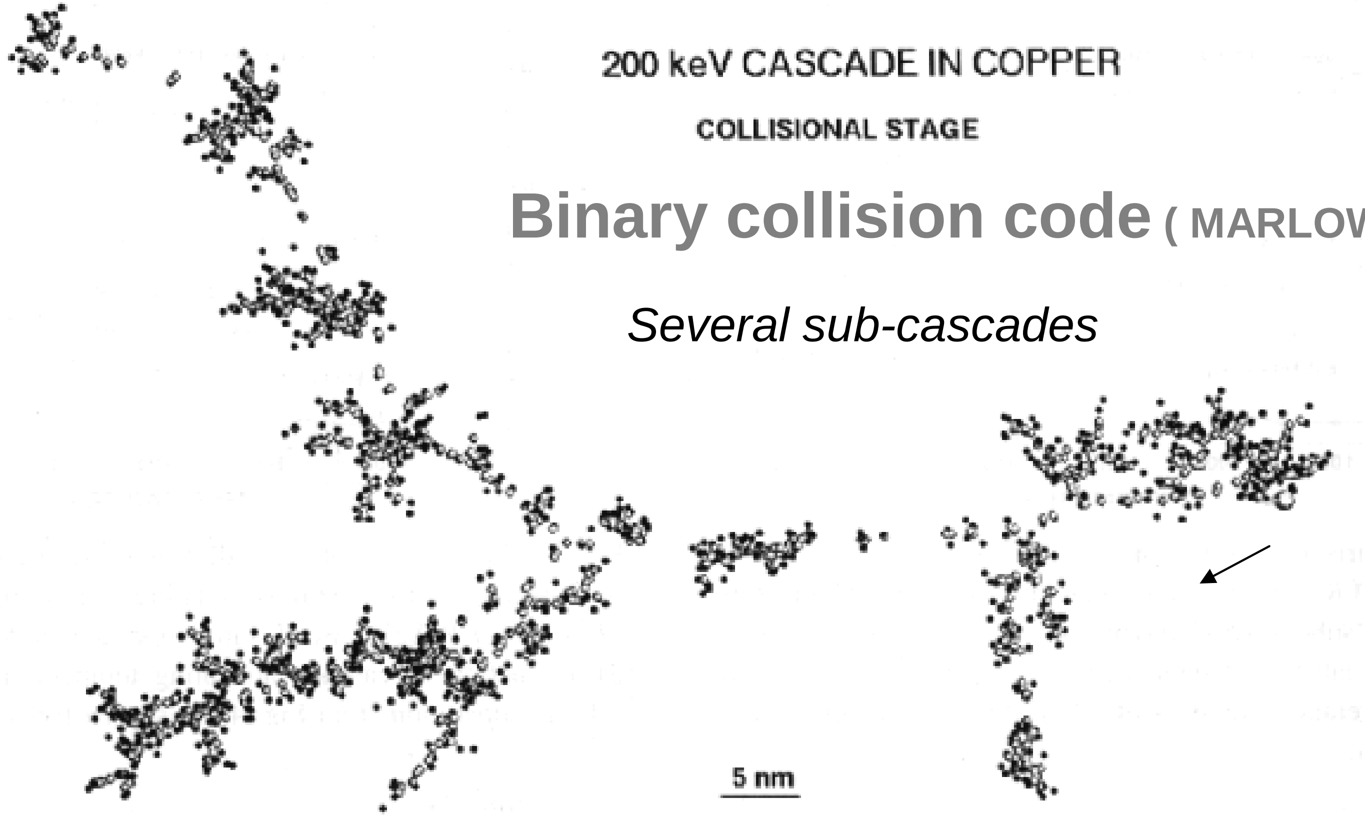


Ultra simple model assuming that the energy is share equally in every collision.

.... As far as $E > E_d$

Frenkel pair number: $n = T/(2.E_d)$

Balistic damage



Binary collision code **(MARLOWE)**

Several sub-cascades

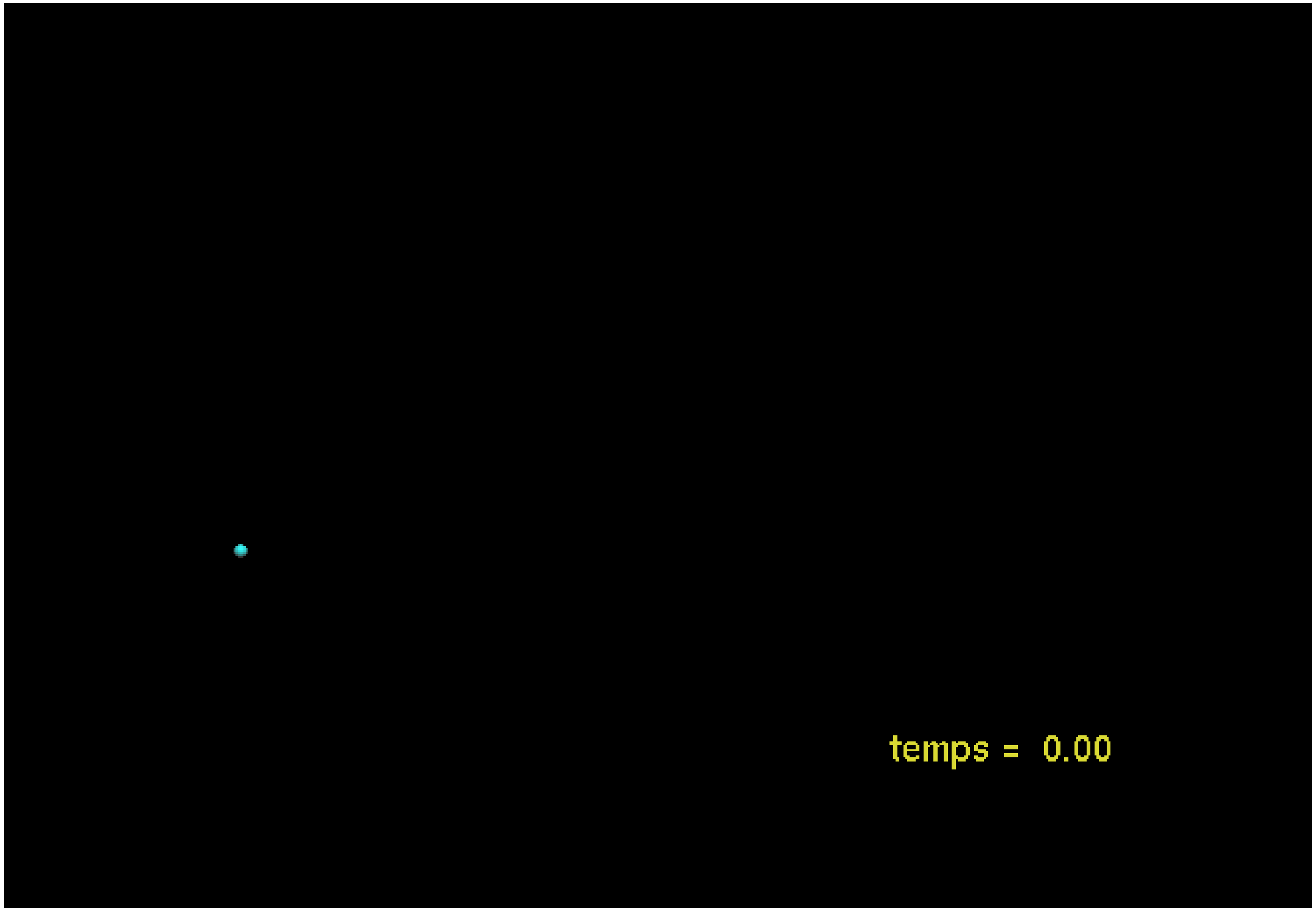
Number of Frenkel pairs $n = 0.8\hat{E}/2E_d$

Ê : Tranfered energy in elastic collision

⇒ **dpa_{NRT}**

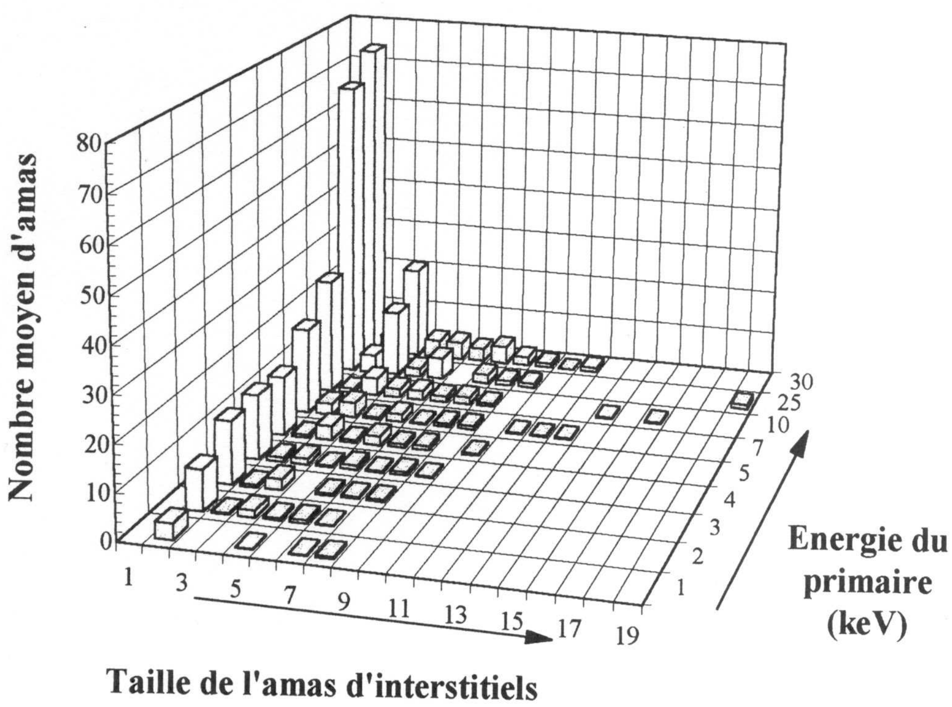
Ballistic damage: displacement cascades

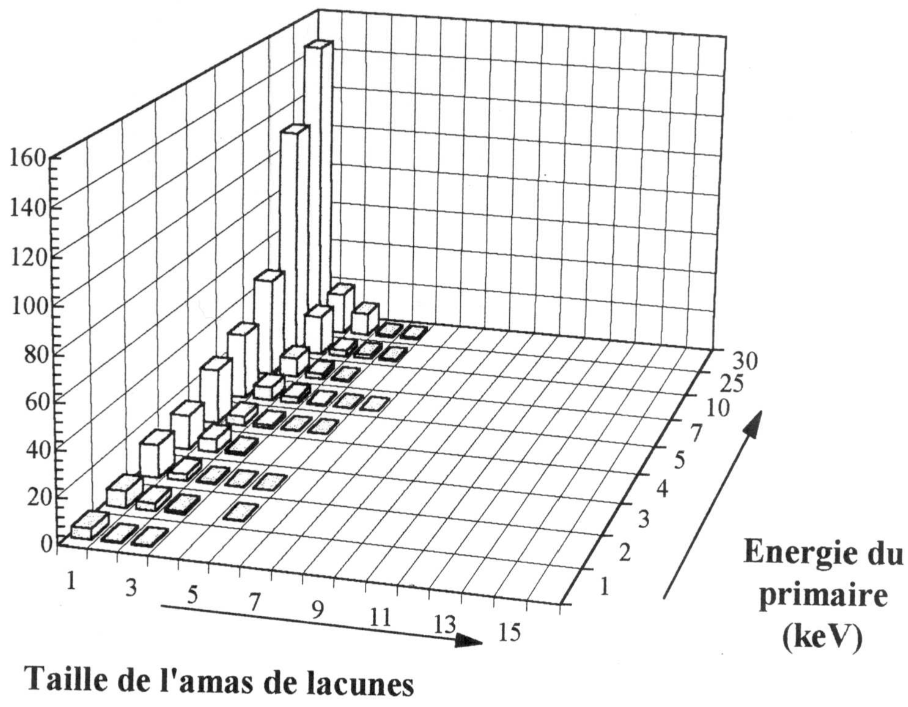
Molecular Dynamics (MD) simulation of a 10 keV cascade in iron (Doan SRMP)



Point defect clusters induced in cascade

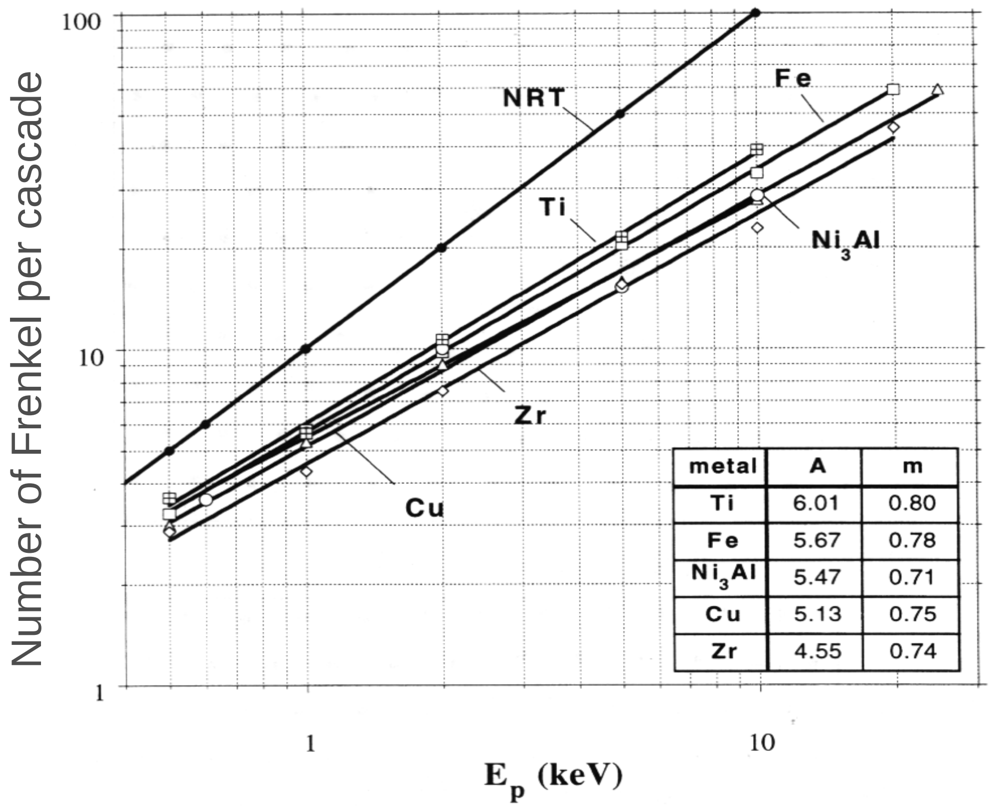
A significant proportion of point defects are in clusters





Vacancy and SIA clusters created in a cascade induced in Fe as a function of the PKA energy

dpa MD versus dpa NRT



metal	A	m
Ti	6.01	0.80
Fe	5.67	0.78
Ni_3Al	5.47	0.71
Cu	5.13	0.75
Zr	4.55	0.74

→ **Dpa's given by MD are significantly smaller than dpa NRT**

Clustering of point defects in cascade cores → **Dpa not sufficient to describe primary damage**

IRRADIATION CONDITIONS

Elément	Matériau	T°C	n m^{-2} s^{-1}	dpa/s	dpa end of life (estimated)
PWR Vessels	Low alloyed ferritic steels	290	10^{15}	3. 10^{-10}	0.2 (60 years)
PWR Fuel claddings	Zirconium alloys	345 - 420	10^{17}	3. 10^{-8}	4 (5 years)
PWR Internals (screws)	Austenitic steels	370	10^{17}	3. 10^{-8}	55 (60 years)
Fuel claddings FBR	Austenitic steels	550°C	10^{17}	1. 10^{-6}	150 (4 years)

Charged particles	dpa/s	
e^- (1-2 MeV) HV Electron Microscope	10^{-3} - 10^{-5}	Thin foil (500 nm)
e^- (1-3 MeV) Accelerators	10^{-8} - 10^{-9}	“Bulk” (0.5 mm)
Ion accelerators (1-30 MeV)	10^{-3} - 10^{-5}	“Bulk” (some microns under the surface)

Disorder induced by the ballistic damage

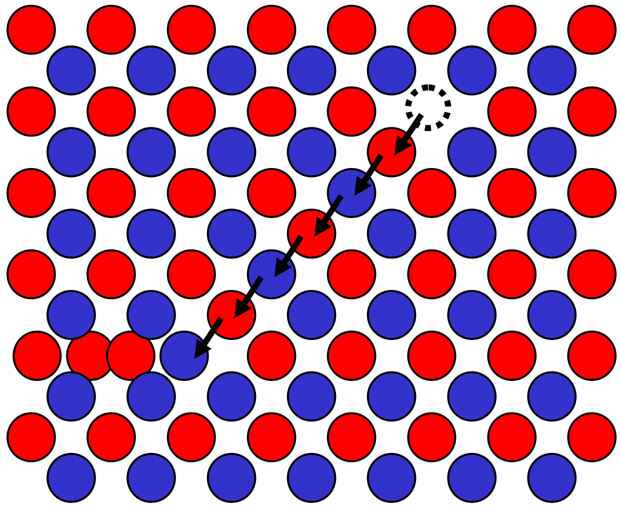
Usually, irradiation and observation at low temperature

Disorder induced by the ballistic damage

Replacement sequences

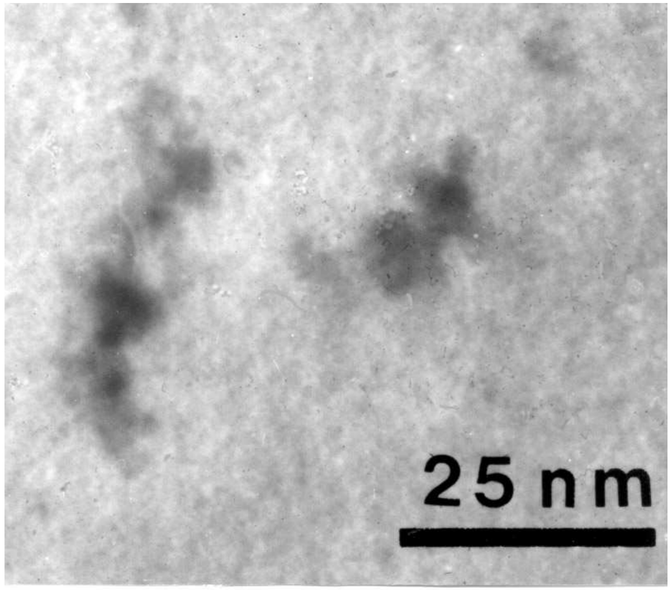
↓

Chemical disorder

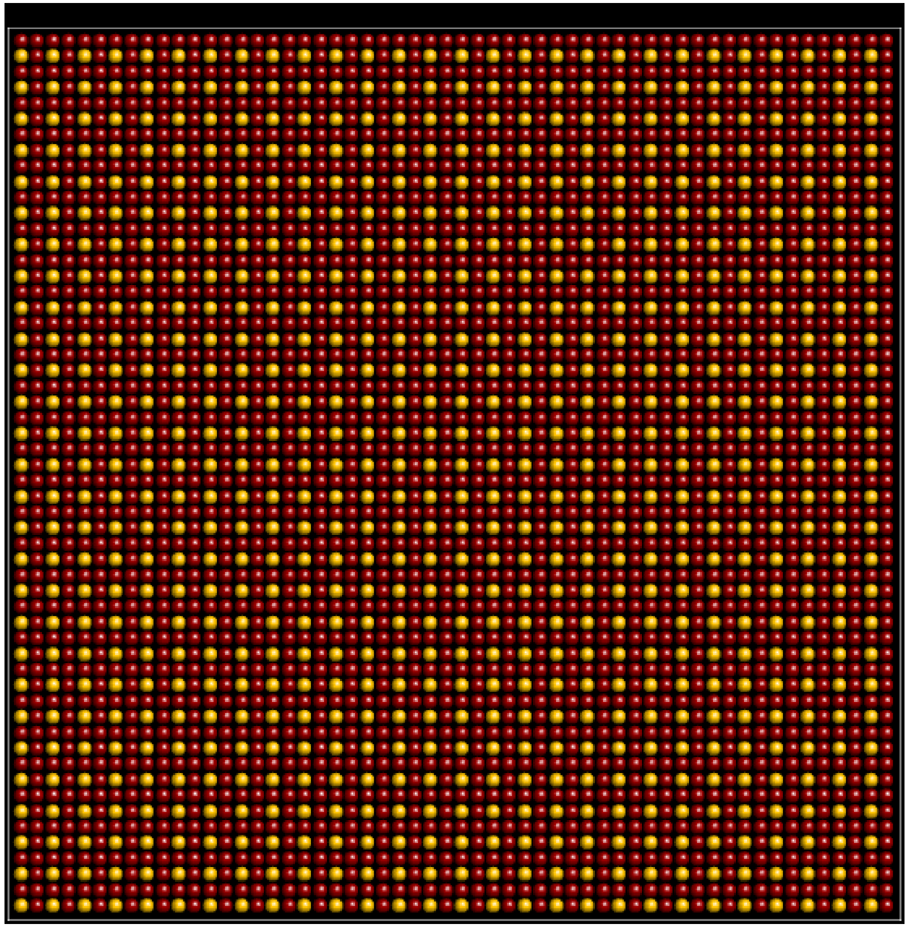


Disorder induced by the ballistic damage

Chemical disorder induced by desplacement cascades in ordered alloys (Cu_3Au)

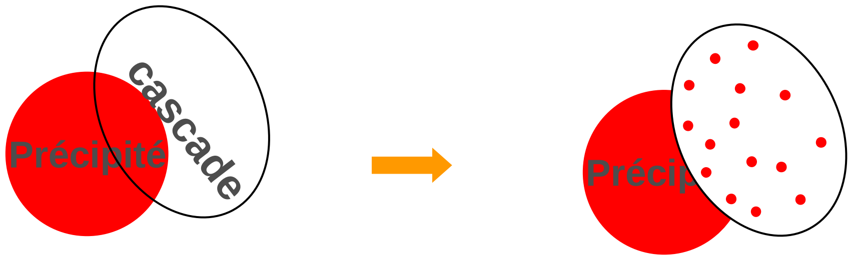


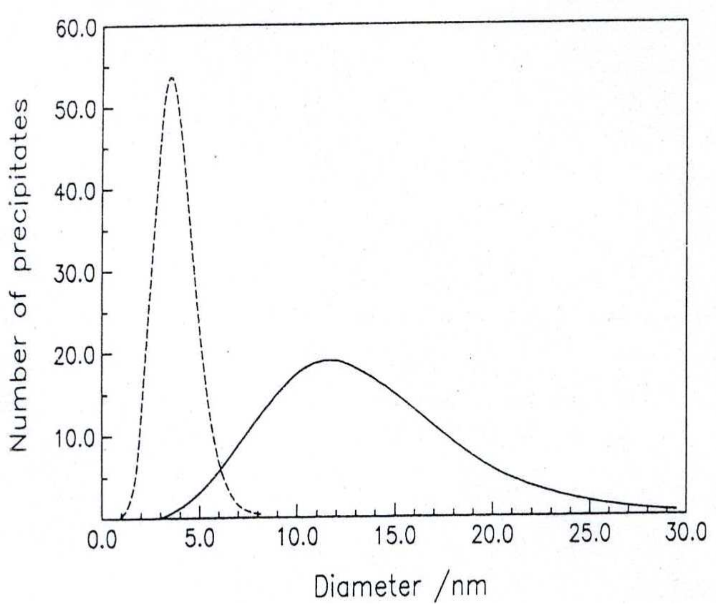
TEM

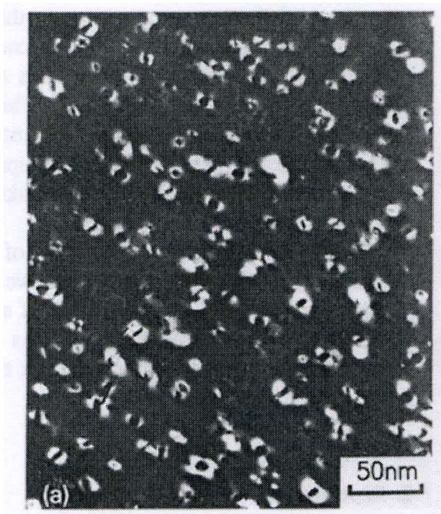


MD simulation

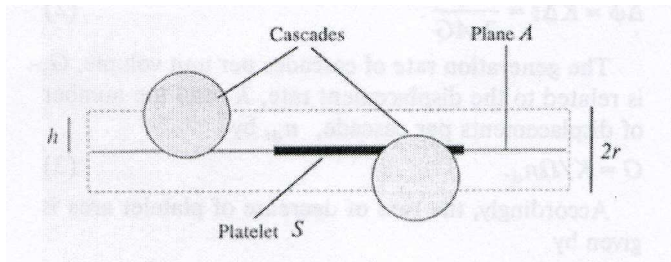
Ballistic resolution of précipitates





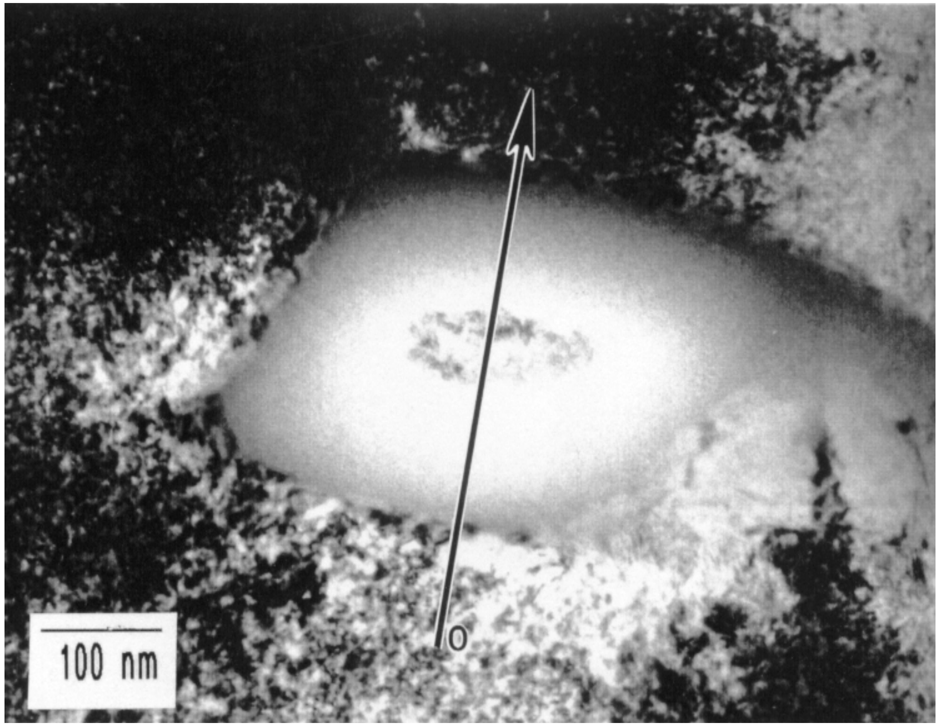


Size distribution of coherent precipitates in the CuCrZrSi alloy before (———) and after irradiation at 80 K, 0.06 dpa (··········) at 2.3e^{-5} dpa/s. No more precipitates observed at 0.1 dpa.

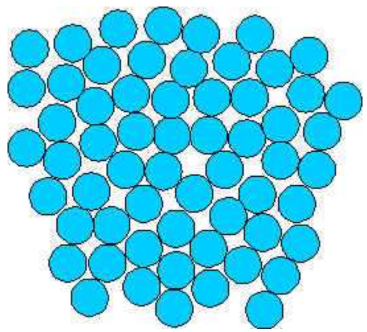


Amorphization

Destruction of the crystalline lattice in cascades



Amorphization under irradiation of a $Zr(Fe,Cr)_2$ precipitate in Zircaloy with resolution of Fe.



Amorphous state

Nuclear reaction

- Capture

  Recoil energy → dpa

$$^{27}Al\ (n, \gamma)\ ^{28}Al \Rightarrow\ ^{28}Si$$

$$^{30}Si\ (n, \gamma)\ ^{31}Si \Rightarrow\ ^{31}P$$

$$^{10}B\ (n, \alpha)\ ^{7}Li$$

- Fission

$$^{A}_{Z}M\ (n^{f}, \alpha)\ ^{A-3}_{Z-2}M'$$

$$^{A}_{Z}M\ (n^{th}, f)\ PF^{1} + PF^{2}$$

$$\begin{cases} ^{58}Ni\ (n^{th}, \gamma)\ ^{59}Ni \\ ^{59}Ni\ (n^{th}, \alpha)\ ^{56}Fe \end{cases}$$

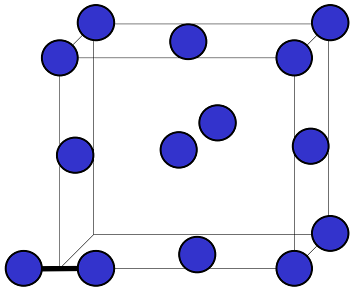
Hélium

in-situ creation of gas

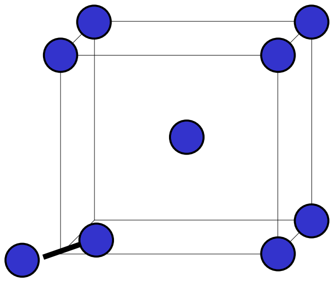
Ballistic damage : summary

- Point defects : vacancies and SIA’s
- Small point defect clusters
- Chemical disordering
- Amorphization for some compounds
- Creation of new chemical species in nuclear reactions

Point defect structures



fcc (austenitic steels)



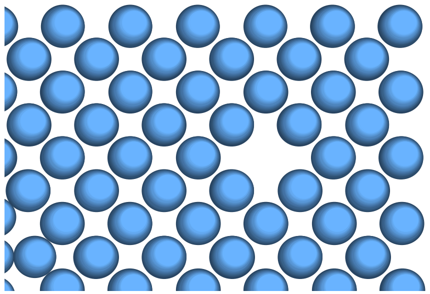
bcc (ferritic steels)

SIA dumbbell in compact cristalline structures

. H^f_I de 3 à 5 eV

Vacancy

H^f_v de 0.6 à 2 eV



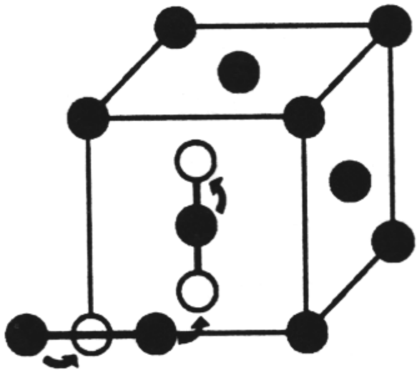
Equilibrium concentration of point defects

$$c_{DPe} = \exp\left(-\frac{H^f_{DP}}{kT}\right)$$

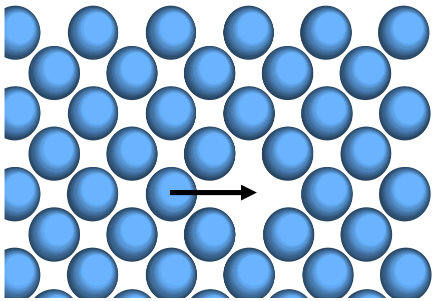
Formation energy

Point defect mobility

fcc SIA



vacancy



$$D_{DP} = D_{DP0} \exp\left(-\frac{H_{DP}^{m}}{kT} \right)$$

Migration energy eV

	VACANCY	SIA
	$H^{m,v}$,	$H^{m,i}$
Ni	1.1	0.15
Fe	1.3 (0.6)	0.3
Zr	0.93	0.06 – 0.15

Point defects mobility: consequences

- **No SIA's at equilibrium**
- **Vacancies at equilibrium**
- **SIA's usually very mobile**
- **Vacancies les mobile**

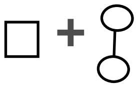
Mobility of point defects $\rightarrow$ slow kinetics

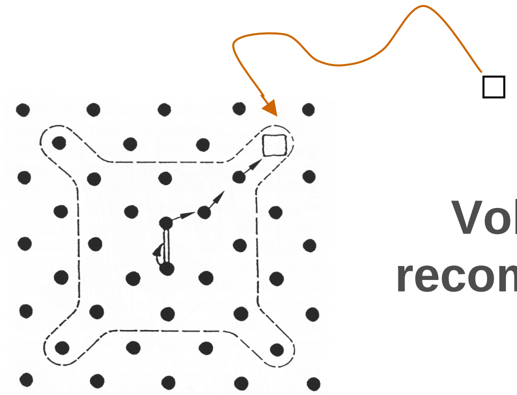
$\downarrow$

Long term evolution of the microstructure

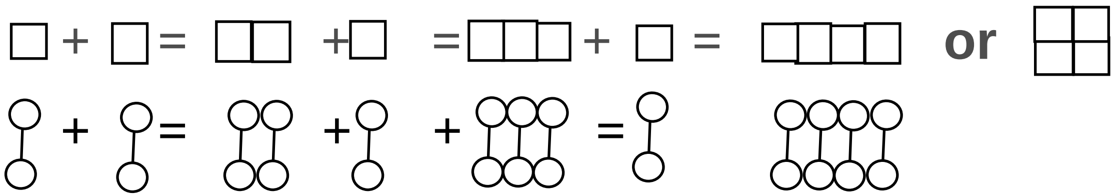
Slow kinetics
Point defects population evolution

- Annihilation (recombinaison)

 = nothing



- clustering

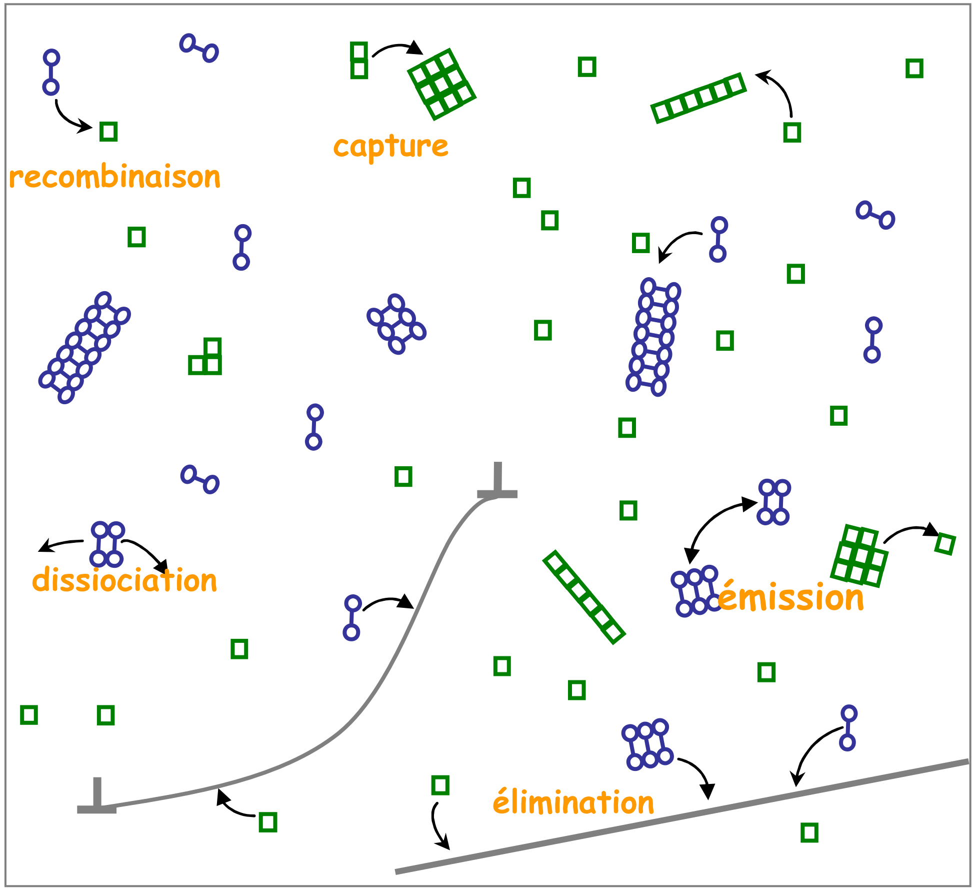


- Elimination on fix sinks
  - Dislocations (including loops)
  - Grain boundaries
  - Free surfaces, voids, bubbles

Slow kinetics

Point defect population evolution

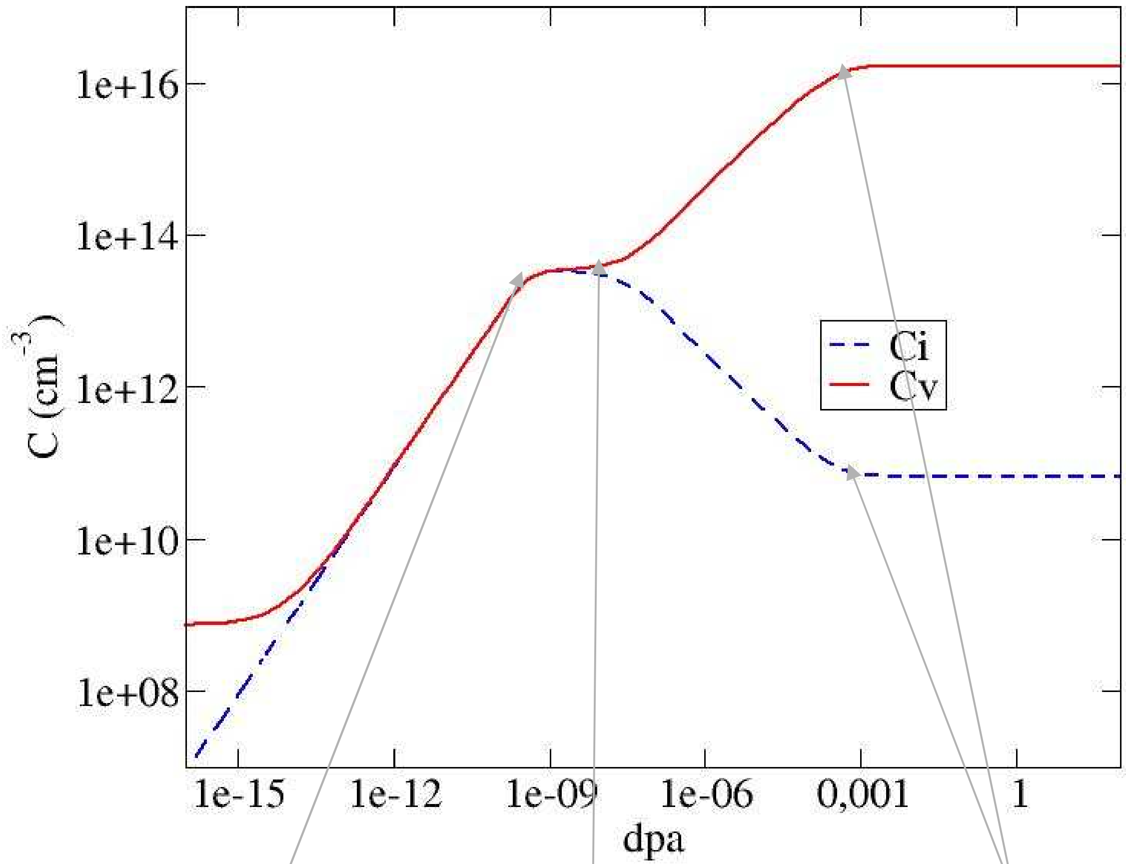
Summary



Slow kinetics

Point defect population evolution

Iron, 2 10^{-8}dpa/s, 300℃, low sink density



recombinations

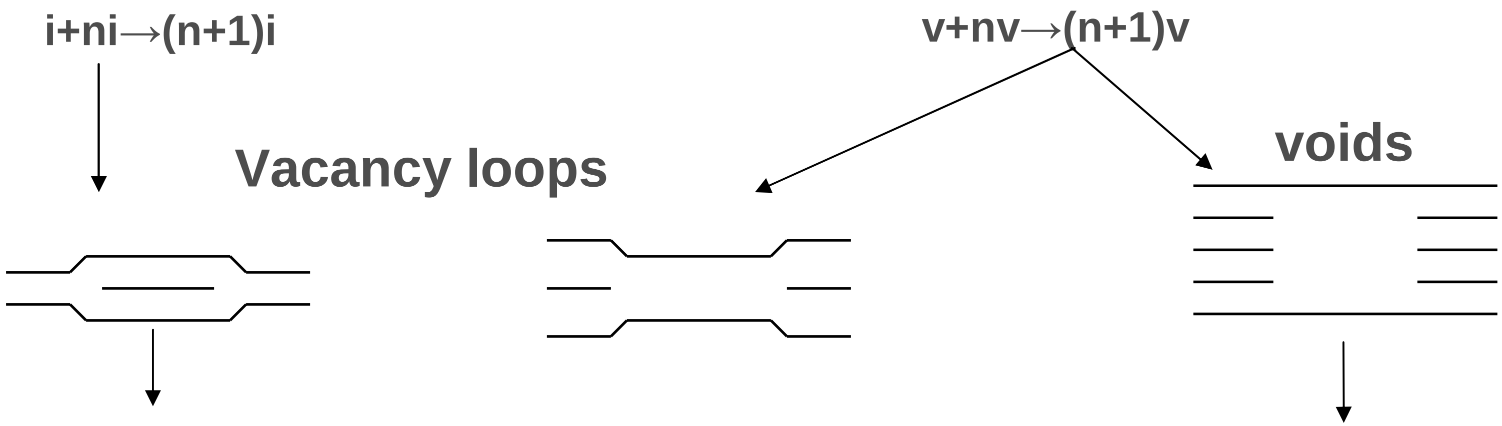
SIA's reach sinks

Vacancies reach sinks

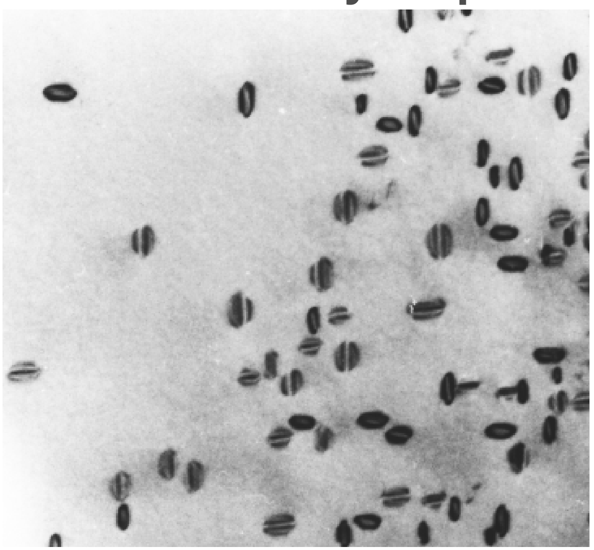
Slow kinetics

Point defect population evolution

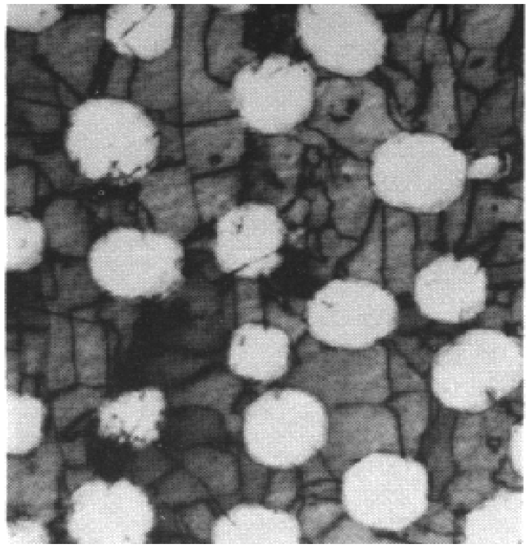
Voids and dislocation loops



Interstitiel vacancy loops in Ni



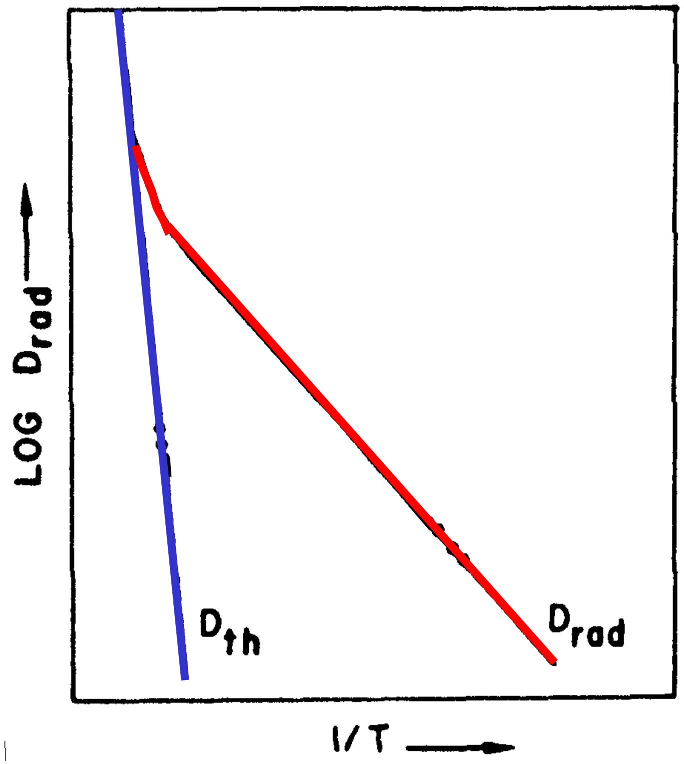
Void in Cu



Slow kinetics

Enhanced diffusionDiffusion accélérée

Enhanced phase transformation



Point defect super-saturation

⇩

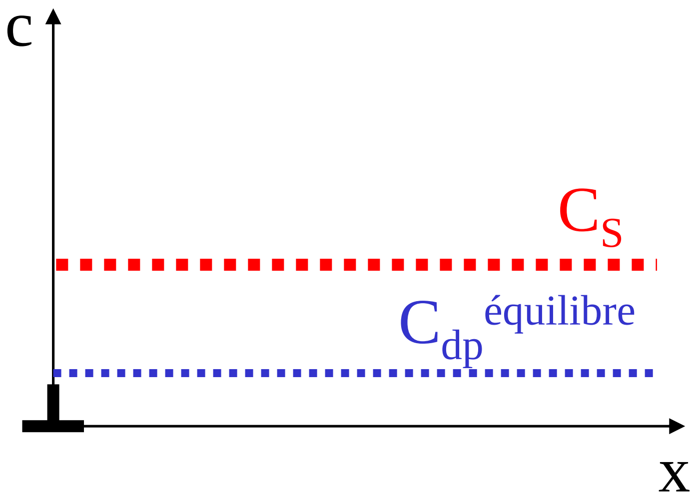
Enhanced diffusion of atoms

⇩

Enhanced precipitation

Point defect fluxes: a specificity of systems under irradiation

No irradiation



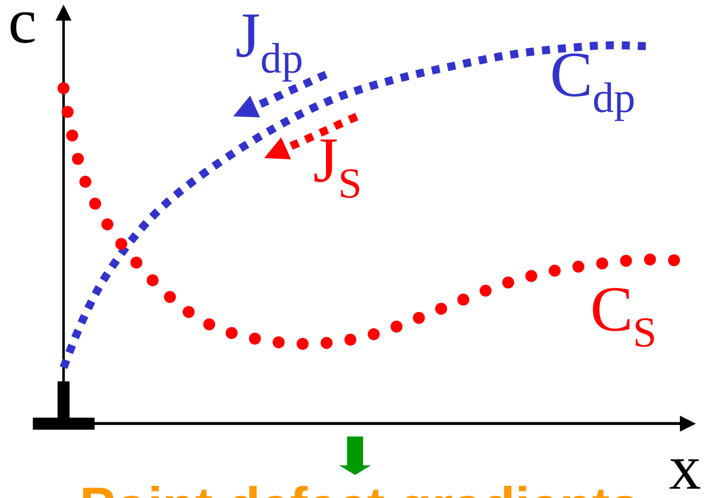
⇩

No point defect gradients

⇩

No point defect fluxes

Under irradiation



⬇

Point defect gradients

⬇

Point defect fluxes

⬇

Coupling between point defect and solute atom fluxes

⬇

Radiation induced segregation

Slow kinetics : example of segregation out of equilibrium induced by irradiation

Cr, Fe, Ni , Si concentration profiles induced by neutron irradiation at 420℃, in an ausrenitic steel

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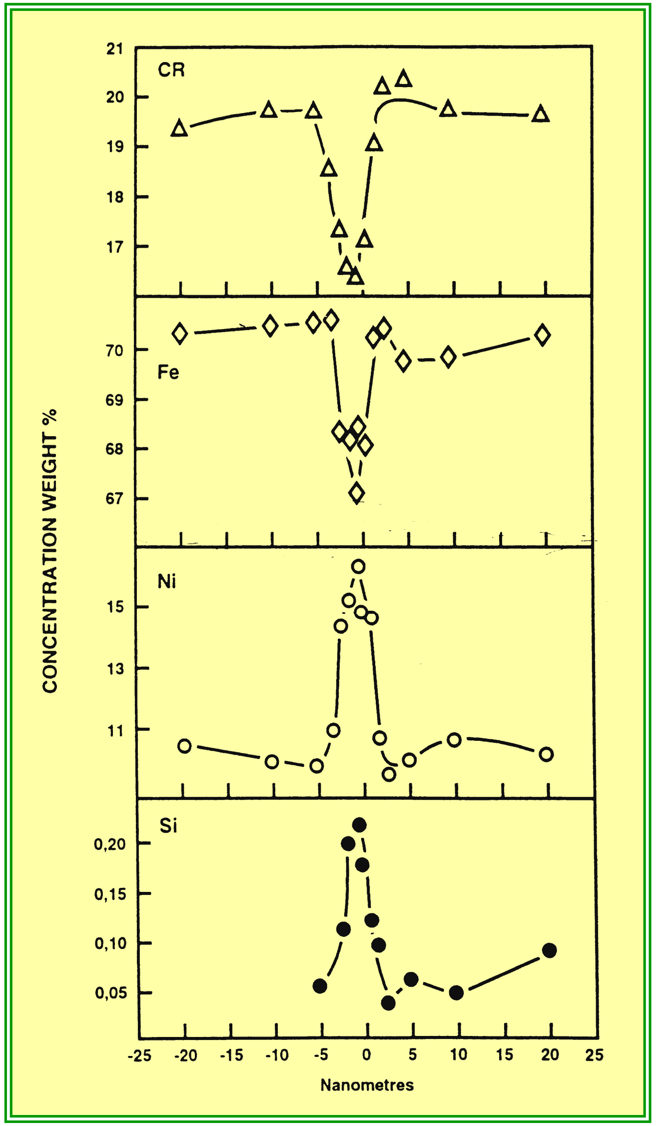
Role of the Cr depletion in the irradiation assisted stress corrosion crack in PWR internals.



Slow kinetics: from the radiation induced segregation to radiation induced precipitation

Radiation induced precipitation in the under saturated solution <u>Ni</u> 4%at Si , (5 10^{-9} dpa/s, 300℃, 8. 10^{-5}dpa, $[Si]_{limite}$ = 10%)

(a) Images of dislocation loops
(b) Precipitates associated to loops

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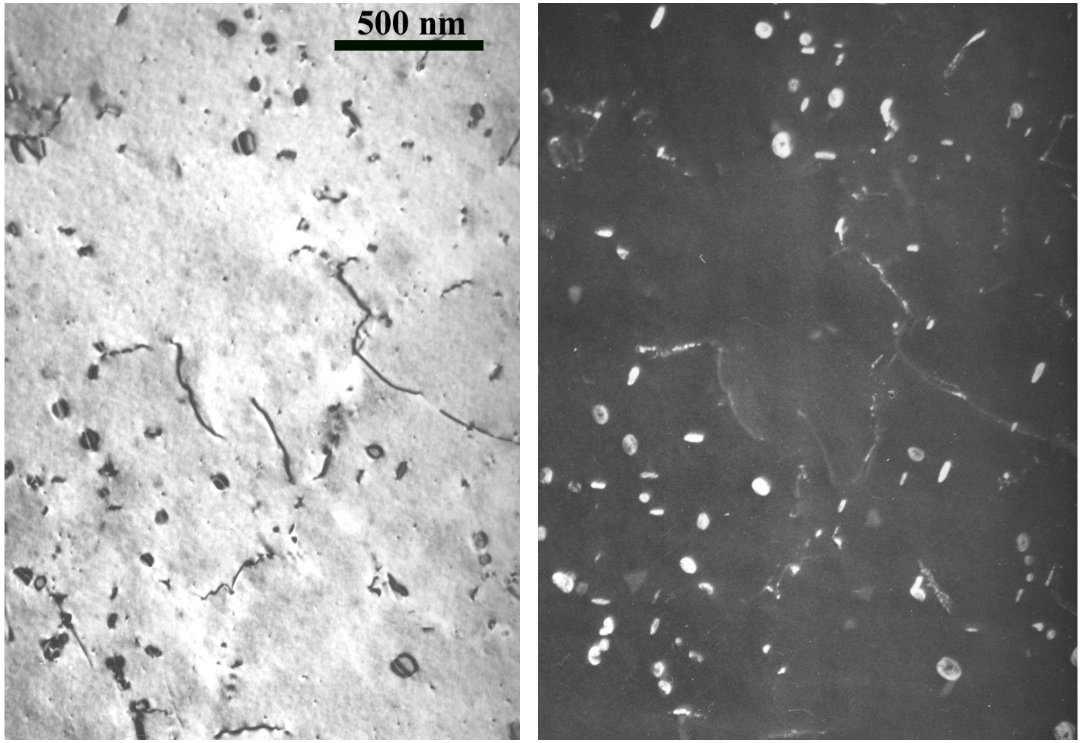
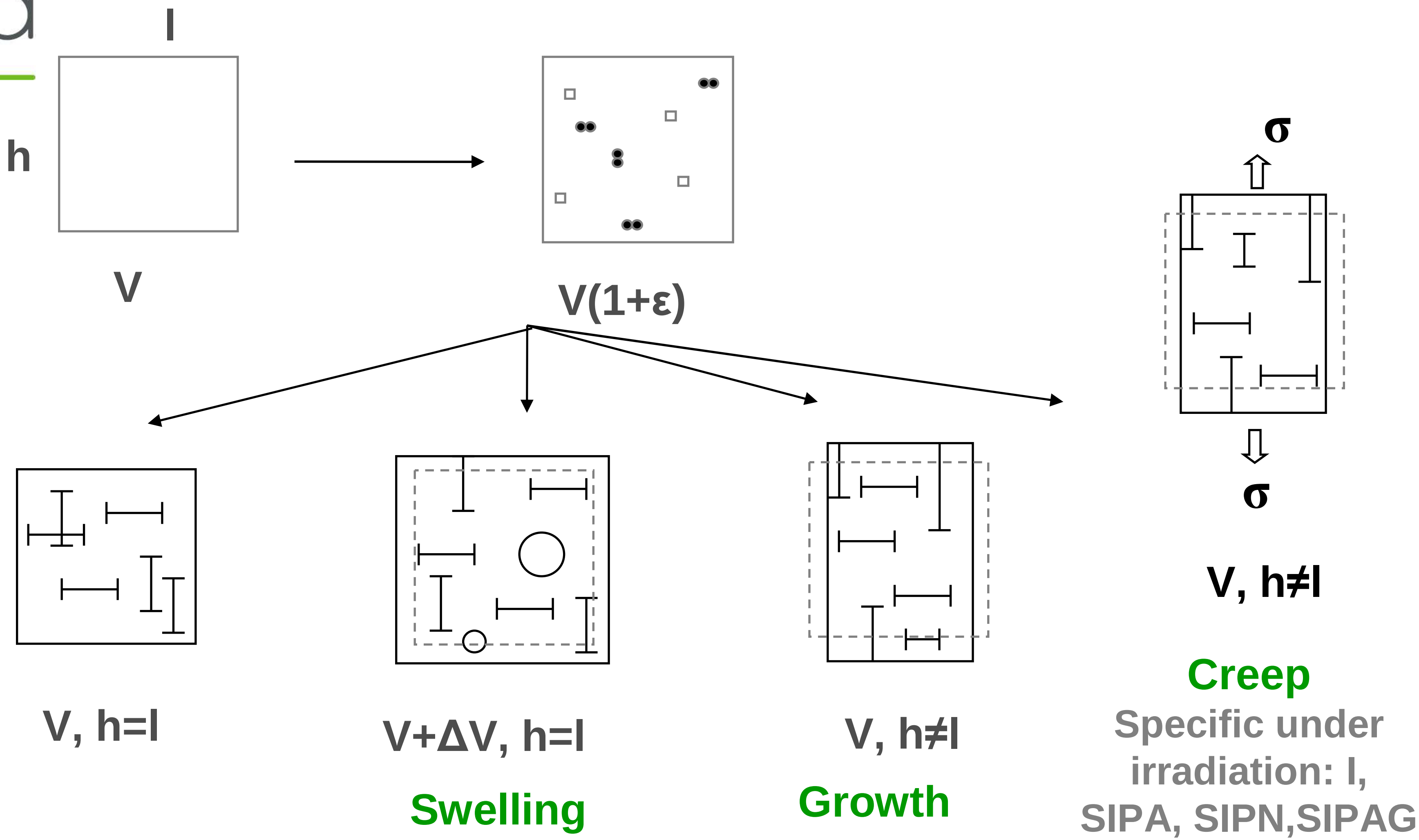
Phase diagrams obtained out of irradiation are no more valid under irradiation

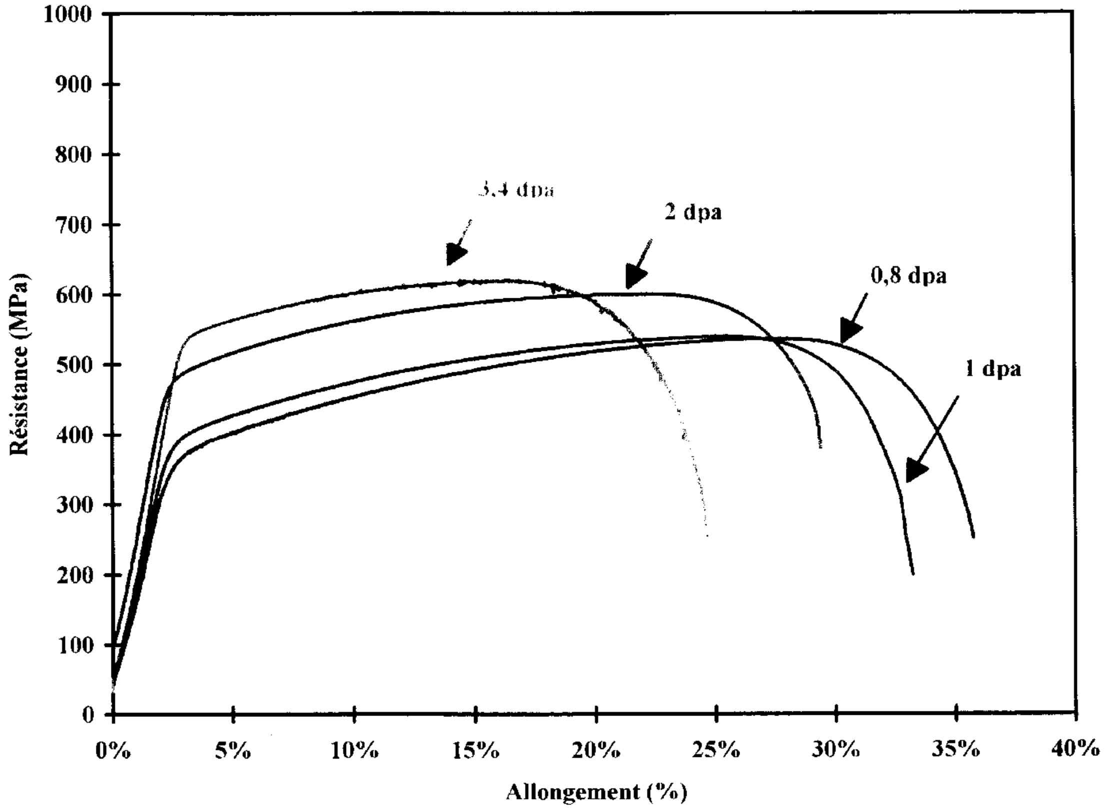


Macroscopic consequences: dimensional changes



Macroscopic consequences: hardening

- **Yield stress increase**
- **Decrease of the rupture elongation**

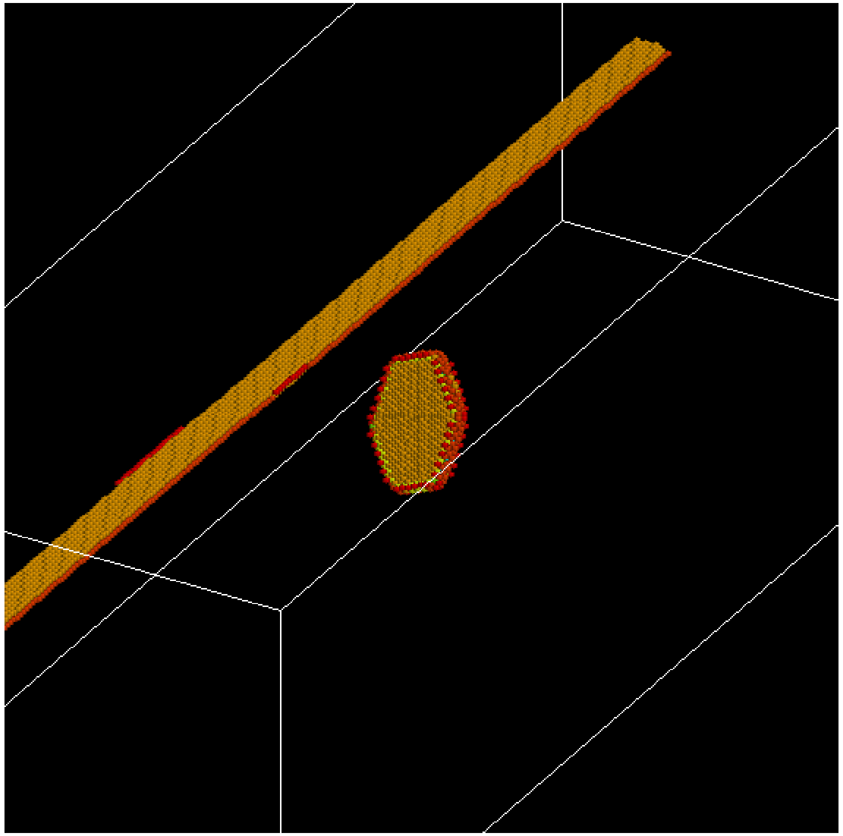


Traction curve evolution of a 316 austenitic steel neutron irradiated at 325°C, as a function of fluences.

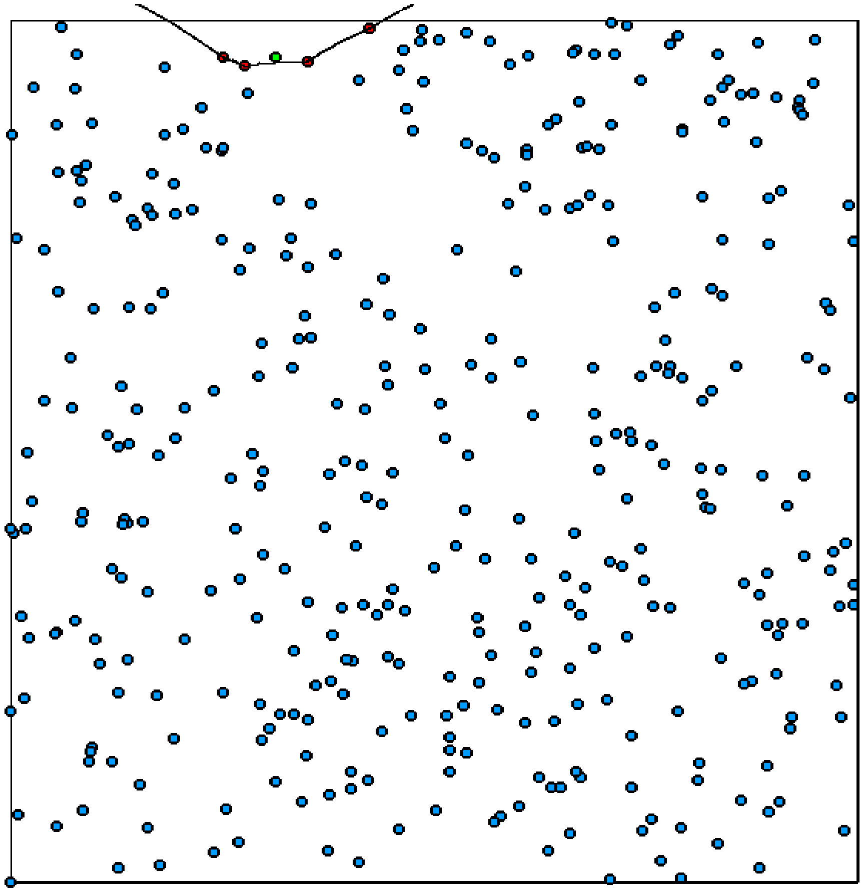
Macroscopic consequences: hardening

Pining of dislocations by radiation induced defects

Elementary mechanism



Collective effect

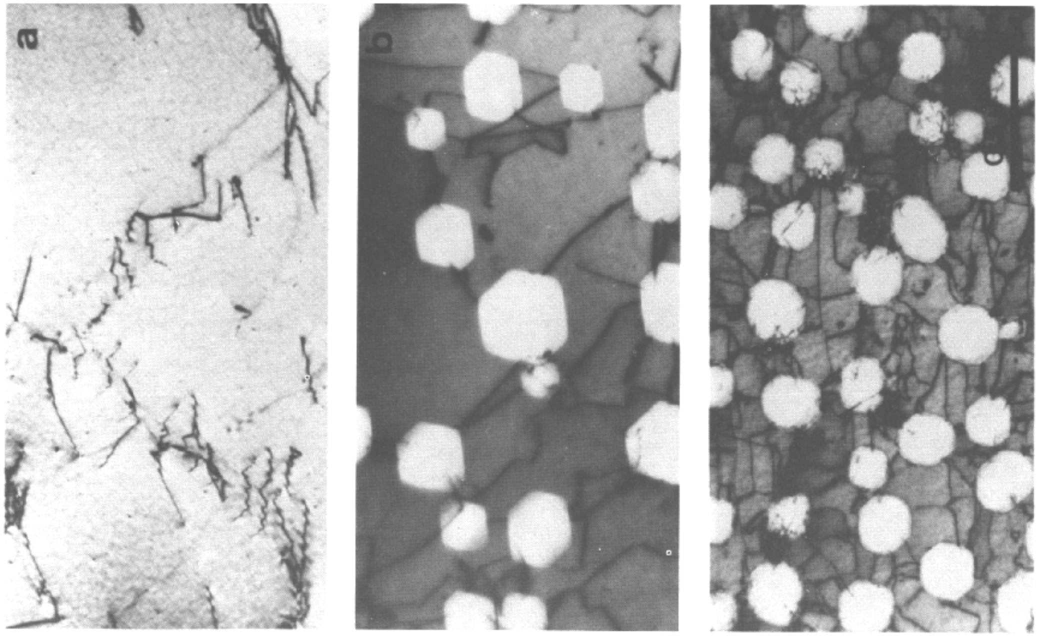


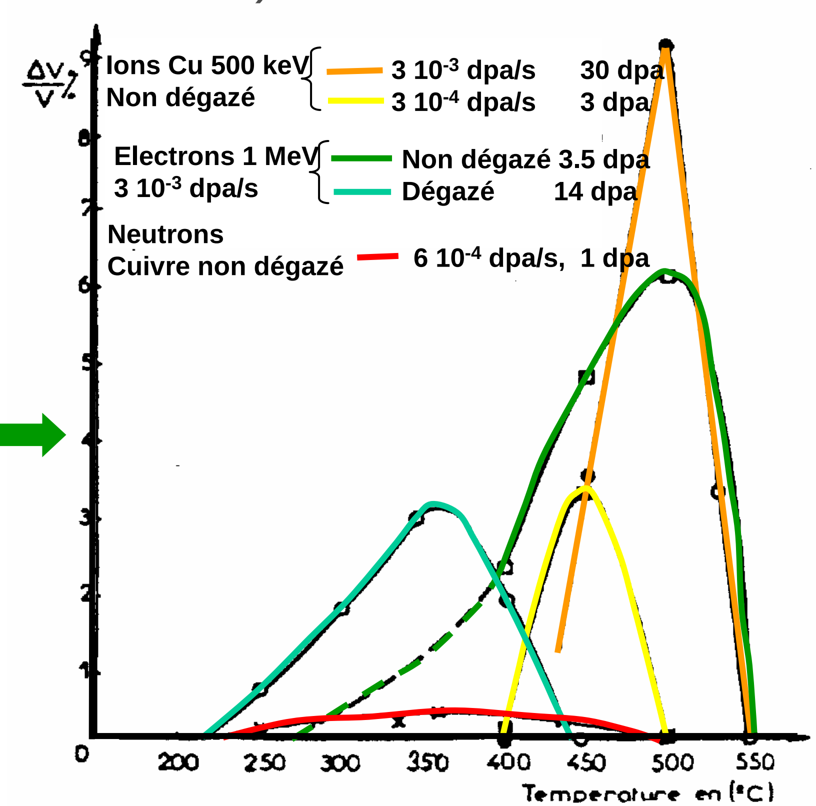
step 12800 - sigma 28 MPa - sigma max 140 MPa

Macroscopic consequences: void swelling

Occurs only when the vacancy flux toward voids is larger than SIA flux (consequence of the preferential attraction of SIA-dislocation interaction).

Cu irradiated at 500℃ with 500 keV Cu ions at 3 10-3dpa/s; Oxygen 30, 70 et 110 ppm (Glowinsky 1976)



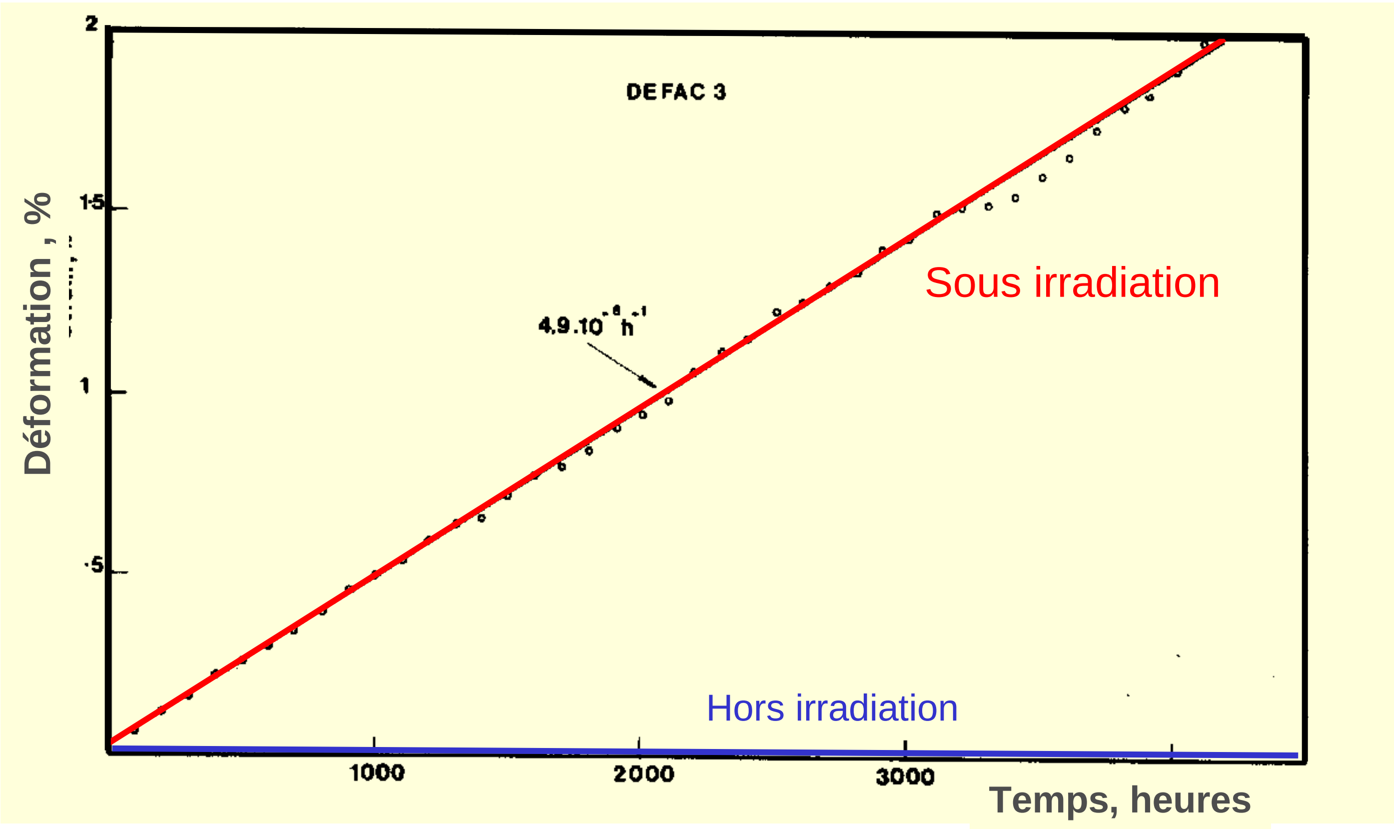


Peak swelling

In reactors, important role of helium produced in nuclear reactions in void nucleation

Macroscopic consequences: irradiation creep

Specific creep mechanism under irradiation efficient at temperature at which the thermal creep is null



316 stainless steel; Uni axial stress; neutron irradiation (Rapsodie); 450°C; 2.6 10 $^{-6}$ dpa/s;

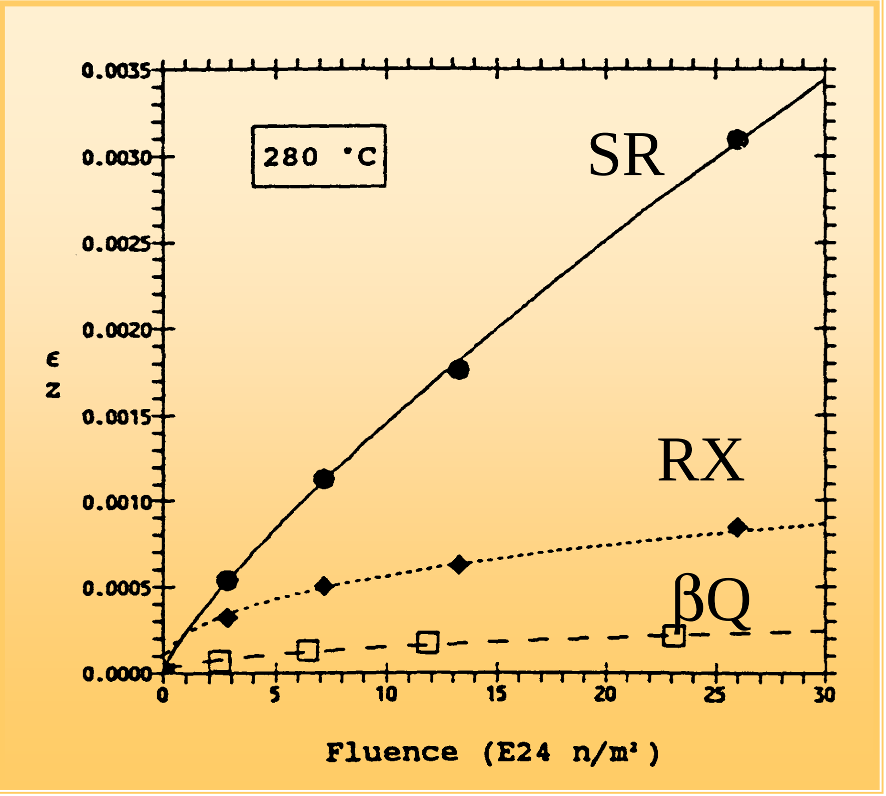
Macroscopic consequences: growth

- Shape change at constant volume
- Only in anisotropic materials : zirconium alloys, graphite, uranium, etc.

βQ : quenched β, isotropic

RX recristallized, few ⊥

SR stress released, many ⊥



Anisotropic nucleation of loops and anisotropic diffusion of point defects

Conclusion

- Irradiation changes material properties
- Associated to deep changes of the microstructure often at nanometric scale.
- Origin in the production of point defects created by irradiation and their slow evolution
- Necessity to be able to predict and extrapolate the materials behavior at large fluences = modeling validated with irradiation carried out with charged particles.
- Final test in reactor
- As the irradiation in experimental reactors are long, heavy and very expensive, multi-scale modeling based on physical phenomena allow to optimize the in reactor tests.