Introduction to Radiation Damage in Metals

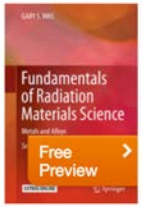
Journey from incoming particles

Most of the slides are borrowed from

22.14 Materials in Nuclear Engineering

22.74 Radiation Damage and Effects in Nuclear Materials

Was. means from Prof. Was' text book:



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Fundamentals of Radiation Materials Science

Metals and Alloys

Authors: **Was**, Gary S.

Radiation Damage

Radiation Damage

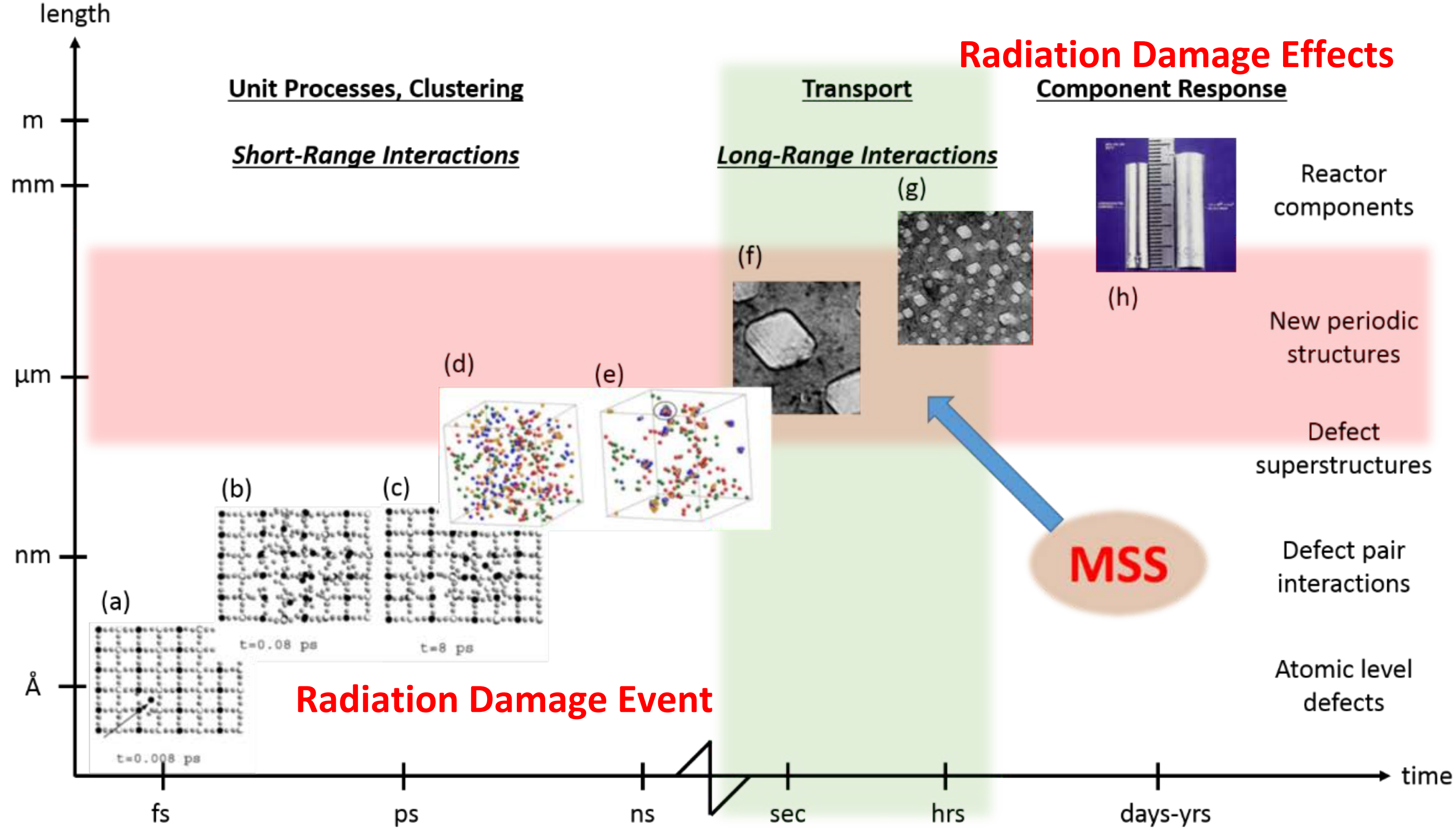
Radiation Damage Event (Monday)

the transfer of energy from an incident projectile to the solid and the resulting distribution of target atoms after completion of the event

Radiation Damage Effects (Wednesday)

Subsequent events involving the migration of the point defects and defect clusters and additional clustering or dissolution of the clusters

From Radiation Damage Event to Radiation Damage Effects



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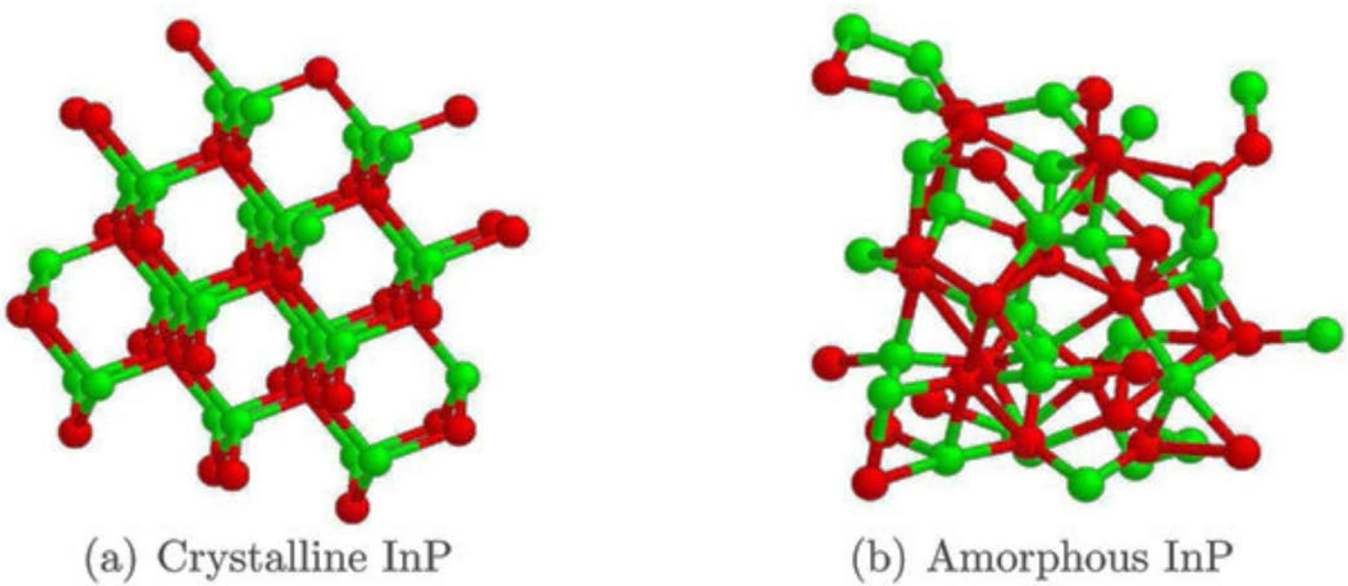
Short & Yip. ***Current Opinions in Solid State Material Science*** **(2015)**

Materials Intro.

- Crystalline vs. Amorphous: The difference is *long-range* order

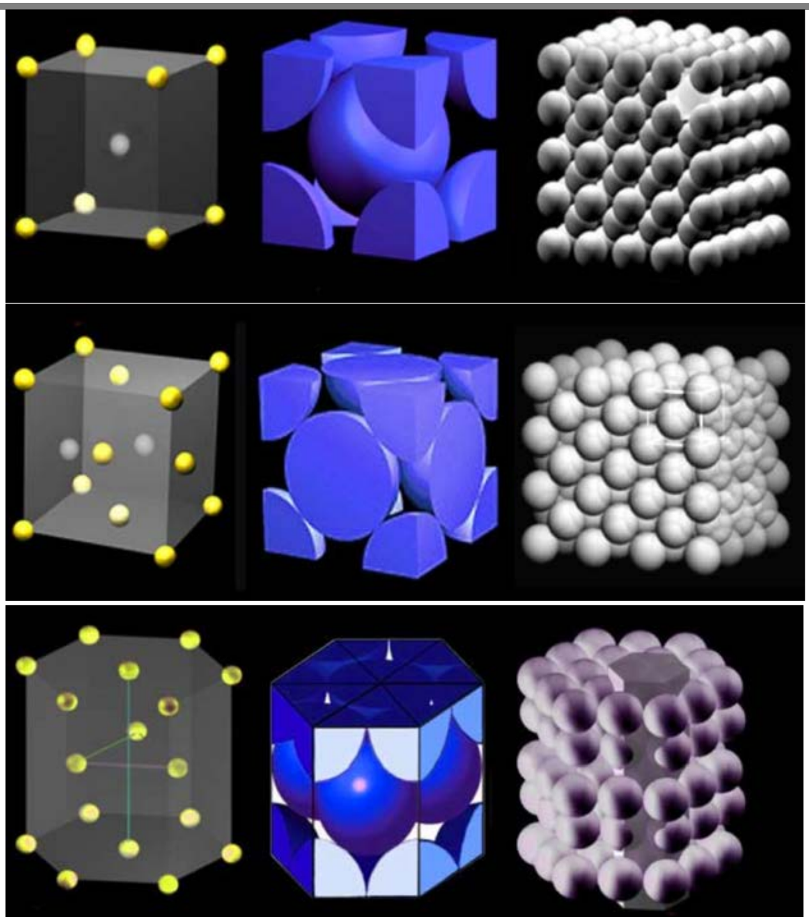


http://physics.anu.edu.au/eme/research/amorphous.php

Materials Intro.

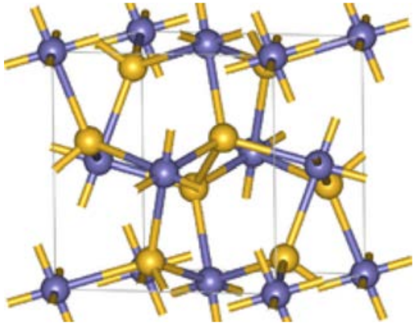
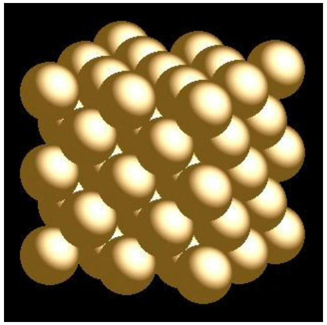
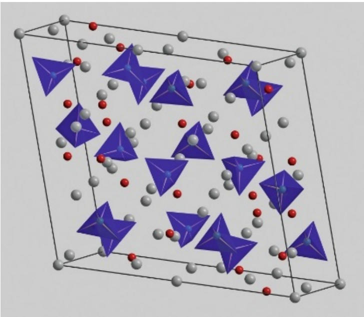
Crystalline materials lattices

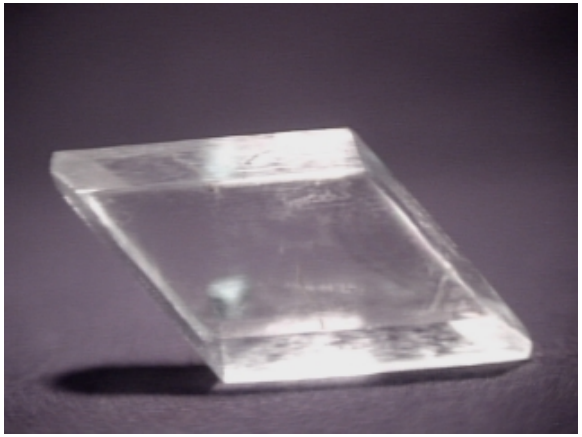
- **Body-Centered Cubic (BCC) Structure**
- **Face Centered Cubic (FCC) Structure**
- **Hexagonal Close Packed (HCP) Structure**



These images showing BCC structure, FCC structure, and HCP structure are reprinted/reused by permission from, © Iowa State University Center for Nondestructive Evaluation (CNDE).

Form Follows Structure

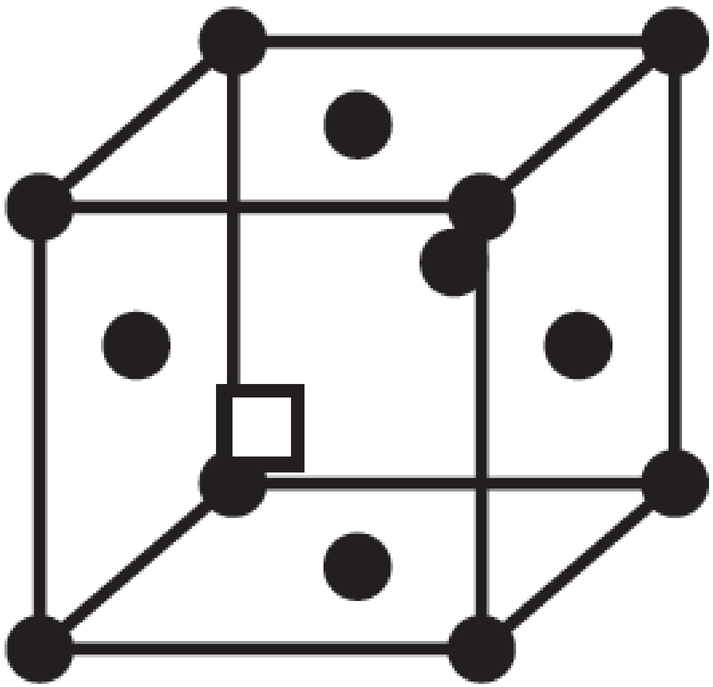
Pyrite (FeS_2), simple cubic (SC)

Gold (Au), face centered cubic (FCC)

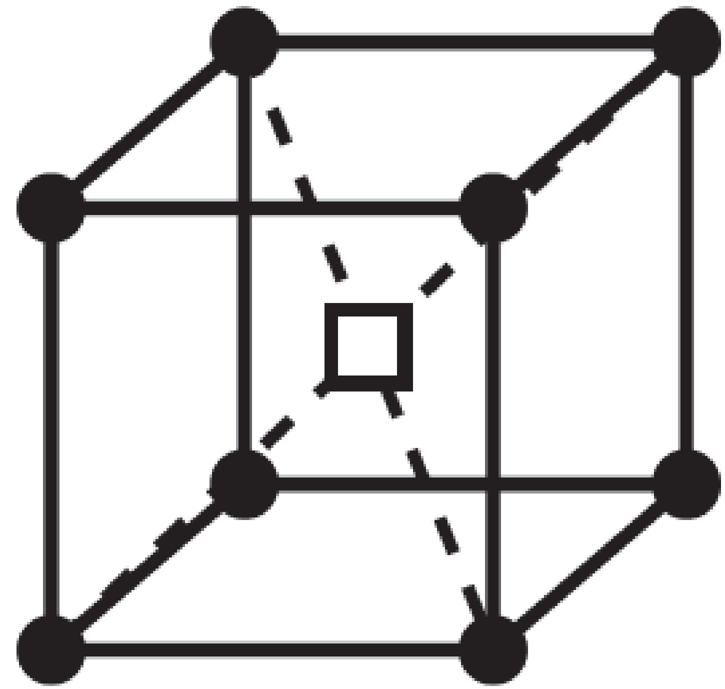
Gypsum, monoclinic

Point Defects (0D) – Vacancies

Was, p. 163



Vacancy in FCC lattice

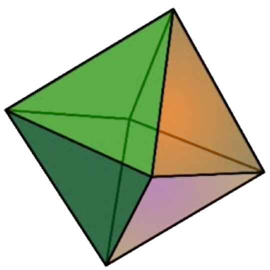


Vacancy in BCC lattice

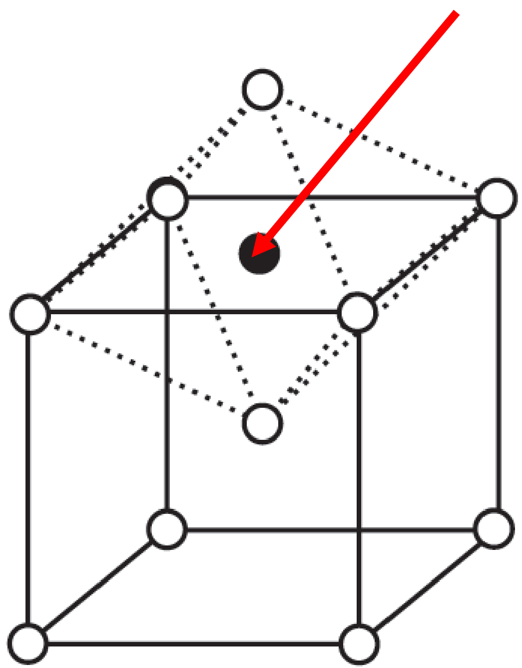
Point Defects (0D) – Interstitials

Was, p. 157

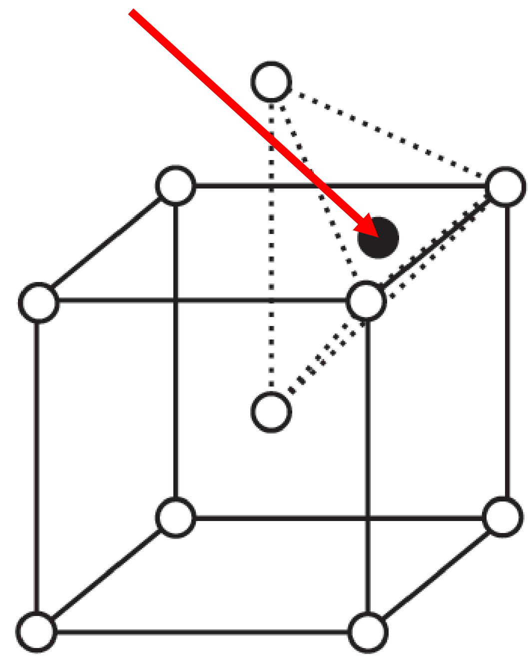
- Extra atoms shoved into the crystal lattice



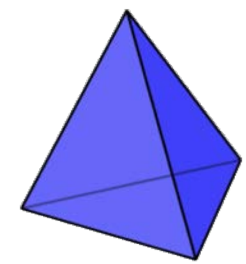
Octahedron



Octahedral interstitial in BCC lattice

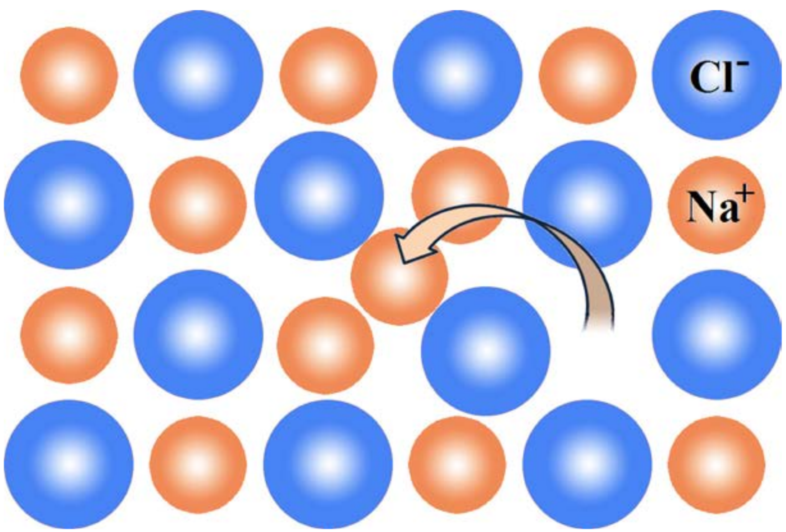


Tetrahedral interstitial in BCC lattice



Tetrahedron

Point Defects (0D) – Frenkel Pair



https://commons.wikimedia.org/w/index.php?curid=25745819

Frenkel Pair: an atom is displaced from its lattice position to an interstitial site, creating a vacancy at the original site and an interstitial defect at the new location

Dislocations (1D)

Was, p. 268

- Two types: Edge & Screw
- **Edge dislocation**: Extra half-plane of atoms shoved into the lattice

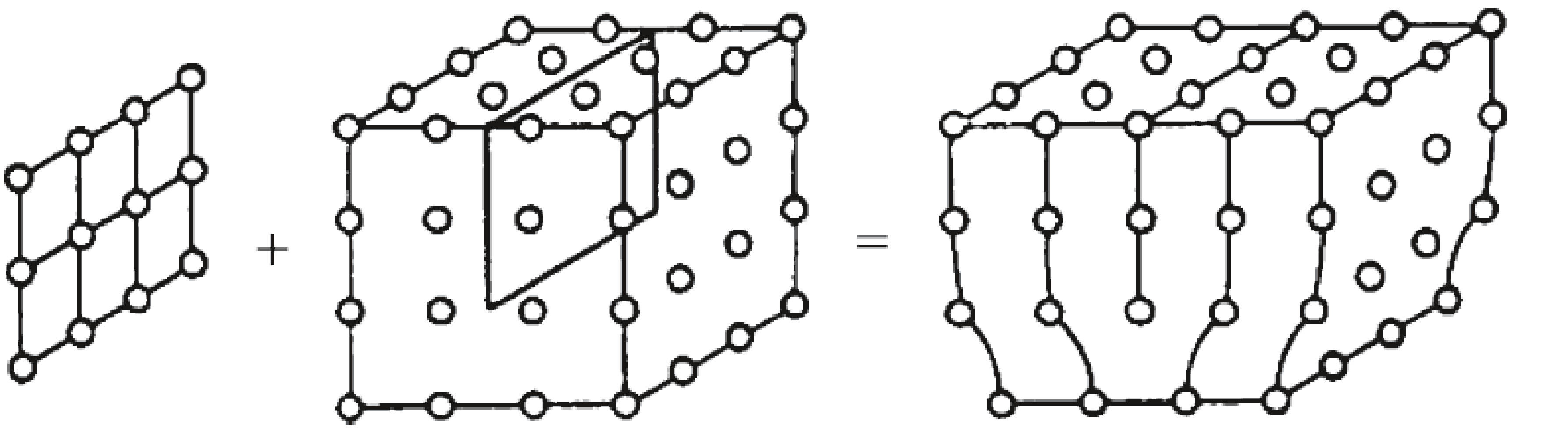


Fig. 7.2. An edge dislocation described as an extra half plane of atoms above the slip plane

Dislocations (1D)

Was, p. 268

- Two types: Edge & Screw
- **Screw dislocation**:

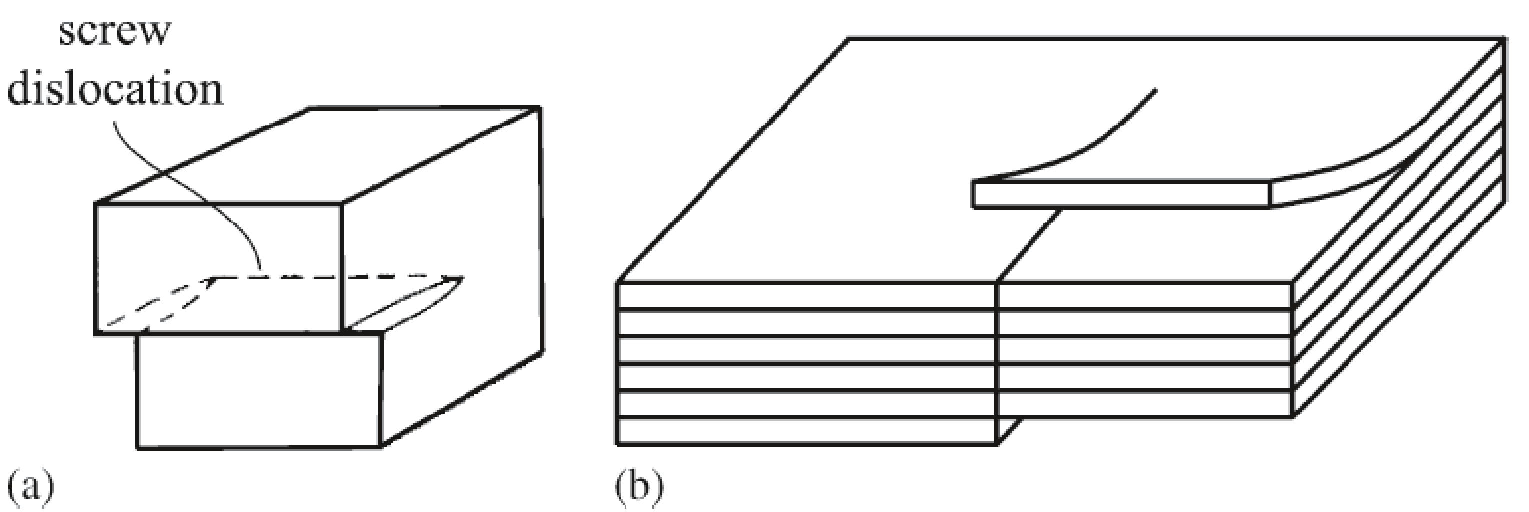
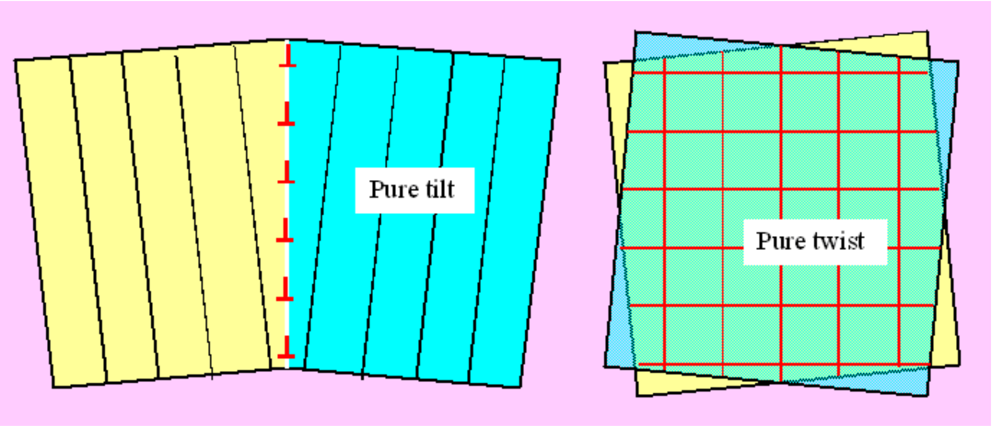


Fig. 7.3. (a) A screw dislocation formed by a cut and a shift of atoms in a direction parallel to the *cut line*. (b) A schematic showing the "parking ramp" nature of a screw dislocation in which atom planes spiral about the dislocation line

Grain Boundaries (2D)

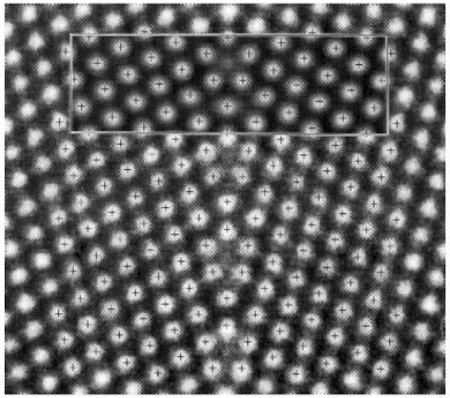
http://www-hrem.msm.cam.ac.uk/gallery/

- Regions of different orientation



http://www.tf.uni-kiel.de/matwis/amat/def_en/kap_7/backbone/r7_2_1.html

Tilt grain boundary in Al

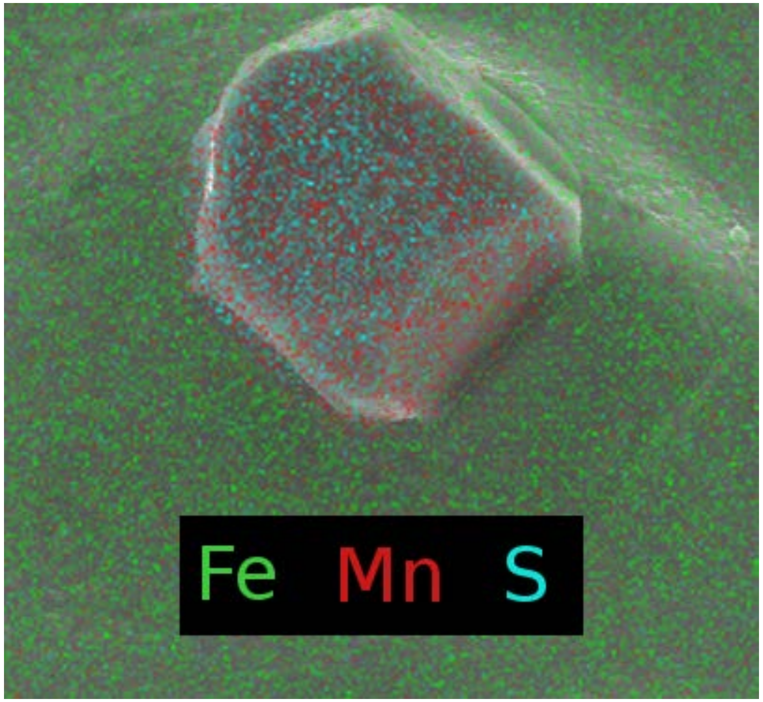


Courtesy of Sandia National Lab.

Inclusions (3D)

From Prof. Short's own image collection

- Other phases trapped within base material
- Examples:
  - Secondary particle precipitates in Zircaloys
  - Carbides in steels
  - Y_2O_3 particles in Oxide Dispersion Strengthened (ODS) steels



Single crystal of MnS, space group $Fm\bar{3}m$, FCC crystal structure embedded in Alcator rotor steel

Summary of Material Intro.

- Crystalline Solids
- 0D Defects
  - Vacancies & Interstitials
- 1D Defects (Dislocations)
- 2D & 3D Defects

We are interested in how many defects (Frenkel pairs & defect clusters) are produced by incoming particles

Radiation Damage Event

The result of a radiation damage event is the creation of a collection of point defects (vacancies and interstitials) and clusters of these defects in the crystal lattice. (10^{-11} s)

This can be described by how many displacements of atoms (DPAs) are created by incoming particles

Displacement Theory

- Define a rate of atomic displacements using flux:

Maximum energy available

Energy dependent flux distribution

$$R = \int_0^{E_{max}} N * \Phi(E_i) * \sigma_D(E_i) dE_i$$

Reaction rate $\frac{displ.}{m^3 - sec}$

Material number density $\frac{atoms}{m^3}$

Displacement cross section

Displacement Theory

- Define a rate of atomic displacements using flux:

$$R = \int_0^{E_{max}} N * \Phi(E_i) * \sigma_D(E_i) dE_i$$

$$\downarrow$$

$$\frac{R}{N} = \frac{DPA}{sec} = \int_0^{E_{max}} \Phi(E_i) * \sigma_D(E_i) dE_i$$

Displacement Theory

- Define a rate of atomic displacements using flux:

$$\frac{DPA}{sec} = \int_0^{E_{max}} \Phi(E_i) * \sigma_D(E_i) dE_i$$

Probability that an atom displaced by a particle with energy E_i leaves with recoil energy T (differential energy transfer cross section)

$$\sigma_D(E_i) = \int_{T_{min}}^{T_{max}} \sigma(E_i, T) v(T) dT$$

Number of atomic displacements from a PKA with energy T

- T is the PKA (displaced atom) recoil energy

Displacement Theory

- Define a rate of atomic displacements using flux:

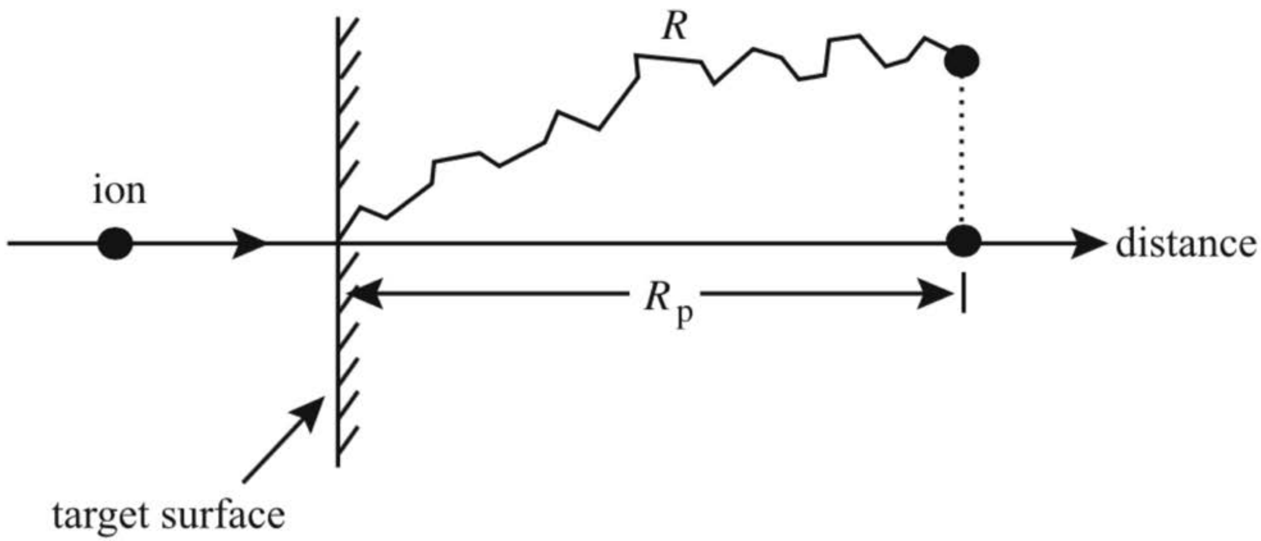
Probability that an atom displaced by a particle with energy E_i leaves with recoil energy T (differential energy transfer cross section)

$$\frac{DPA}{sec} = \int_0^{E_{max}} \int_{T_{min}}^{T_{max}} \Phi(E_i) * \sigma(E_i, T)\nu(T)dTdE_i$$

Number of atomic displacements from a PKA with energy T

Journey of an incoming ion

(E_i)



Total path length R and projected range R_p for an ion incident on a target

What happened until it stops

Explanation on board

$(\sigma_n , \sigma_e , \mathrm{T_n}, \mathrm{T_e})$

Journey of multiple incoming ions

(f_{E_i}) → Continuous description of a large quantity discrete events

↓

Stopping power

Coulombic/Nuclear Stopping Power

- *Stopping Power* is defined as differential energy loss as a function of energy:

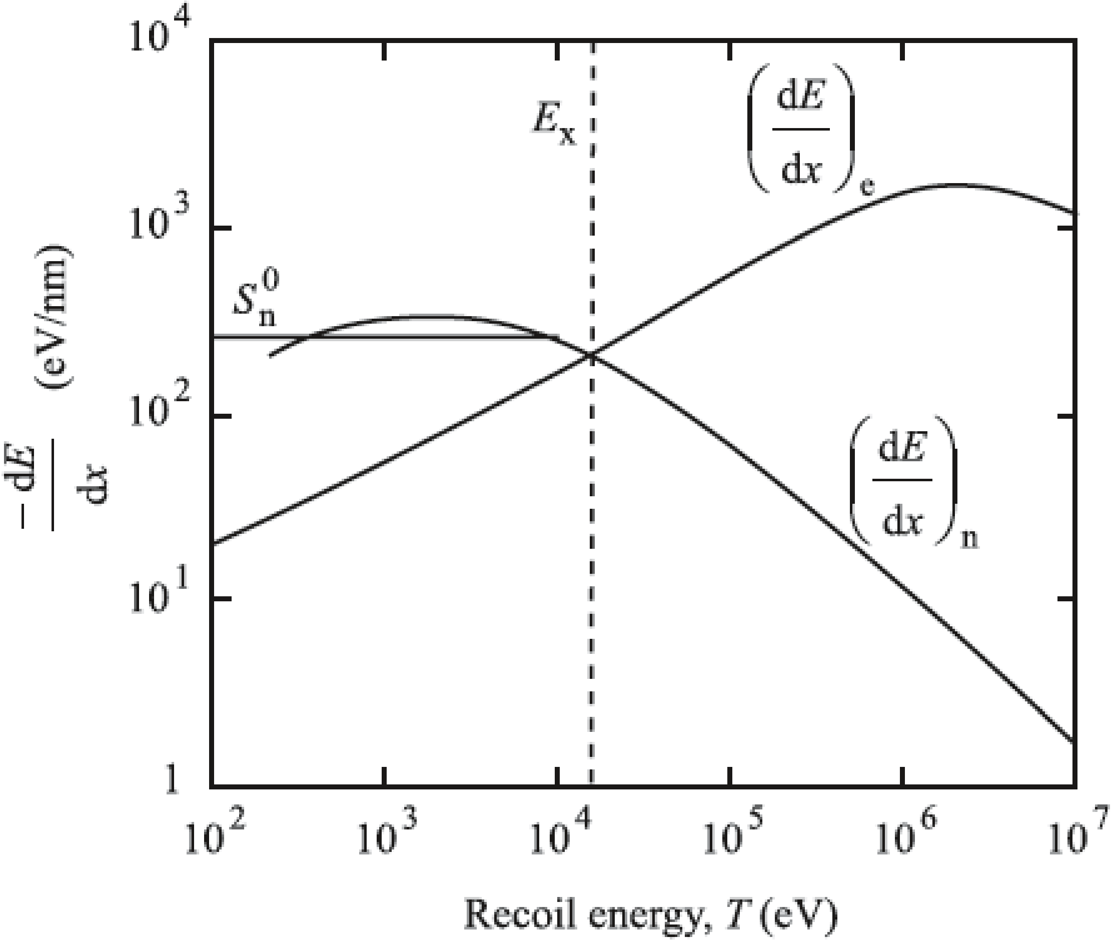
$$N * S(E) = -\frac{\partial E}{\partial x}$$

- Separable components due to nuclear (screened nucleus Coulombic), electronic, and radiative terms:

$$N * S(E) = -\left(\frac{\partial E}{\partial x}\right)_{nucl.} - \left(\frac{\partial E}{\partial x}\right)_{elec.} - \left(\frac{\partial E}{\partial x}\right)_{rad.}$$

Relative Stopping Powers

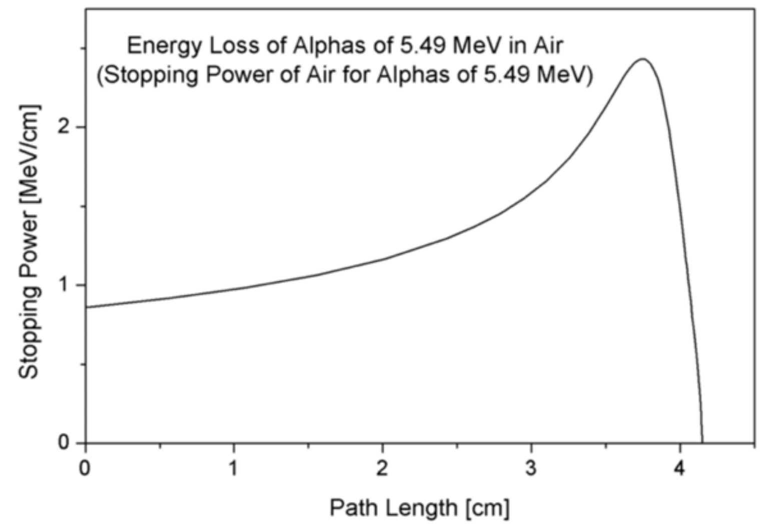
Was, p. 84



Range

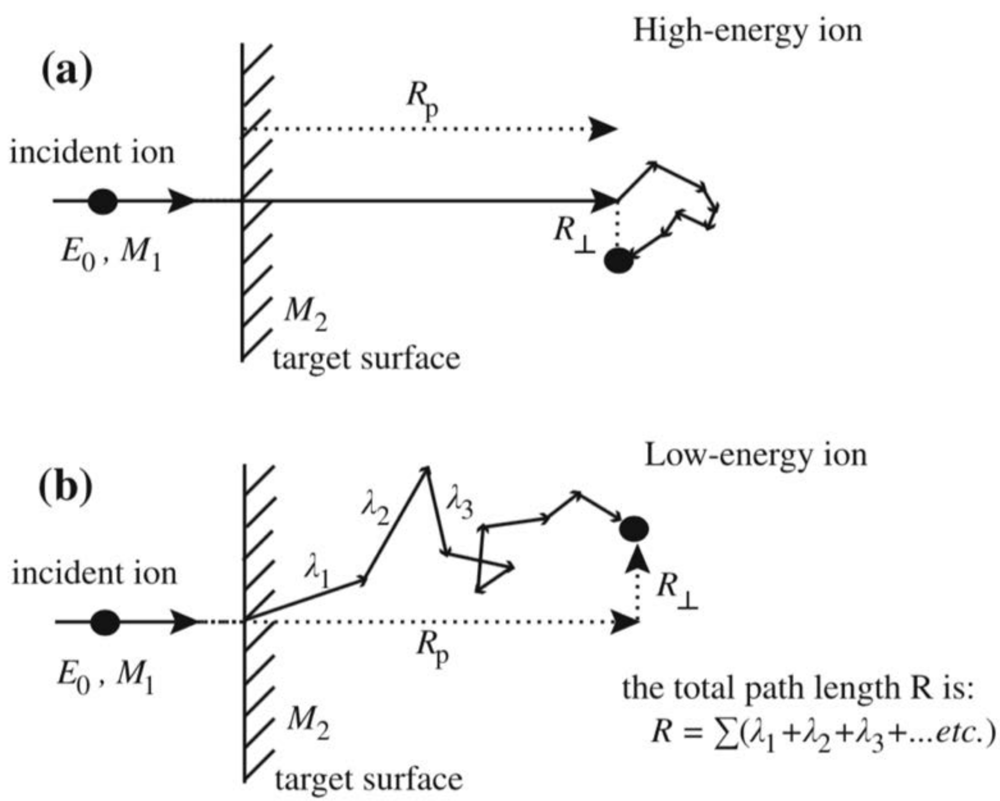
Source: Wikimedia Commons

- Integrate inverse of stopping power over the energy range of the particle: $Range = \int_0^{E_{max}} \frac{1}{S(E)} dE$
- Not all particles have identical range, *straggling* describes this variation



Range of the incoming ions

Was, p. 66



journey continues by PKAs

PKAs → How many atoms will be displaced by a PKA of energy T, $\nu(T)$

↓

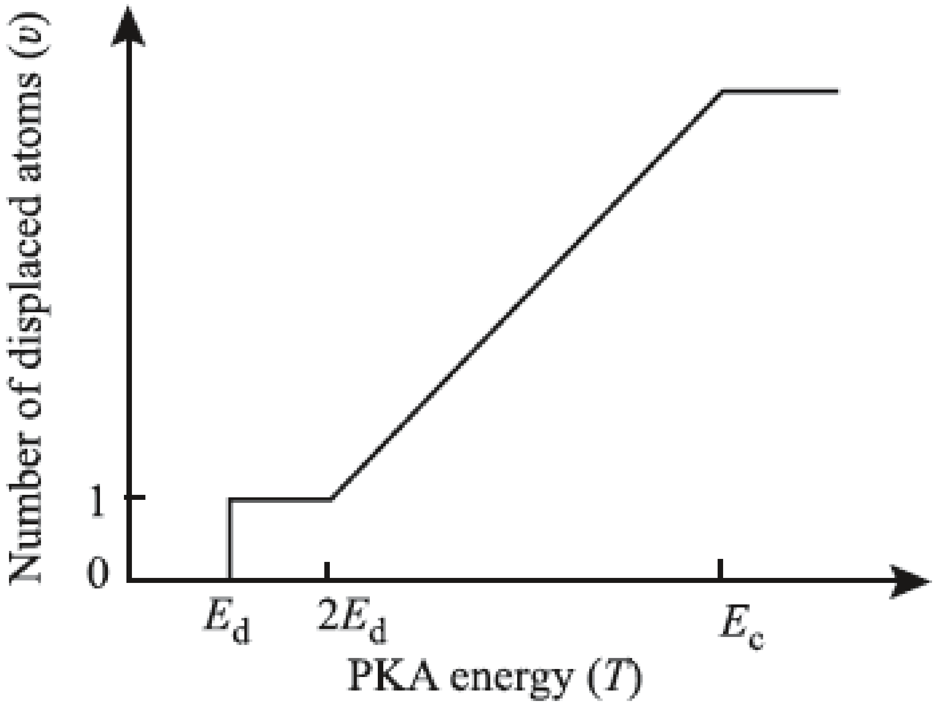
Kinchin and Pease Model (K-P Model)

Kinchin-Pease Model

Was, p. 77

- Final formulation:

$$\nu(T) = \begin{cases} 0 & \text{for } T < E_d \\ 1 & \text{for } E_d < T < 2E_d \\ \dfrac{T}{2E_d} & \text{for } 2E_d < T < E_c \\ \dfrac{E_c}{2E_d} & \text{for } T \geq E_c \end{cases}$$



Assumptions made by K-P model

1. The cascade is created by a sequence of two-body elastic collisions between atoms.
2. The displacement probability is 1 for $T \geq E_d$
3. No energy passes to the lattice during the collision process
4. For all energies less than cutoff energy E_c , electronic stopping is ignored
5. The energy transfer cross section is given by the hard sphere model
6. Effects due to crystal structure are neglected

K-P Mods – Modifications by release the assumptions

Was, p. 109

Table 2.4. Modifications to the displacement cross section

Assumption	Correction to $\nu(T)$	Equation in text
3: Loss of E_d	$0.56\left(1+\frac{T}{2E_d}\right)$	Eq. (2.31)
4: Electronic energy loss cut-off	$\xi(T)\left(\frac{T}{2E_d}\right)$	Eq. (2.50)
5: Realistic energy transfer cross section	$C\frac{T}{2E_d}, \quad 0.52 < C \leq 1.22$	Eqs. (2.33), (2.39)
6: Crystallinity	$\frac{1-P}{1-2P}\left(\frac{T}{2E_d}\right)^{(1-2P)} - \frac{P}{1-2P}$	Eq. (2.104)
	$\sim\left(\frac{T}{2E_d}\right)^{(1-2P)}$	Eq. (2.105)

The Real σ_D Is Ugly!

Was, p. 108

$$\sigma_D = \frac{\sigma_s(E_i)}{\gamma E_i} \int_{E_d}^{\gamma E_i} \left[\frac{1-P}{1-2P} \left(C'\xi(T) \frac{T}{2E_d} \right)^{(1-2P)} - \frac{P}{1-2P} \right]$$
$$\times \left[1 + a_1(E_i) \left(1 - \frac{2T}{\gamma E_i} \right) \right] dT$$
$$+ \sum_j \frac{\sigma_{sj}(E_i, Q_j)}{\gamma E_i} \left[1 + \frac{Q_j}{E_i} \left(\frac{1+A}{A} \right) \right]^{-1/2}$$
$$\times \int_{\check{T}_j}^{\hat{T}_j} \left[\frac{1-P}{1-2P} \left(C'\xi(T) \frac{T}{2E_d} \right)^{(1-2P)} - \frac{P}{1-2P} \right] dT$$
$$+ \int_0^{E_i - E'_m} \sigma_{(n,2n)}(E_i, T) \left[\frac{1-P}{1-2P} \left(C'\xi(T) \frac{T}{2E_d} \right)^{(1-2P)} - \frac{P}{1-2P} \right] dT$$
$$+ \sigma_\gamma \left[\frac{1-P}{1-2P} \left(C'\xi(T) \frac{E_\gamma^2}{8E_d(A+1)c^2} \right)^{1-2P} - \frac{P}{1-2P} \right] . \qquad (2.117)$$

Example σ_D

Was, p. 109

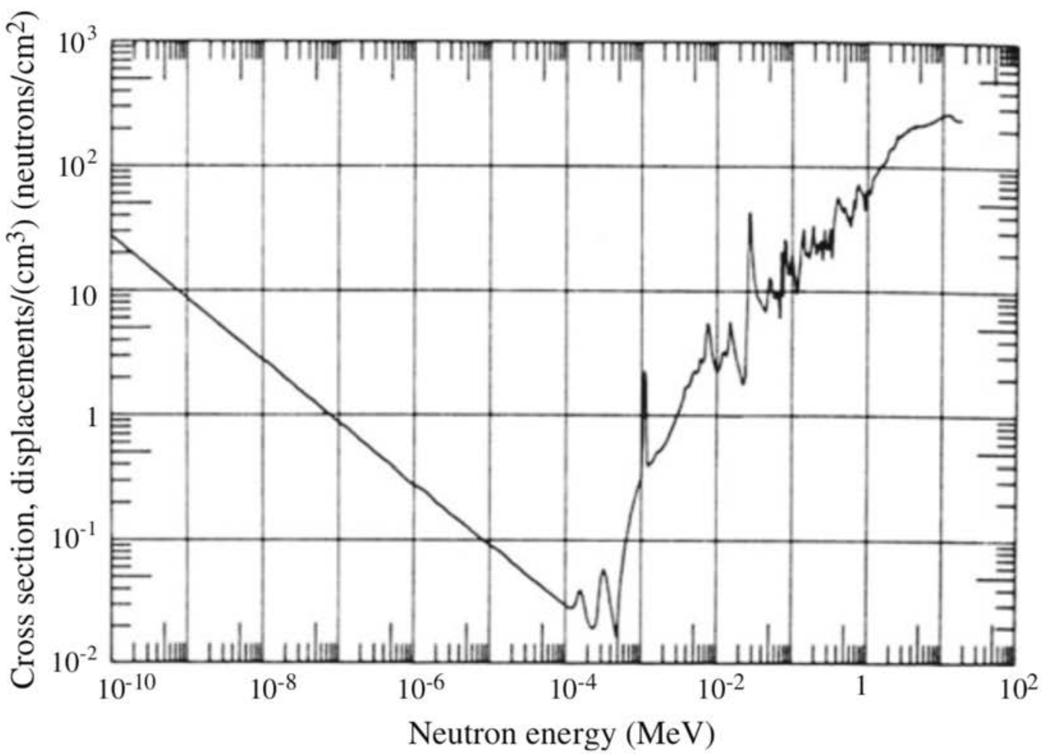


Fig. 2.18 The displacement cross section for stainless steel based on a Lindhard model and ENDF/B scattering cross sections (after [21])

Displacement Theory

- Define a rate of atomic displacements using flux:

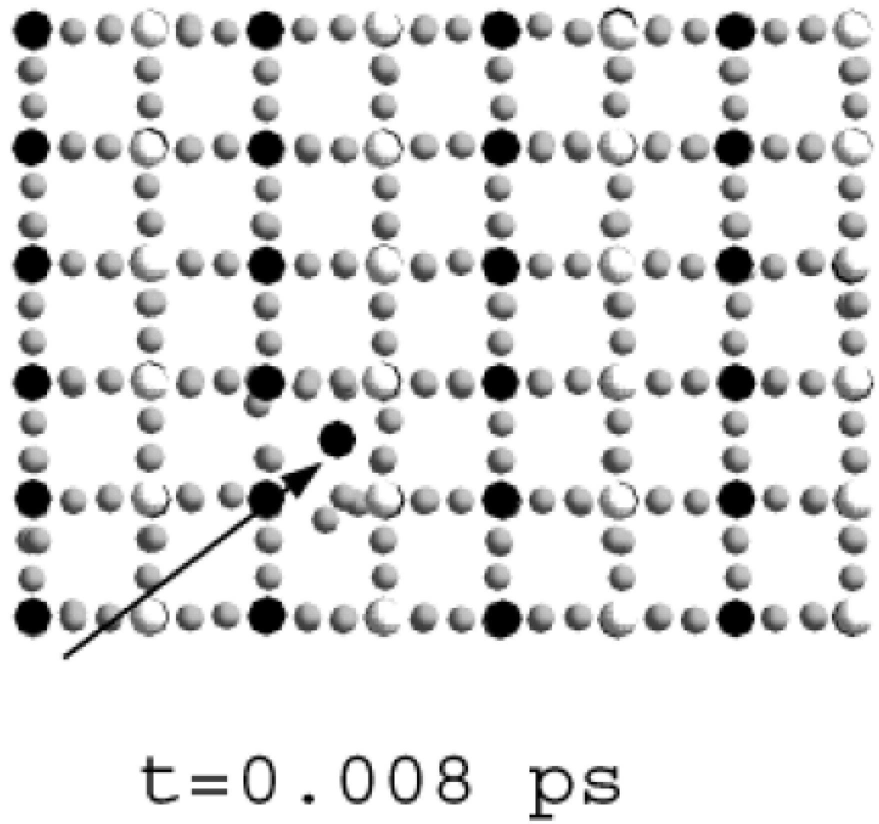
$$\frac{DPA}{sec} =$$

Does the number of displacements per atom per second we calculated this way equal with the stable defects (Frenkel pairs & defect clusters) introduced by incoming particles?

displacements
from a PKA with
energy T

Cascade Stages – Ballistics

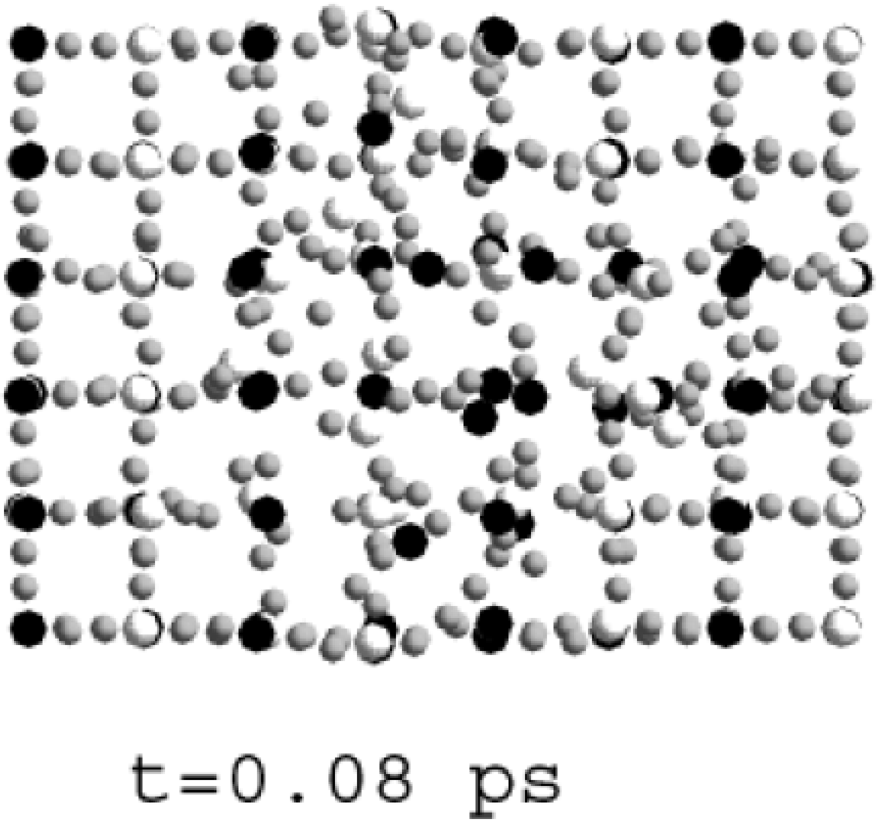
- PKA initiates a cascade of displacive collisions
- Ends until no atom contains enough energy to create further displacements



K. O. Trachenko, M. T. Dove. E. K. H. Salje. ***J. Phys. Condens. Matter*****, 13:1947 (2001)**

Cascade Stages – Thermal Spike

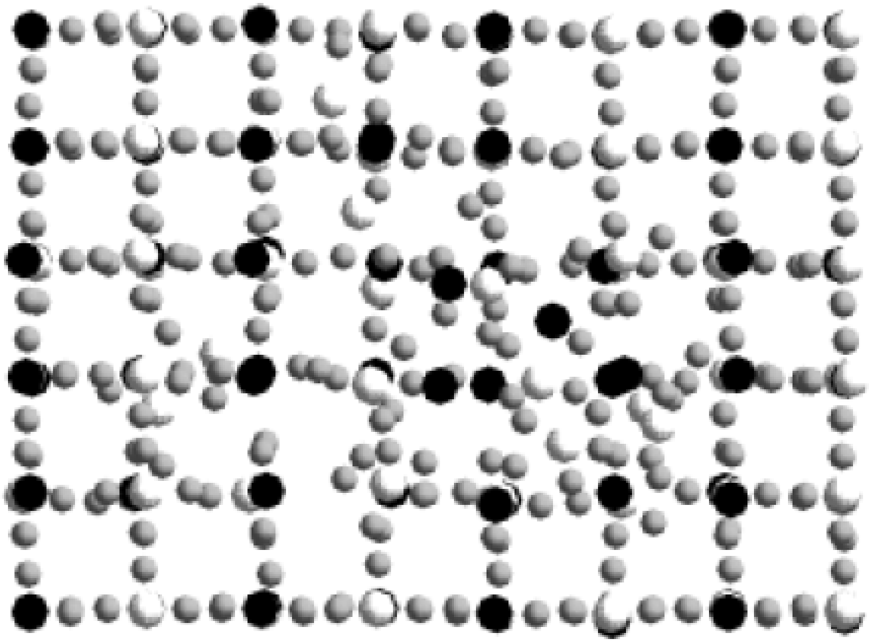
- Collisional energy of the displaced atoms is shared among their neighboring atoms
- Temperature rises very locally for a very short time



K. O. Trachenko, M. T. Dove. E. K. H. Salje. *J. Phys. Condens. Matter*, 13:1947 (2001)

Cascade Stages – Quench

- Heat is conducted away EXTREMELY quickly
- Stable lattice defects form either as point defects or defect clusters



t=8 ps

K. O. Trachenko, M. T. Dove. E. K. H. Salje. *J. Phys. Cond. Matter*, 13:1947 (2001)

Radiation Damage Event

- Radiation damage event completes after the quench stage
- We define the *displacement efficiency* ξ, as the fraction of the "ballistically" produced displacements that survive the cascade quench
- $\xi * \frac{DPA}{sec} = \xi * \int_0^{E_{max}} \int_{T_{min}}^{T_{max}} \Phi(E_i) * \sigma(E_i, T) \nu(T) dT dE_i$

Simulation Methods – MD

http://www-personal.umich.edu/~gsw/movies.html

3.1 Molecular Dynamics (MD) simulation of the development of a cascade from a 200 keV recoil in iron at 100K.

Note the extension of the cascade to the lower right hand corner of the screen. This lobe likely results from channeling of a knock-on atom. Also note the striking difference in defect density between the peak of the ballistic regime (~0.7 ps) and that at the end of the quench (~21.6 ps). (courtesy, R. Stoller, Oak Ridge National Laboratory)

Types of Radiation

Was, p. 138

- Different radiation produces different cascades

Mass & Charge

Increasing mass, same charge

Moderate mass, no charge

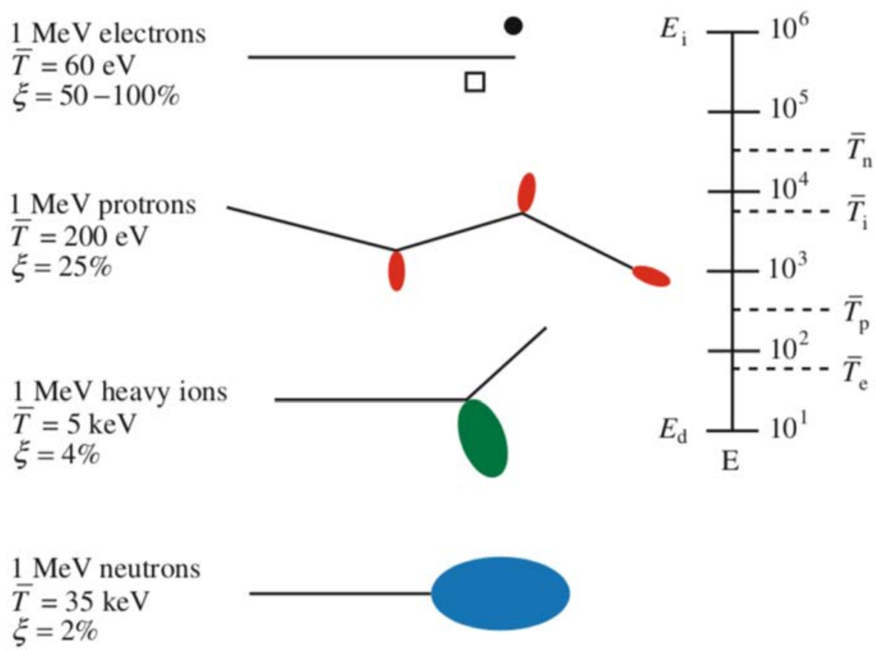


Fig. 3.7 Difference in damage morphology, displacement efficiency, and average recoil energy for 1 MeV particles of different types incident on nickel (after [6])

Stopping Mechanism

Almost All electronic

Mostly electronic

Nuclear and electronic

Entirely nuclear

Radiation damage effects

Radiation Damage Effects

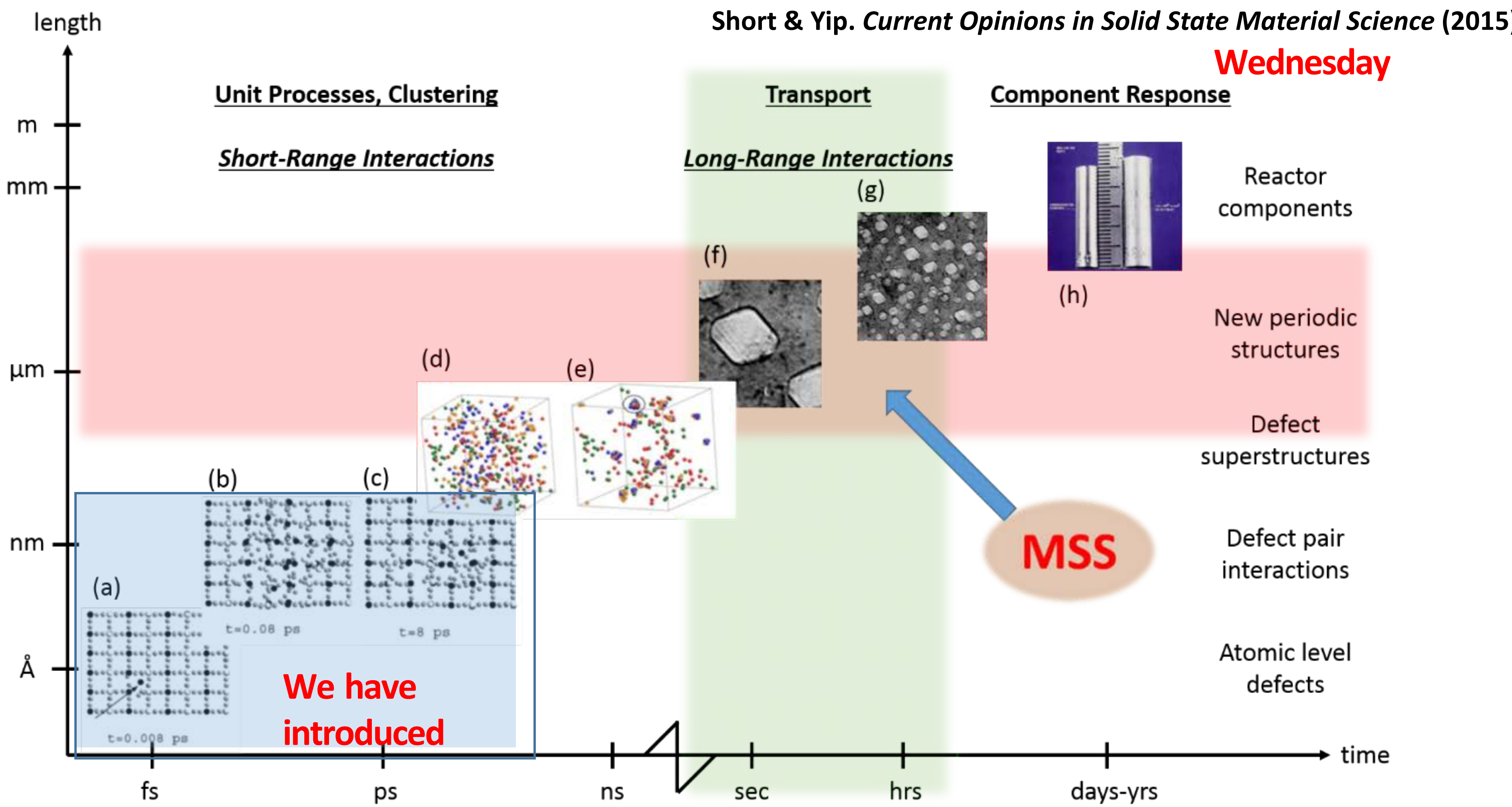
Physical effects

- RIS (Radiation-induced segregation)
- Nucleation and growth of dislocation loops and voids
- Phase Stability

Mechanical effects: when stress is applied

- Irradiation hardening and deformation
- Fracture and Embrittlement
- Irradiation Creep and Growth
- IRSCC (irradiation-assisted stress corrosion cracking)

Building Up to Radiation Effects



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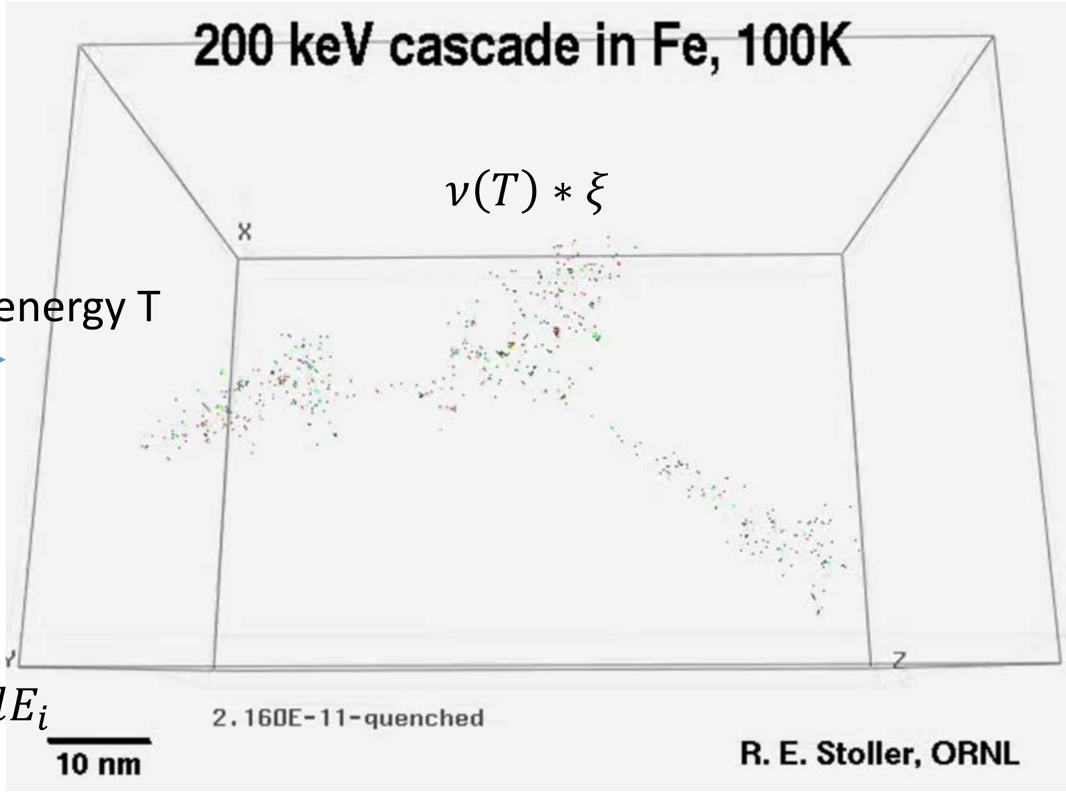
Radiation Damage Event: from incoming particles to point defects and defect clusters

http://www-personal.umich.edu/~gsw/movies.html

Incoming particle → $\sigma(E_i, T)$ → PKA with energy T

$\Phi(E_i)$



$\nu(T) * \xi$

$$\xi * \int_0^{E_{max}} \int_{T_{min}}^{T_{max}} \Phi(E_i) * \sigma(E_i, T) \nu(T) dT dE_i$$

Courtesy of Oak Ridge National Laboratory, U.S. Dept. of Energy.

Damage After the Cascade

- What happens to damage after the cascade?
  - Production
  - Recombination (One interstitial finds one vacancy and they annihilate)
  - Absorption at sinks (migrate to the sink and get trapped)
  - Migration (keep moving)

Point Defect Balance

Change = Gain – Loss (where have we used this format before?)

- What are the possible gain terms?
  - Displacement production
- What are the possible loss terms?
  - Recombination
  - Loss to sinks
  - Diffusion

Point Defect Balance

Change = Gain – Loss

- What are the possible sinks?
  - Grain boundaries
  - Dislocations
  - Impurities
  - Free surfaces
  - Incoherent precipitates

For point defects, sinks are higher dimensional defects

Cluster Dynamics

- Equations that govern the growth and shrinkage of clusters via defect emission and absorption

- Same way to use equation to describe it:

$$\frac{\partial Cluster}{\partial t} = Gains - Losses$$

Cluster Dynamics (for Vacancies)

$$\frac{\partial v_j}{\partial t} = Gains - Losses$$

- What are the gain terms?
  - Direct production
  - Absorption of same type defects & clusters from smaller clusters
  - Emission of same type defects & clusters from larger clusters
  - Absorption of different types of defects & clusters from larger clusters

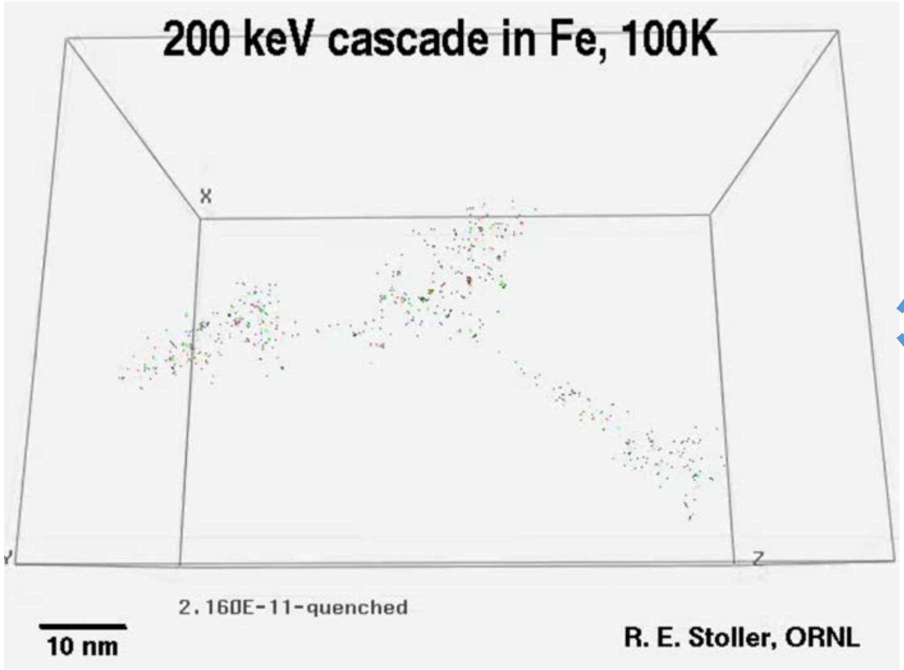
Cluster Dynamics (for Vacancies)

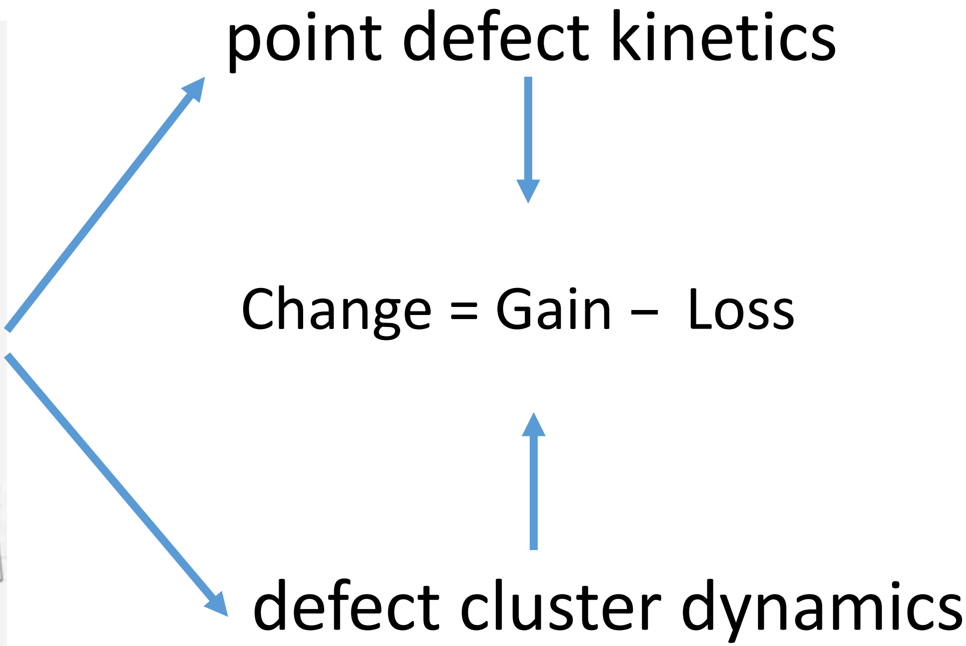
$$\frac{\partial v_j}{\partial t} = Gains - Losses$$

- What are the loss terms?
  - Direct destruction (loop collapse, etc.)
  - Absorption of same type defects & clusters from size *j*
  - Emission of same type defects & clusters from size *j*
  - Absorption of different types of defects & clusters from size *j*

Bridging radiation damage event to radiation damage effects

http://www-personal.umich.edu/~gsw/movies.html





Bridging radiation damage event to radiation damage effects

point defect kinetics

Change = Gain – Loss

defect cluster dynamics

Population and **distribution** of point defects and defect clusters

Radiation damage effects

Population and **distribution** of point defects and defect clusters

↓

Radiation Damage Effects

Physical effects

- RIS (Radiation-induced segregation)
- Nucleation and growth of dislocation loops and voids
- Phase Stability

Mechanical effects: when stress is applied

- Irradiation hardening and deformation
- Fracture and Embrittlement
- Irradiation Creep and Growth
- IRSCC (irradiation-assisted stress corrosion cracking)

RIS (Radiation induced segregation)

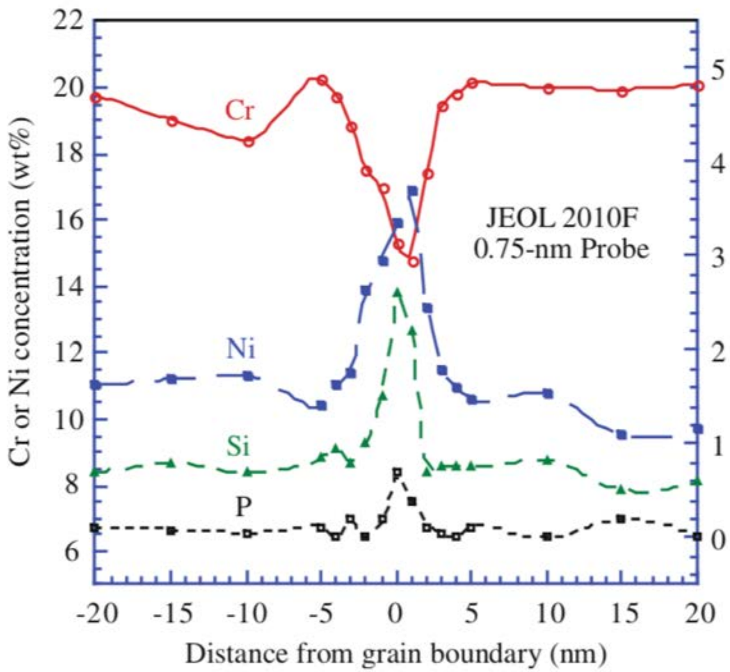


Fig. 6.1 Radiation-induced segregation of Cr, Ni, Si, and P at the grain boundary of a 300 series stainless steel irradiated in a light water reactor core to several dpa at ~300 °C (after [1])

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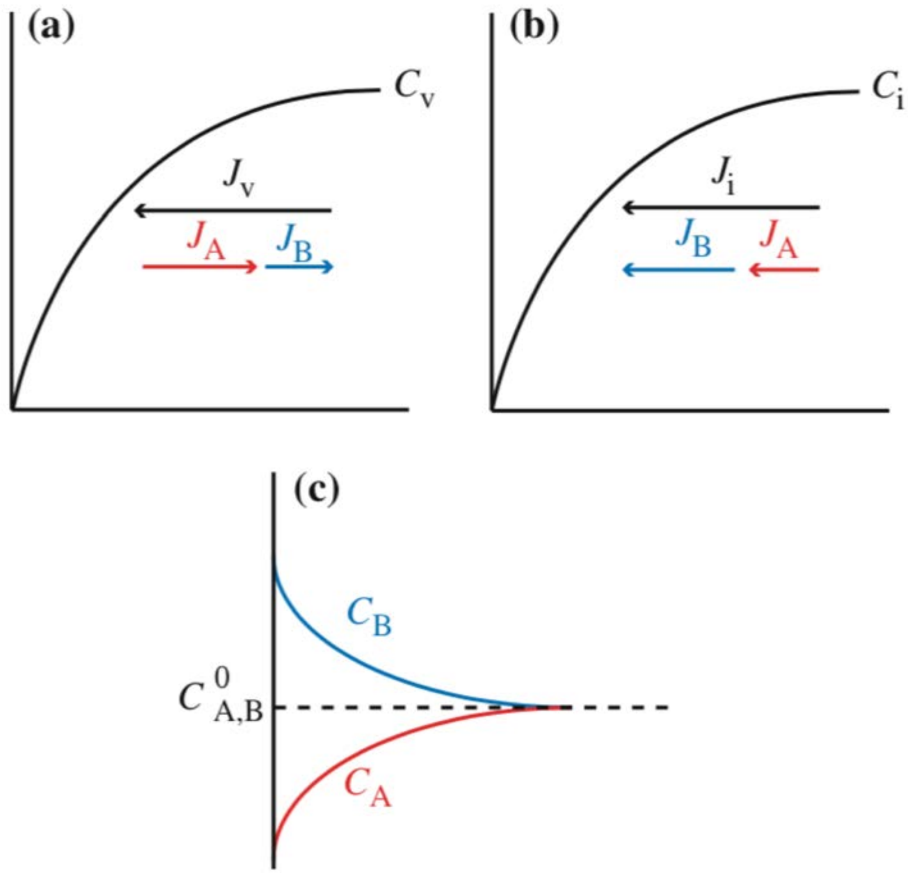
RIS: the spatial redistribution of solute and impurity elements in the metal at elevated temperature under irradiation

Influence: changes in the local properties of the solid, which may induce susceptibility to a host of processes that can degrade the integrity of the component.

RIS was discovered around 1970s

Mechanism of RIS



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Excess point defects produced from radiation

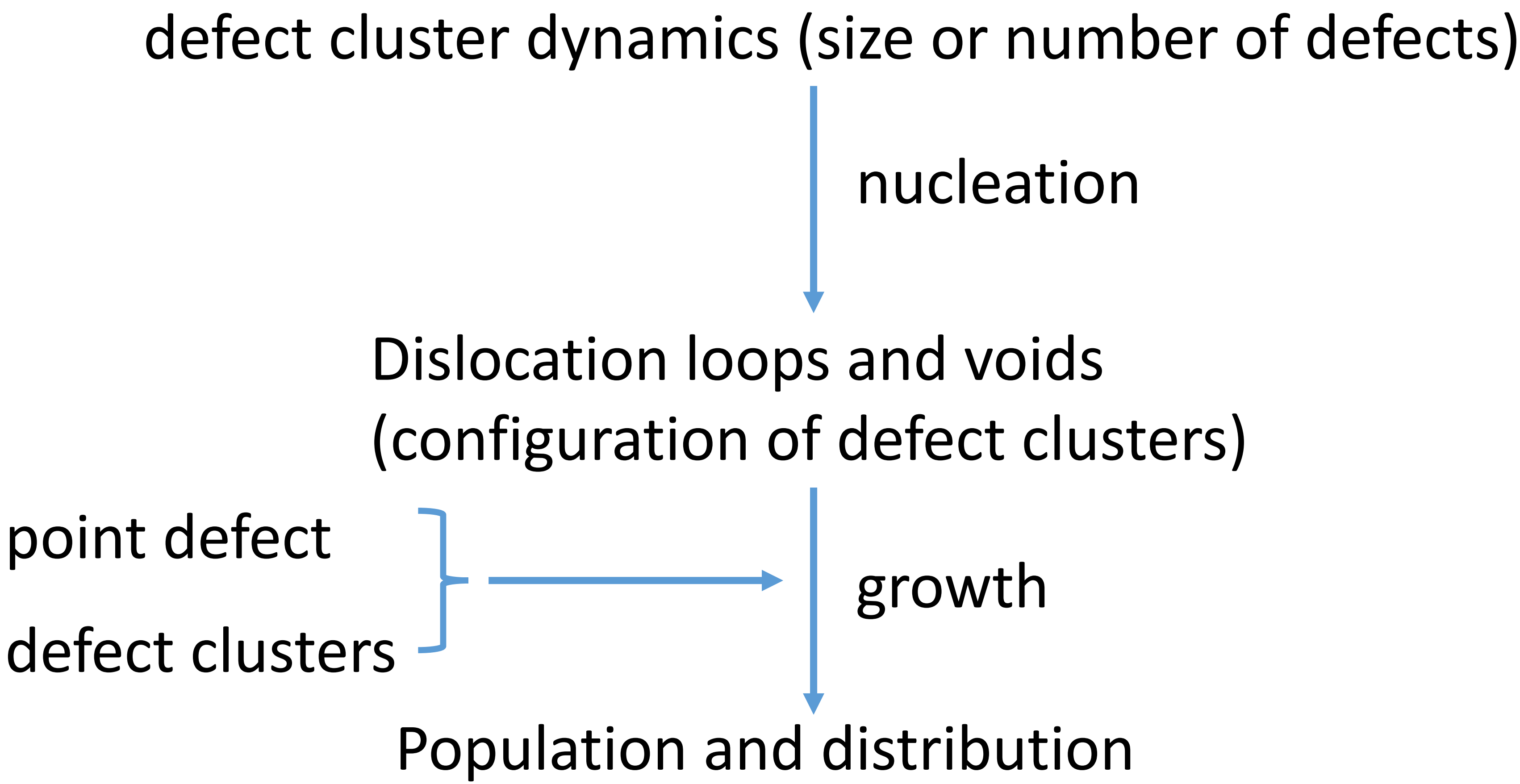
Defect fluxes to defect sinks (GB)

Different atoms couple differently to the defect fluxes

RIS Effect on Material Properties

- Corrosion
  - **Cr depletion**
- Embrittlement, Fracture Toughness Reduction
  - **P, S segregation** to microstructural sinks
- Hardening, Strengthening
  - **Precipitate formation** by smaller vacancy solutes (Mo, Cu, Si) towards sinks
  - **Precipitate formation** by interstitial solutes (C, N) towards sinks

Nucleation and growth of dislocation loops and voids



Example dislocation loops

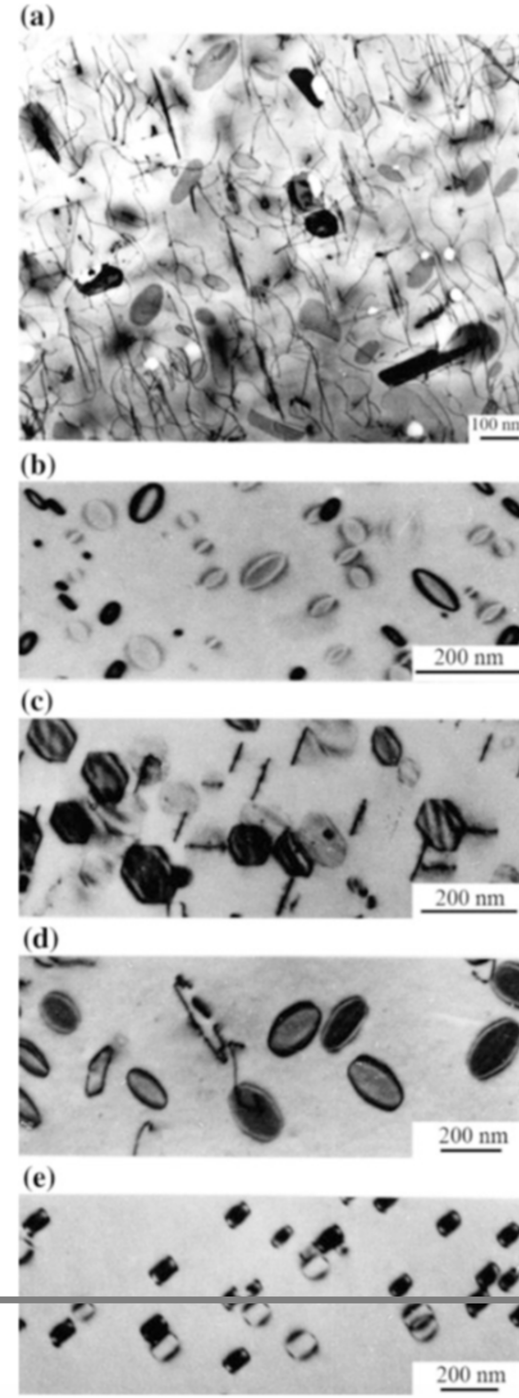


Fig. 7.66 Images of large Frank loops in (**a**) a 300 series stainless steel irradiated at 500 °C to a dose of 10 dpa (from [42]), and (**b**)–(**e**) in irradiated aluminum, copper, nickel, and iron, respectively (after [18])

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Example voids

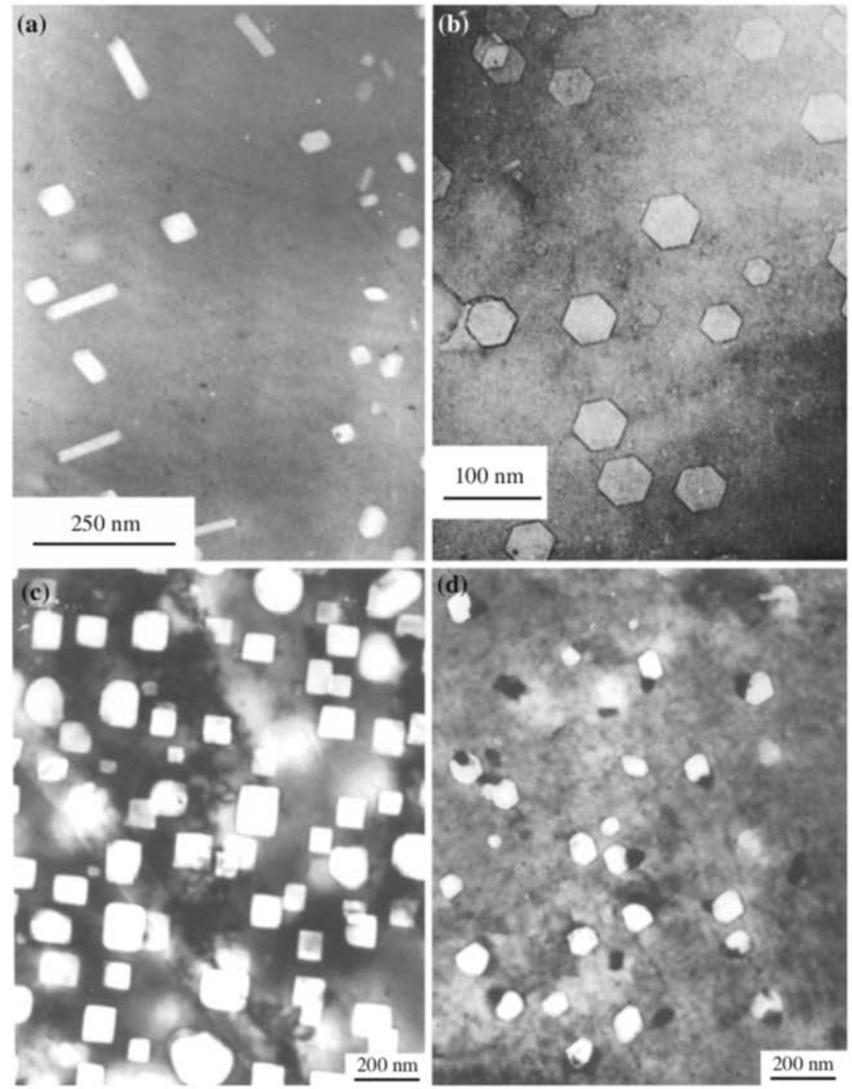

Fig. 8.1 Micrographs of irradiation-induced voids in **(a)** stainless steel, **(b)** aluminum, **(c)** and **(d)** magnesium [2, 3]

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Voids lead to Swelling (dimensional change)

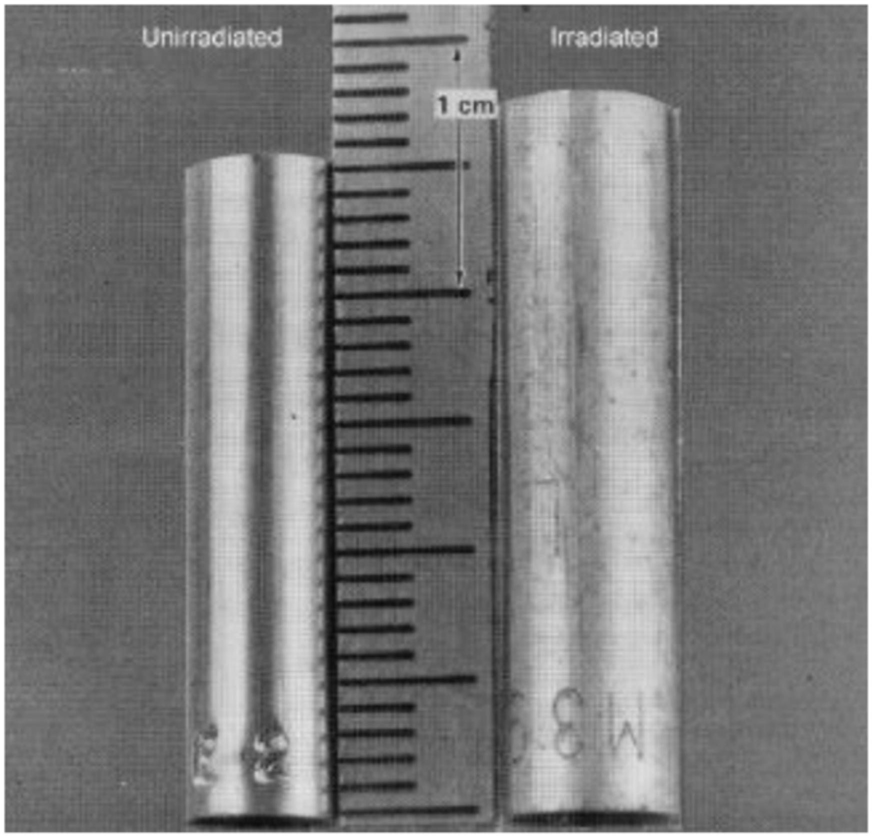
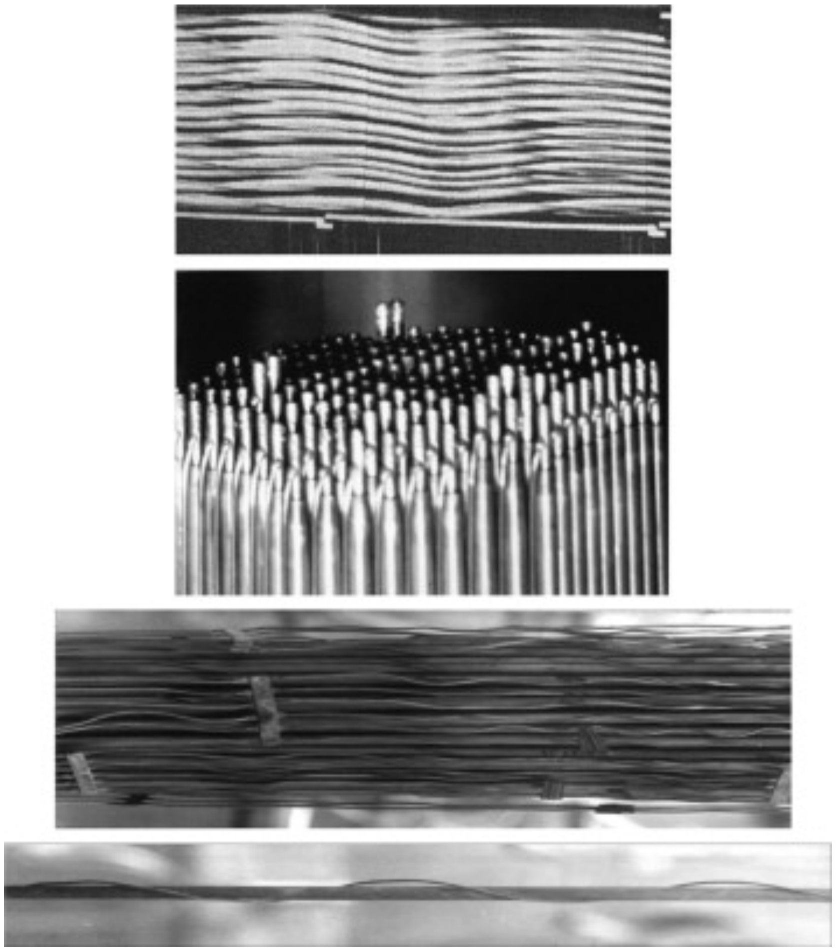


Figure 4.1: Photograph of 20% cold-worked 316 stainless steel rods before (left) and after (right) irradiation at 533°C to a fluence of 1.5×10^{23} neutrons m^{-2} in the EBR-11 reactor (Mansur, 1994).

Swelling is a big issue



- Spiral distortion of 316-clad fuel pins induced by swelling and irradiation creep in an FFTF fuel assembly where the wire wrap swells less than the cladding.

- Reproduced from Makenas, B. J.; Chastain, S. A.; Gneiting, B. C. In *Proceedings of LMR: A Decade of LMR Progress and Promise*; ANS: La Grange Park, IL, 1990; pp 176–183; (middle) Swelling-induced changes in length of fuel pins of the same assembly in response to gradients in dose rate, temperature, and production lot variations as observed at the top of the fuel pin bundle. Reproduced from Makenas, B. J.; Chastain, S. A.; Gneiting, B. C. In *Proceedings of LMR: A Decade of LMR Progress and Promise*; ANS: La Grange Park, IL, 1990; pp 176–183; (bottom) swelling-induced distortion of a BN-600 fuel assembly and an individual pin where the wire swells more than the cladding. Reproduced from Astashov, S. E.; Kozmanov, E. A.; Ogorodov, A. N.; Roslyakov, V. F.; Chuev, V. V.; Sheinkman, A. G. In *Studies of the Structural Materials in the Core Components of Fast Sodium Reactors*; Russian Academy of Science: Urals Branch, Ekaterinburg, 1984; pp 48–84, in Russian.

Phase instability

Local enrichment or depletion of solute atoms

↓

Formation or dissolution of phases

↓ Extreme case

Amorphization

Radiation damage effects

Population and **distribution** of point defects and defect clusters

↓

Radiation Damage Effects

Physical effects

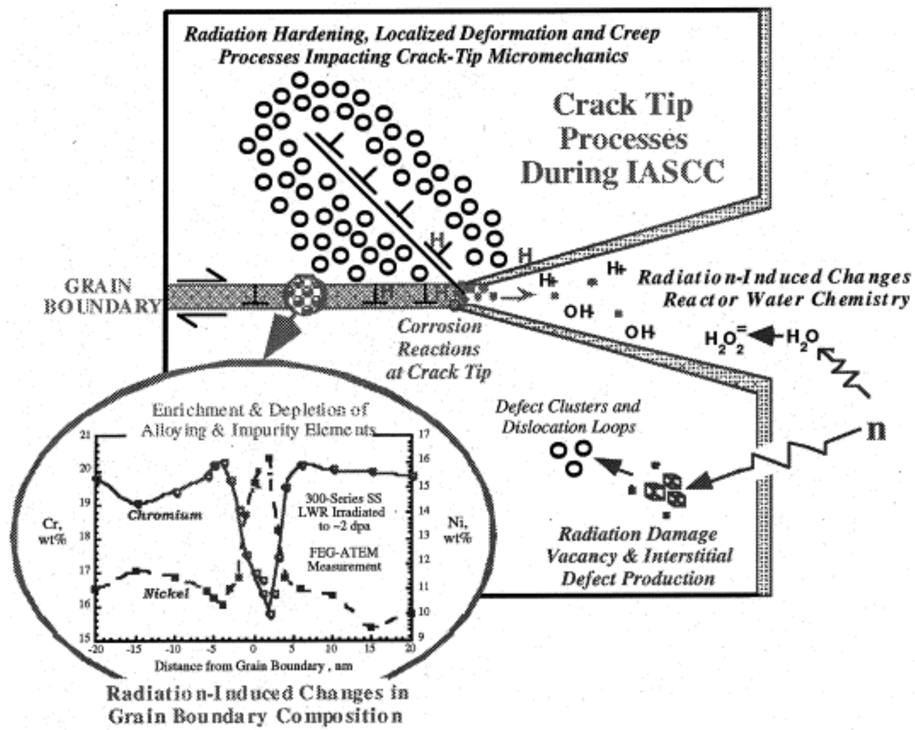
- RIS (Radiation-induced segregation)
- Nucleation and growth of dislocation loops and voids
- Phase Stability

Mechanical effects: when stress is applied

- Irradiation hardening and deformation
- Fracture and Embrittlement
- Irradiation Creep and Growth
- IRSCC (irradiation-assisted stress corrosion cracking)

Irradiation Assisted Stress Corrosion Cracking (IASCC)

- Stress corrosion cracking requires:
  - Tensile stress
  - Susceptible material
  - Aggressive environment
- Radiation can:
  - Increase susceptibility
  - Generate stresses
  - Induce hydrolysis, free radicals, more corrosion



Courtesy Elsevier, Inc., https://www.sciencedirect.com. Used with permission.

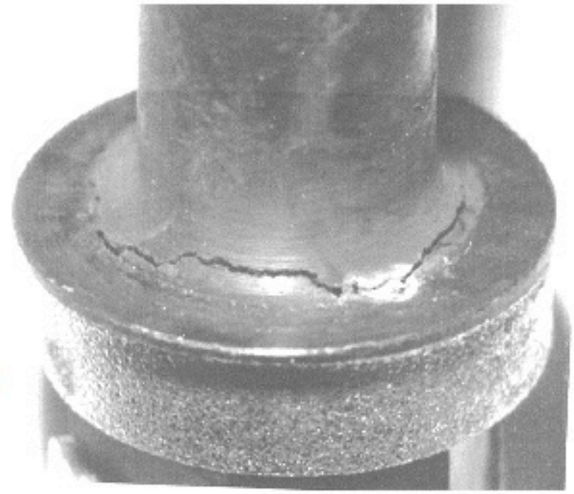
Schematic illustrating mechanistic issues believed to influence crack advance during IASCC of austenitic stainless steels

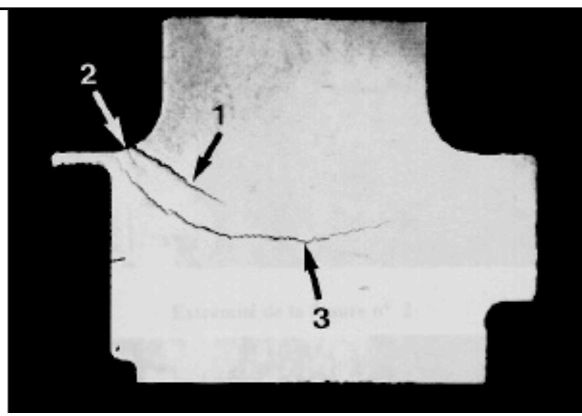
S.M Bruemmer et al.,, *J. Nucl. Mater.*, 274(3):299-314 (1999)

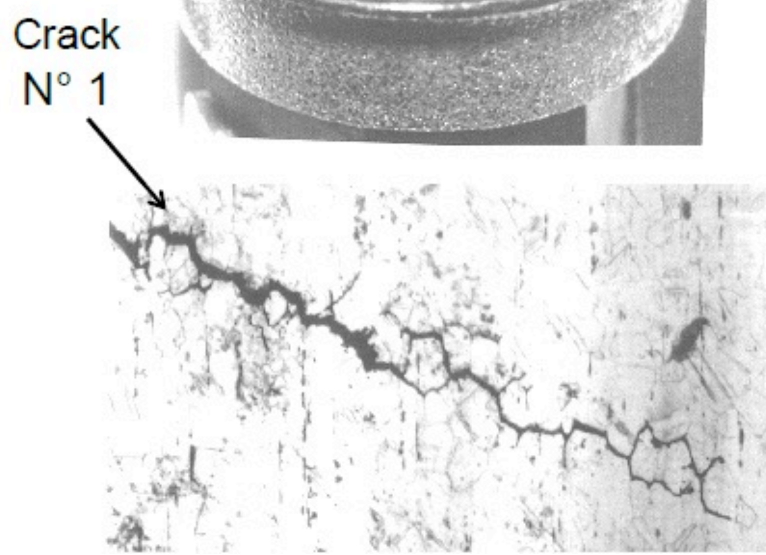
IASCC of PWR Baffle Bolts

(http://www.sciencedirect.com/science/article/pii/S1369702110702200)

Two big problems here: (1) The bolts can break, loosening the baffle. (2) The bolt heads get swept up by the coolant, becoming foreign material.







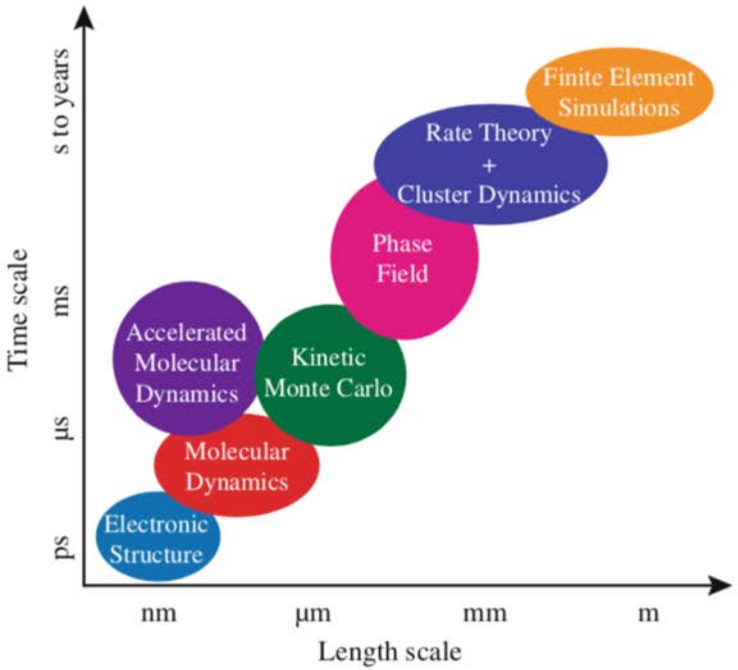
General challenges

- Quantify radiation damage
  - Problems using DPA
    - DPA is calculated, not measurable
    - Too many steps from DPA to the effects
    - Difficult to compare different conditions
  - What is a good unit?
    - Not that general (not too far away from effects)
    - Not that specific (should not only for one specific effect)
    - Can be calculated
    - Can be measured

General challenges

- Simulation limitations

Fig. 3.13 Time and length scale for radiation damage evolution and the corresponding simulation methodologies (courtesy F. Gao)



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General challenges

- Experimental constrains
  - Limitations for experiments
    - Time
    - Cost
    - Safety
    - Sample examination
  - What is a good experimental design
    - Not that simplified (not too different with application conditions)
    - Not that complex (not involving too many variables)