Fundamentals of Radiation Damage

Bangor University
Michael J.D. Rushton
m.rushton@bangor.ac.uk

What is Radiation Damage?

Radiation Damage:

The disruption to the initial (undamaged) structure of a solid caused by high-energy radiation passing through it (defect production).

electronic defects and structural defects

each damage event occurs over ~10^{-11} seconds

Why should we care?

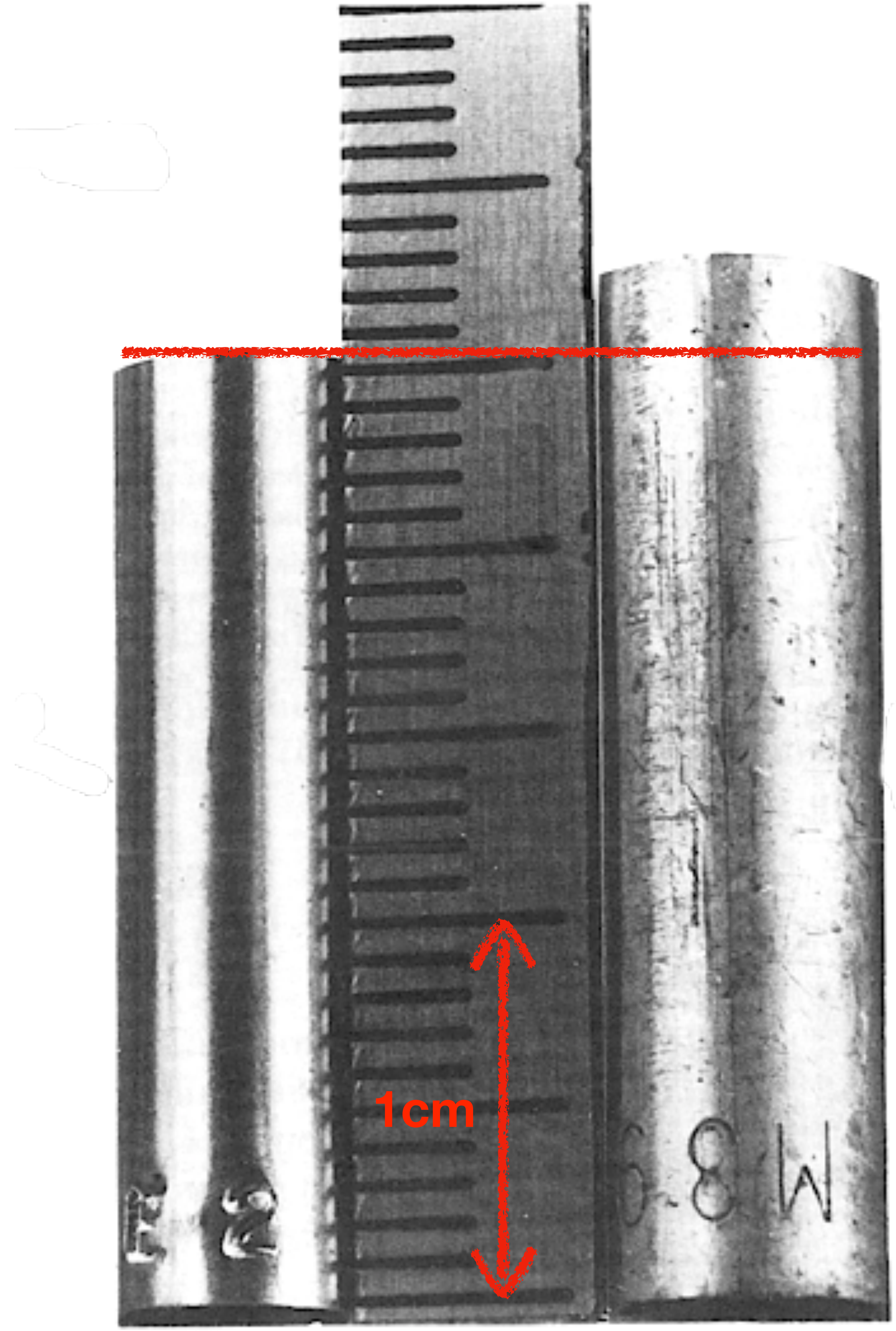
- Radiation damage is the major degradation and ageing issue for materials used in the nuclear industry. It restricts materials performance and defines lifetime.
- Radiation damage (mostly irradiation induced displacements) can affect the physical, mechanical and chemical properties of the solid.
- Examples:
    - dimensional changes, phase changes, amorphization, optical
    - embrittlement, hardening, creep
    - phase separation, re-solution, corrosion

Examples of Adverse Effects of Irradiation

- Possible **dissociation** and/or **activation** of coolant
- **Thermal** - softening, creep of fuel cladding
- **Embrittlement** e.g. RPV welds
- **Enhanced corrosion of cladding and RPV**
- **Fuel/clad interaction**
- **Fission gas release** → increase rod internal pressure
- **Segregation of elements**
- → changes in thermo-physical and mechanical properties, and appearance of new elements must be understood and accommodated by the designer.

Easily Observed: Swelling

316 Stainless Steel
20% Cold Work

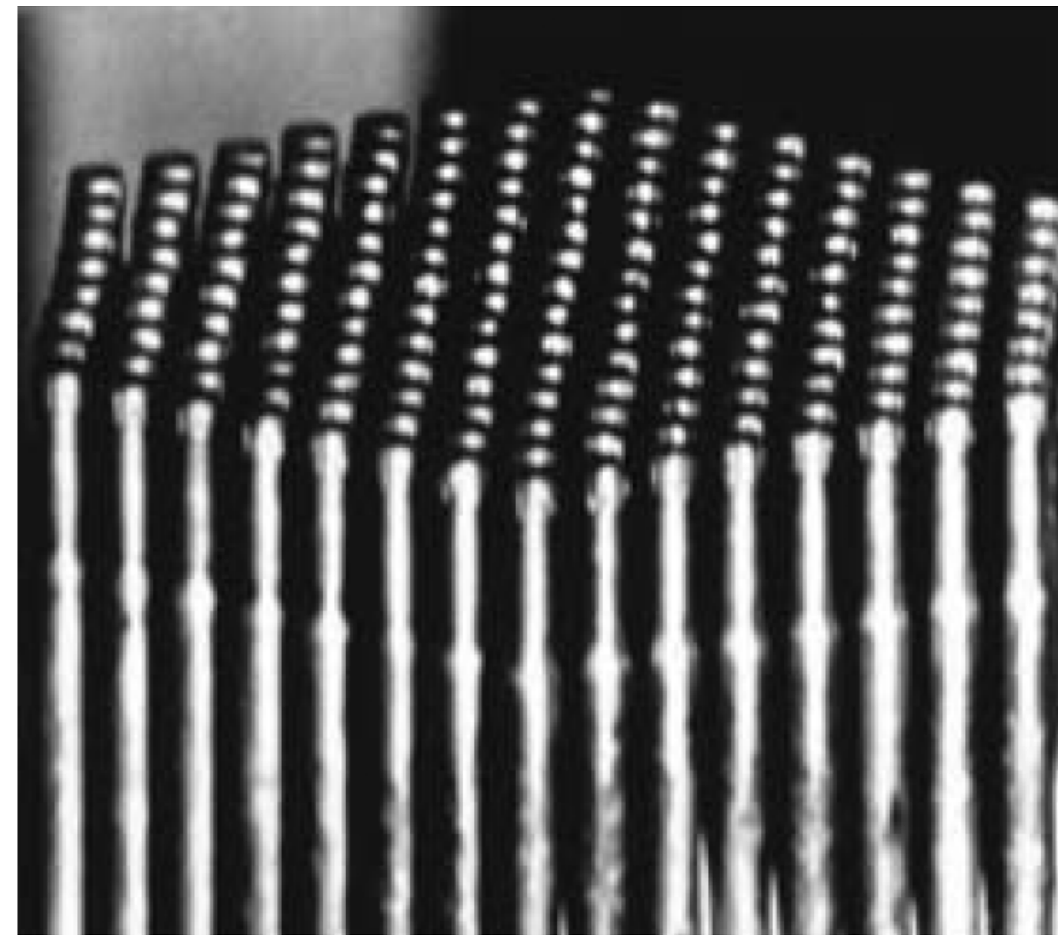


Unirradiated (control)

Irradiated (high fluence)

Dimensional Stability is Compositionally Dependent

D-9 stainless steel
austenitic
15Cr-15Ni stabilised with Ti



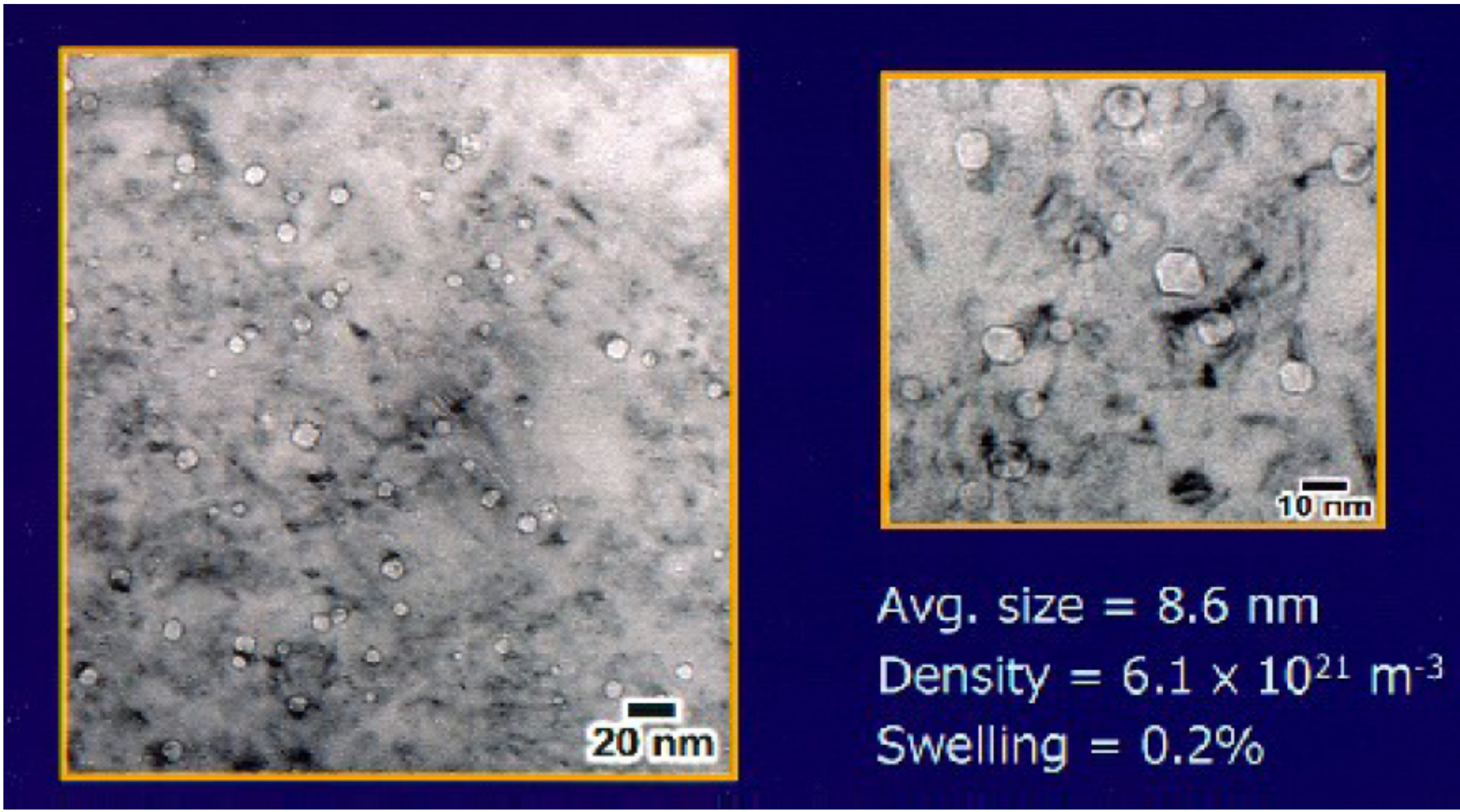
HT-9 ferritic-martensitic steel
limited fracture toughness and high temperature strength

Irradiated to 75 dpa a FFTF

After F.A. Garner, in Nuclear Materials (1996) p 420

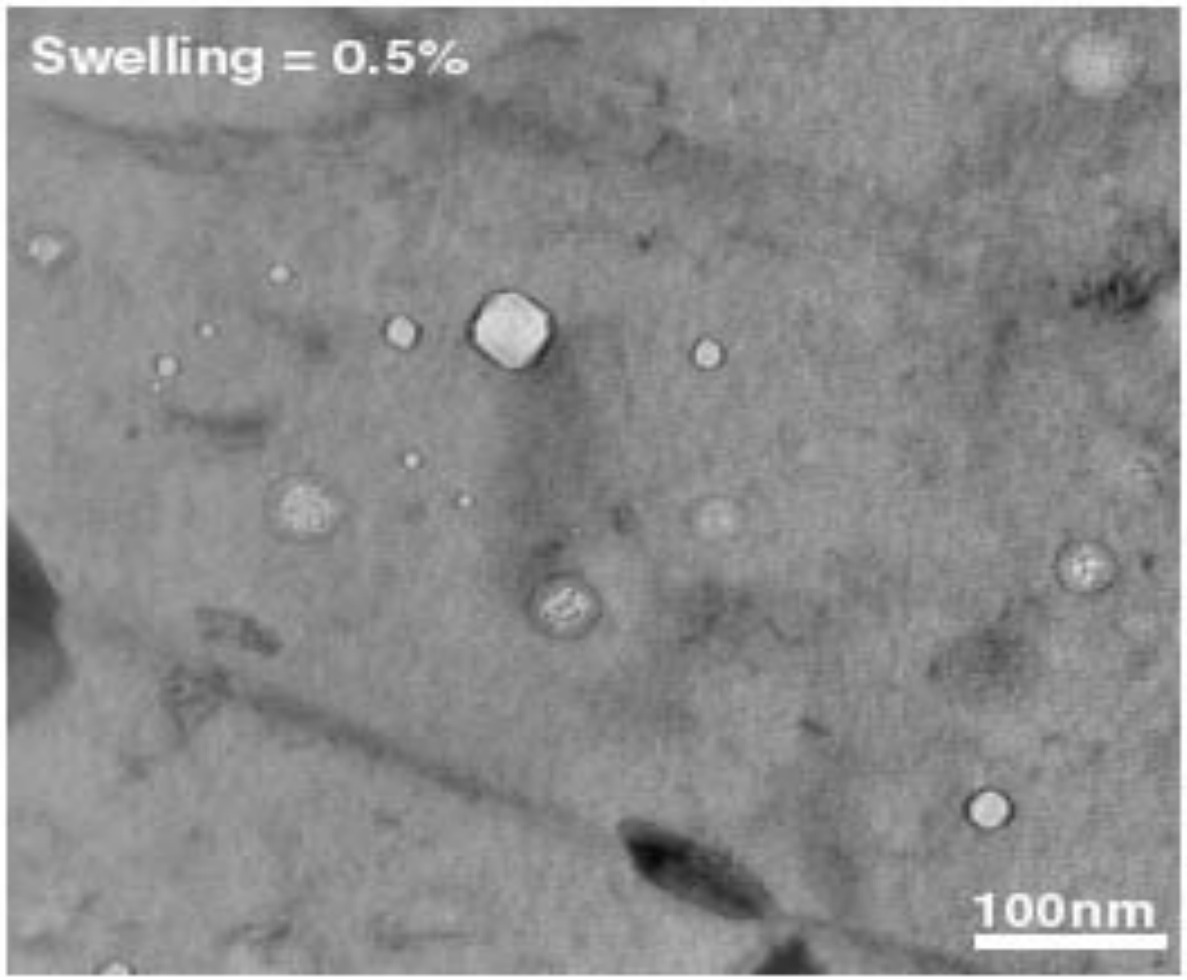
Cavities in 316 Stainless Steel

Voids and helium/hydrogen bubbles in a baffle-bolt extracted from Tihange 1 (Belgium), a 962 MWe PWR (TEM carried out at PNNL)

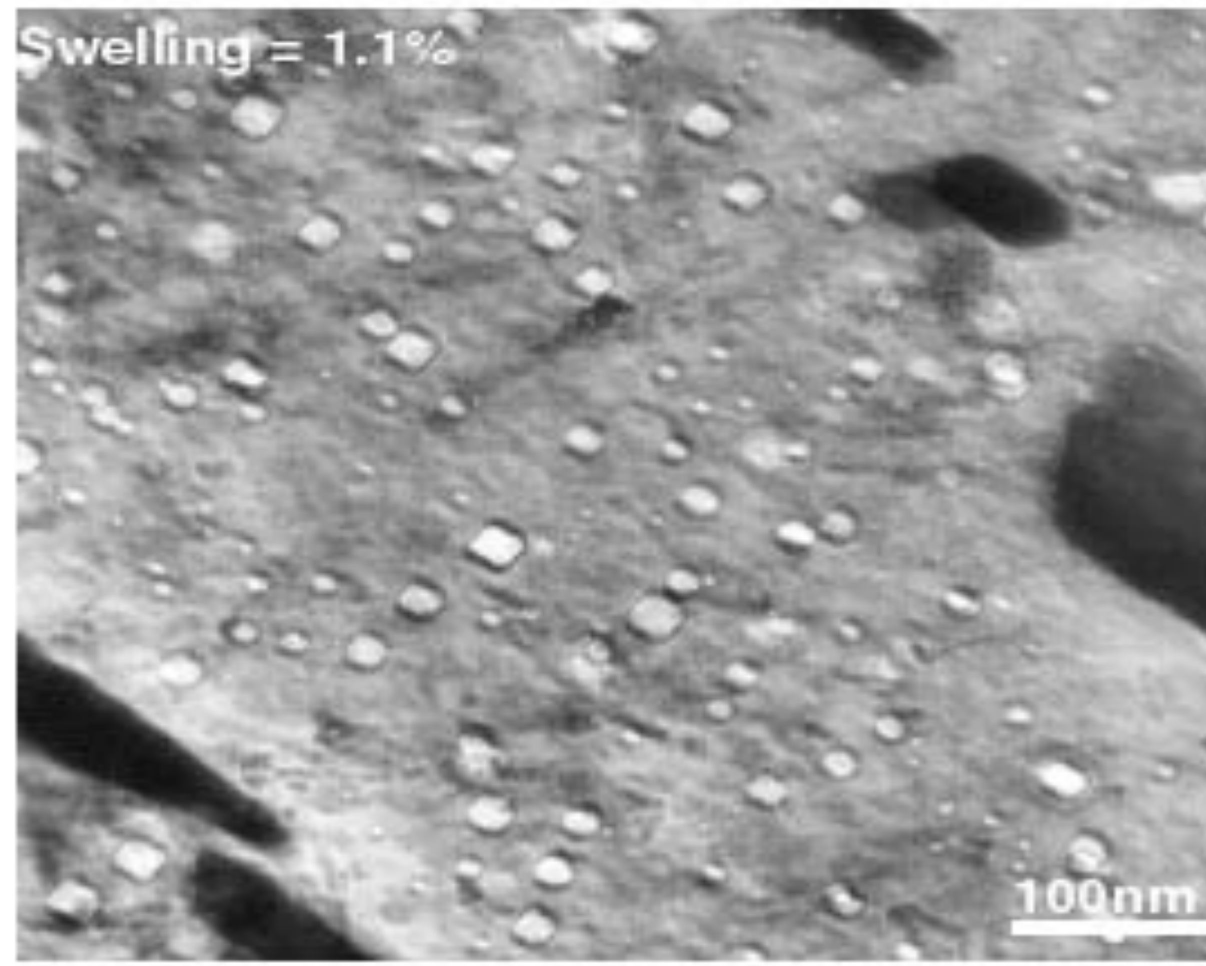


Radiation Induced Bubbles

HFIR Irradiation at 400ºC to 51 dpa



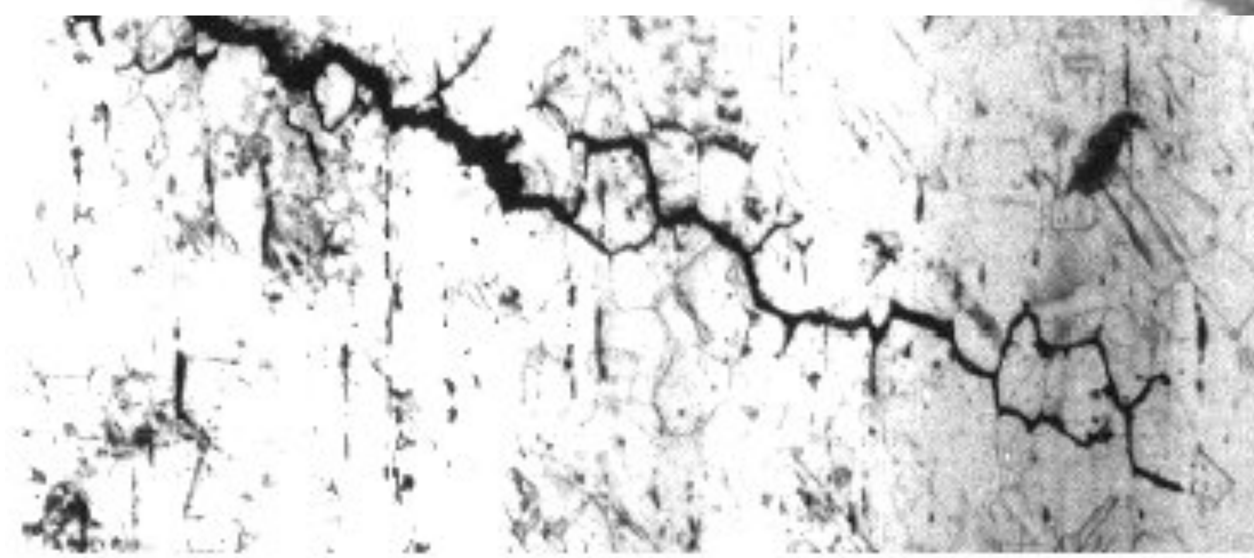
F82H (36 appm He)



^{10}B-doped F82H (330 appm He)

$$^{10}_{5}\mathrm{B} + ^{1}_{0}n \longrightarrow ^{7}_{3}\mathrm{Li} + ^{4}_{2}\mathrm{He}$$

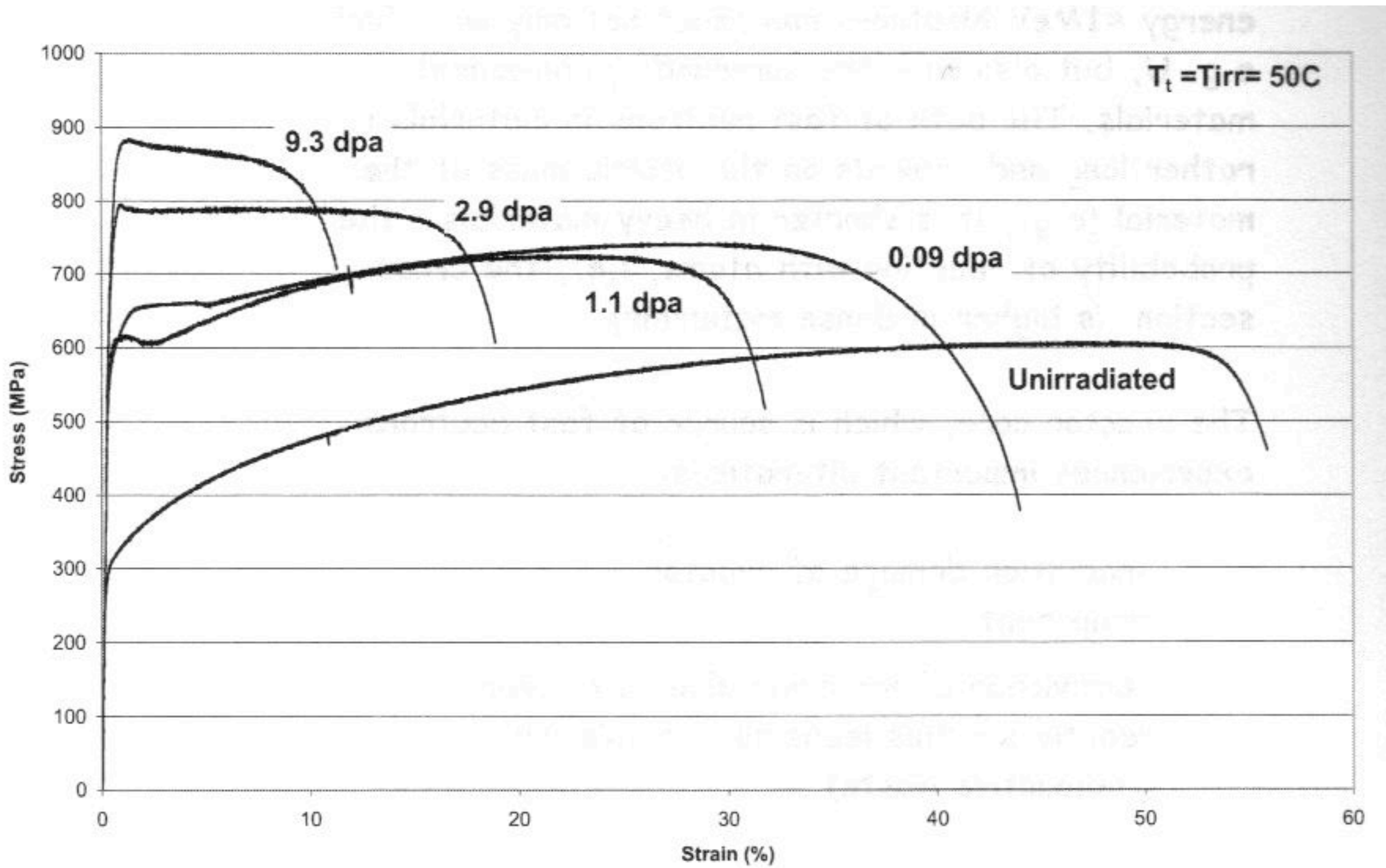
E. Wakai et al. J. Nucl. Mater. 283-287 (2000) 799



Irradiation Assisted Stress Corrosion Cracking

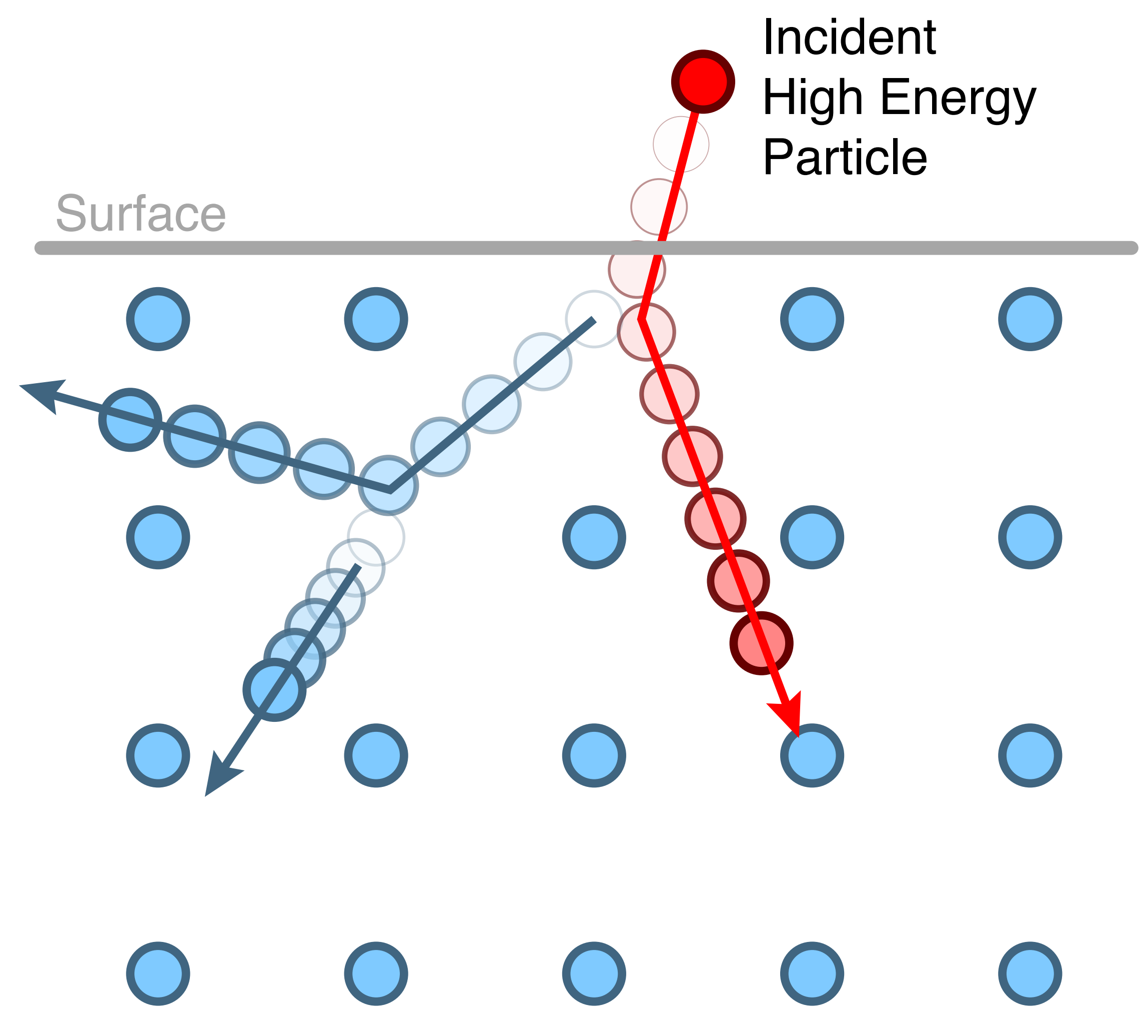
A form of inter-granular stress corrosion cracking that occurs in materials that are subject to high neutron fluences. Probably associated with radiation induced segregation e.g. depletion of Cr at the grain boundaries.

Stress/Strain Curves show increases in yield stress and decrease in elongation in 316L steel after irradiation.



Radiation Damage at the Atomic Scale

- High energy particles travel through a material.
- Energy is transferred to the material:
  - Electronic Stopping.
  - Nuclear Stopping.
  - Radiative.



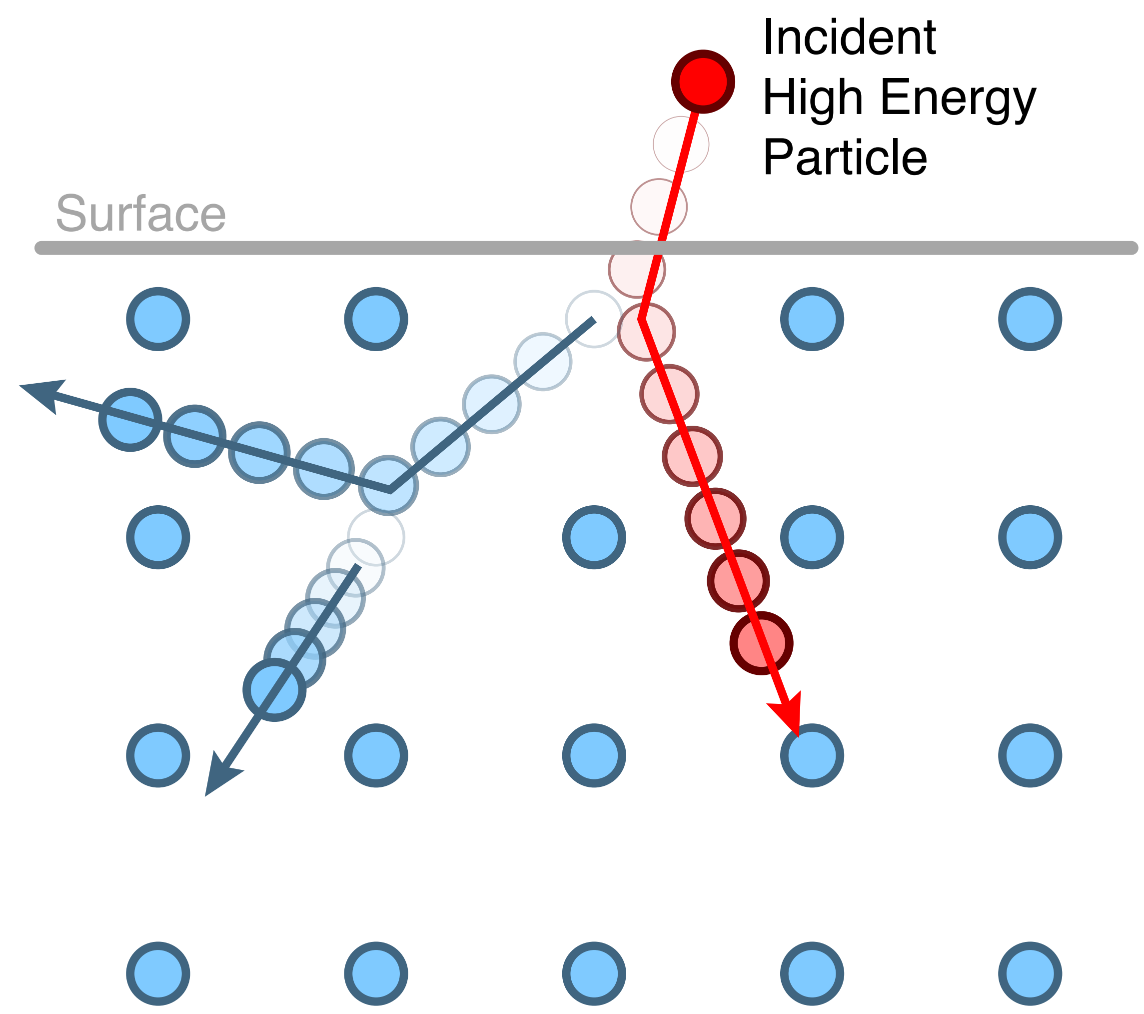
Radiation Damage at the Atomic Scale: Nuclear Stopping

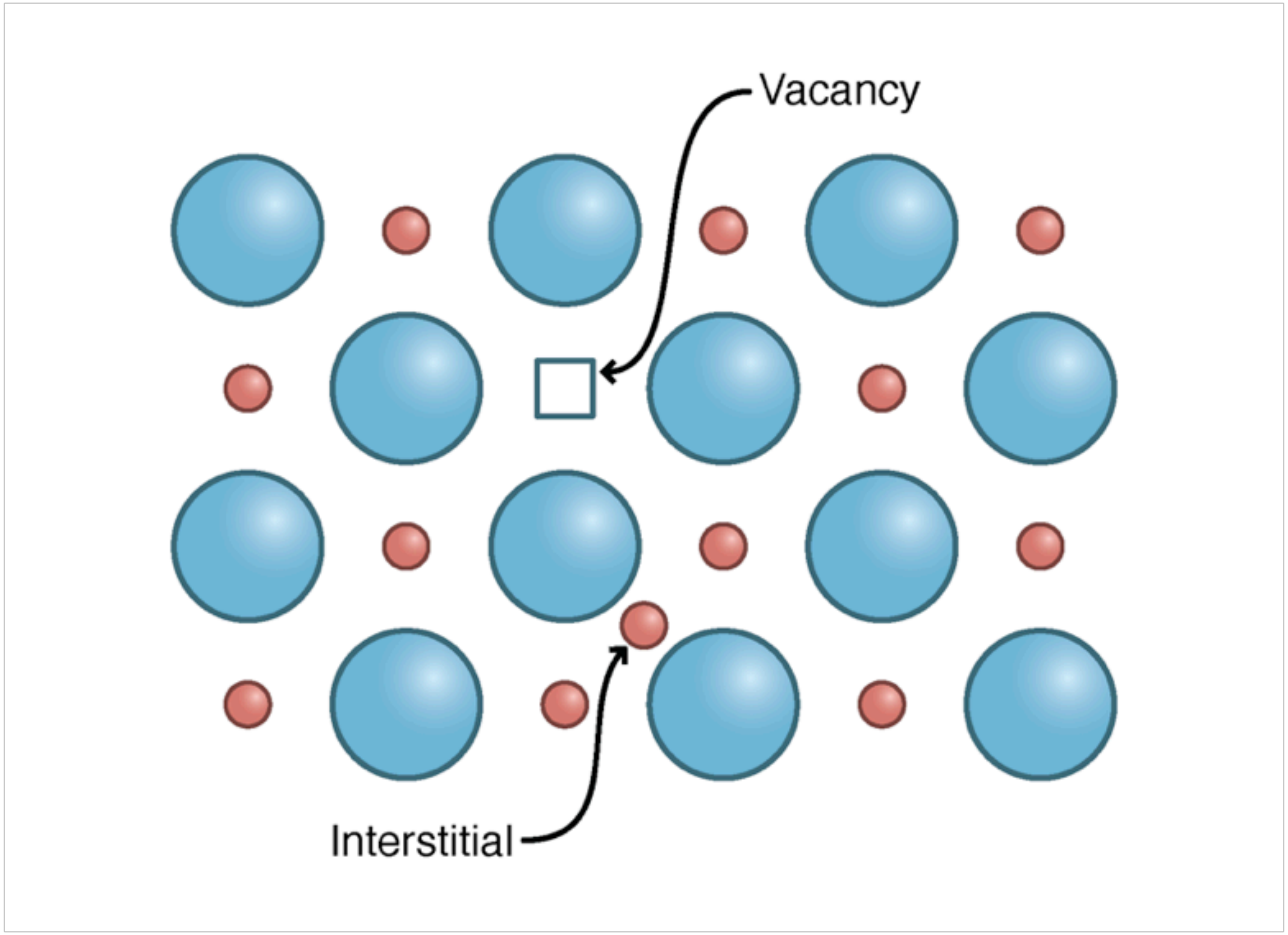
- The figure gives a schematic representation of a **collision cascade**. Nuclear stopping can be thought of as **atomic scale billiards.**

- If it is moving slowly enough the incident particle may collide with an atom in the material imparting energy to it. This first point of impact is the **primary knock-on atom (PKA).**

- A series of further collisions and even sub-cascades will take place until the energy of PKA has been dissipated.

- Fundamentally the **kinetic energy** of the incident particle is being converted into **potential energy** stored in the lattice (e.g. Wigner energy).





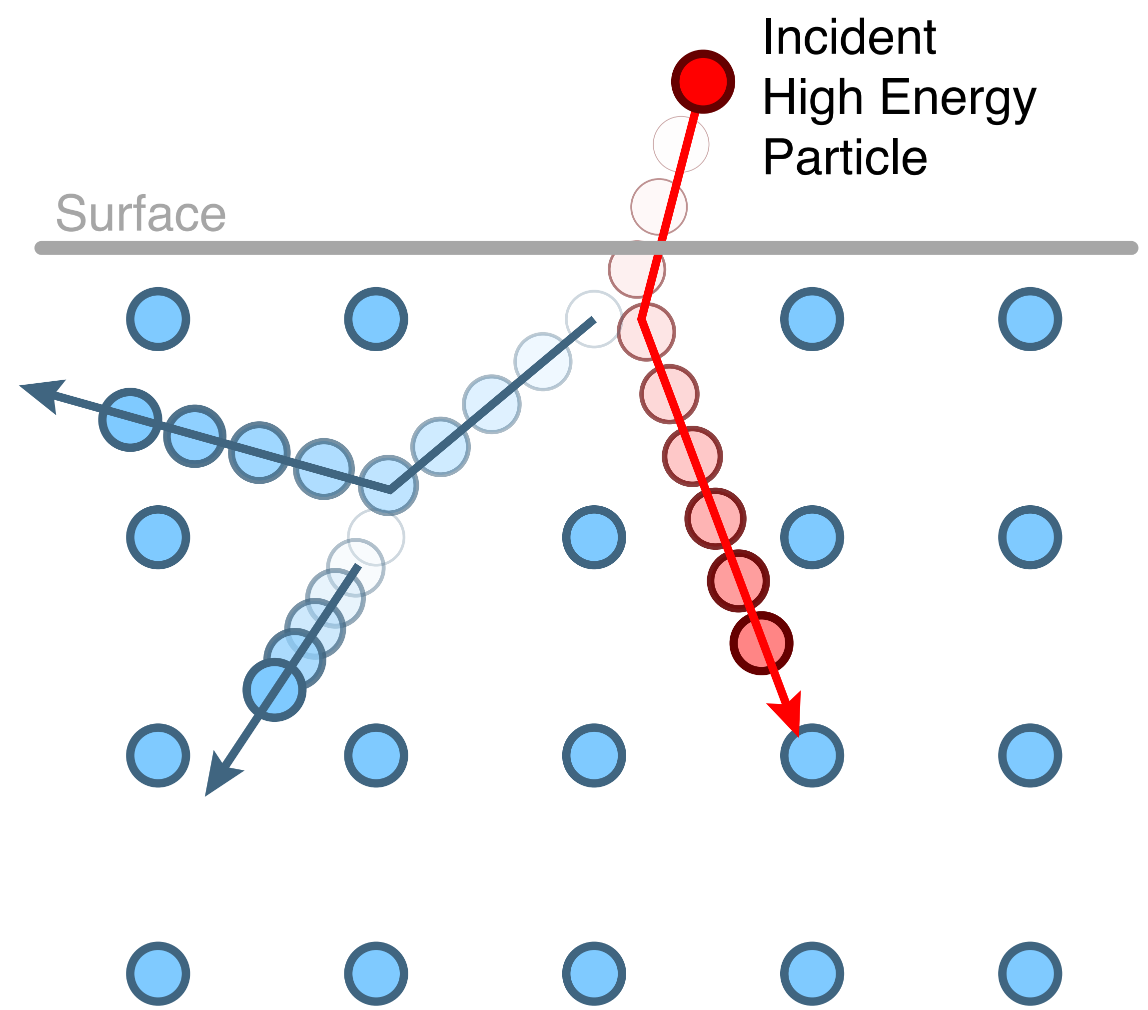
Defect Processes: The Frenkel Reaction

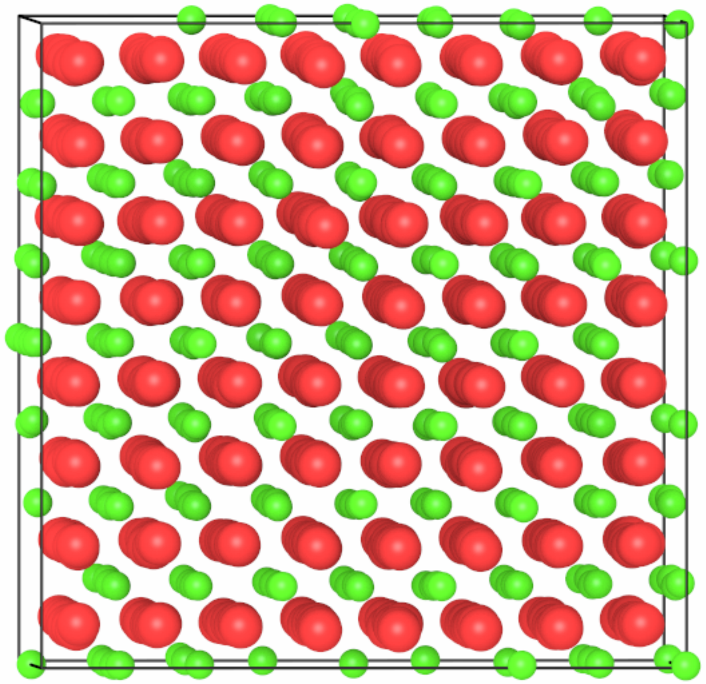
A lattice ion is displaced from its regular position in the crystal to form an interstitial, leaving a gap (or vacancy) in the lattice.

Radiation Damage at the Atomic Scale: Nuclear Stopping

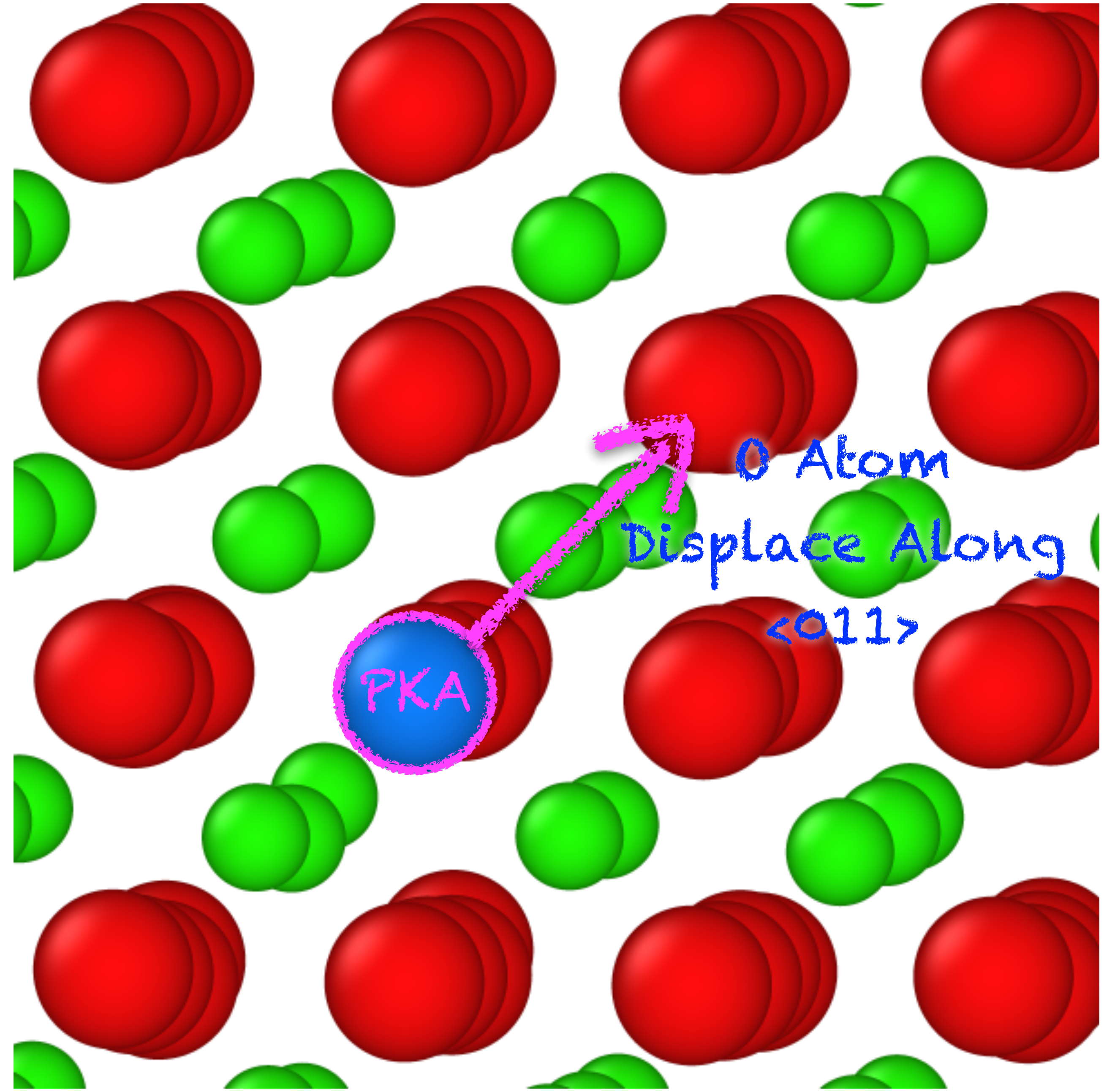
- This type of energy transfer is known as **nuclear stopping** because energies are high enough that **positively charged** atomic **nuclei** undergo Coulombic/electrostatic interaction.

- This can be described well using a shielded Coulomb interaction (e.g. the ZBL potential in the SRIM/TRIM code).





Simulation Cell
4×4×4 UO_2



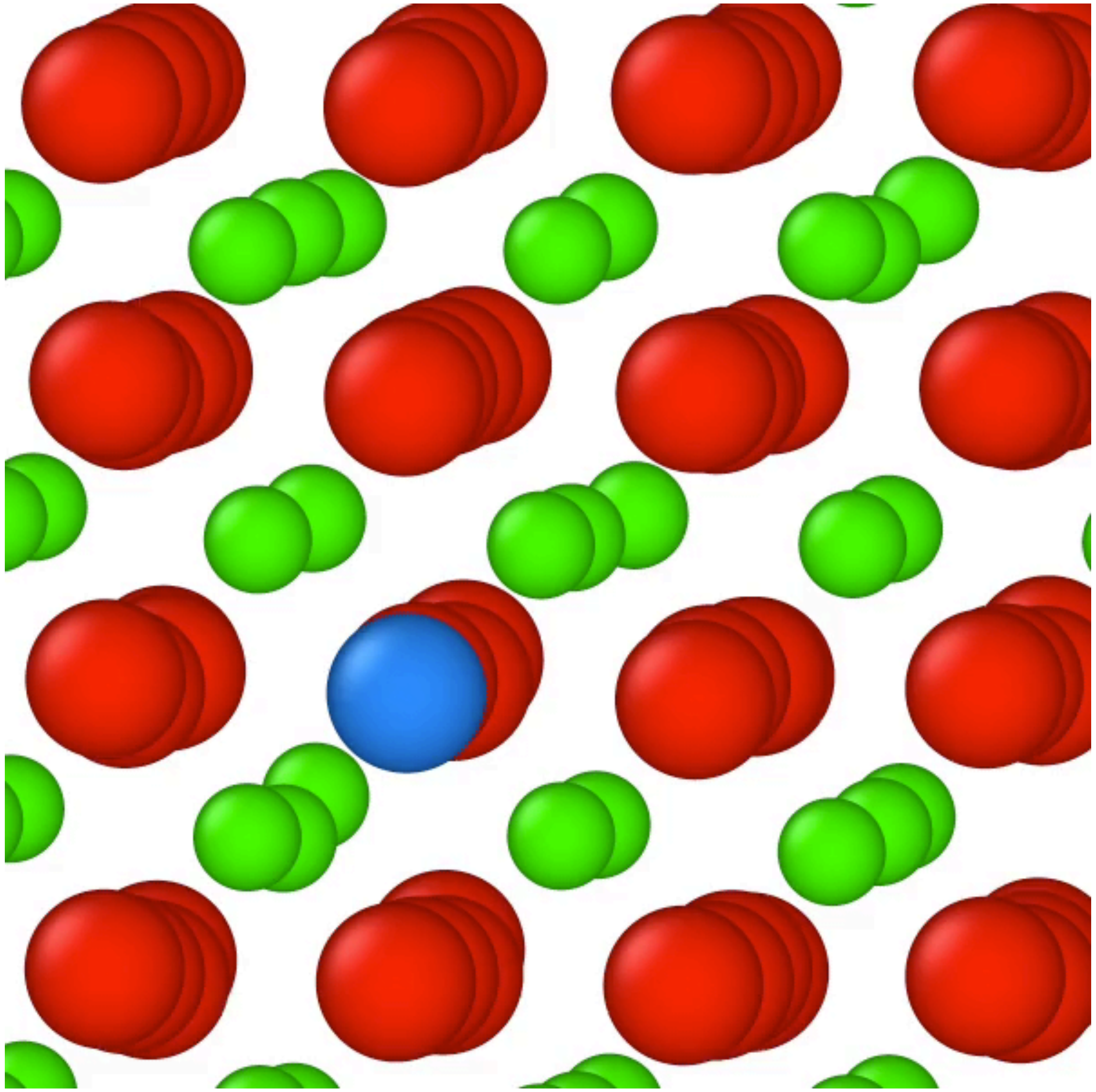
Oxygen

Uranium

View along <100> (yz plane)

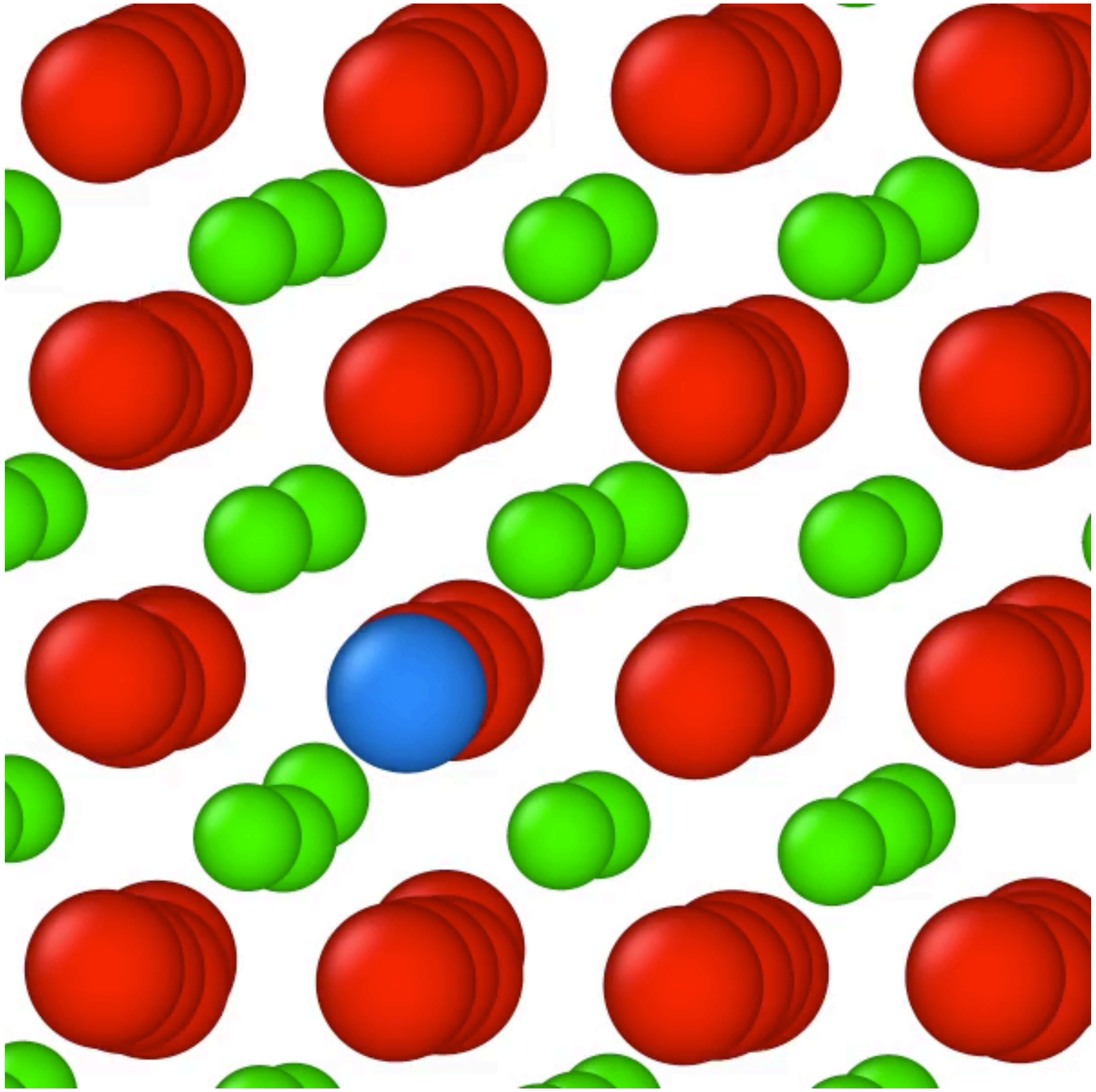
Threshold Displacement Energy (E_d)

The energy required to permanently displace an atom from its lattice site.



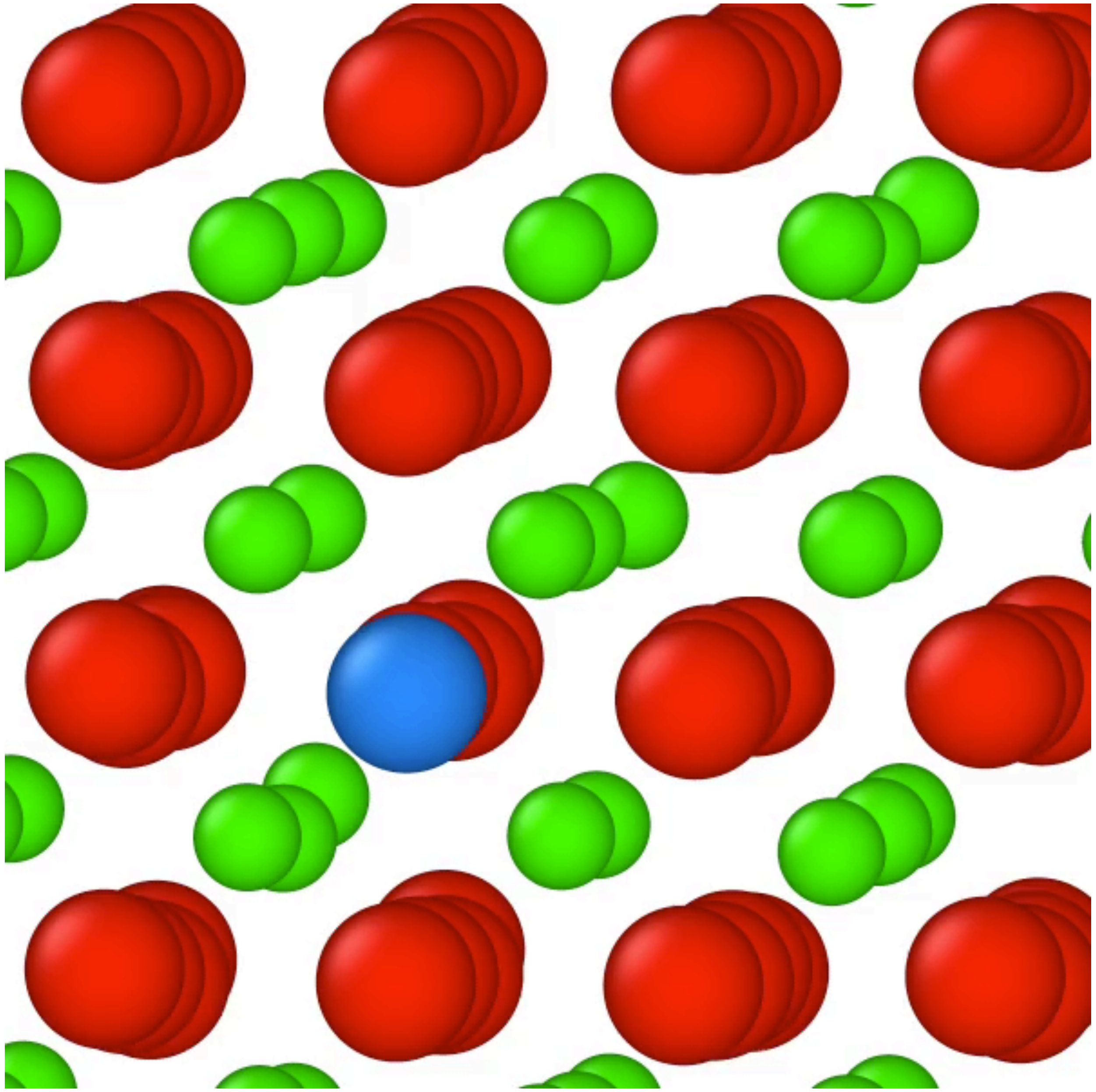
Threshold Displacement Energy (E_d)

Energy = 20eV



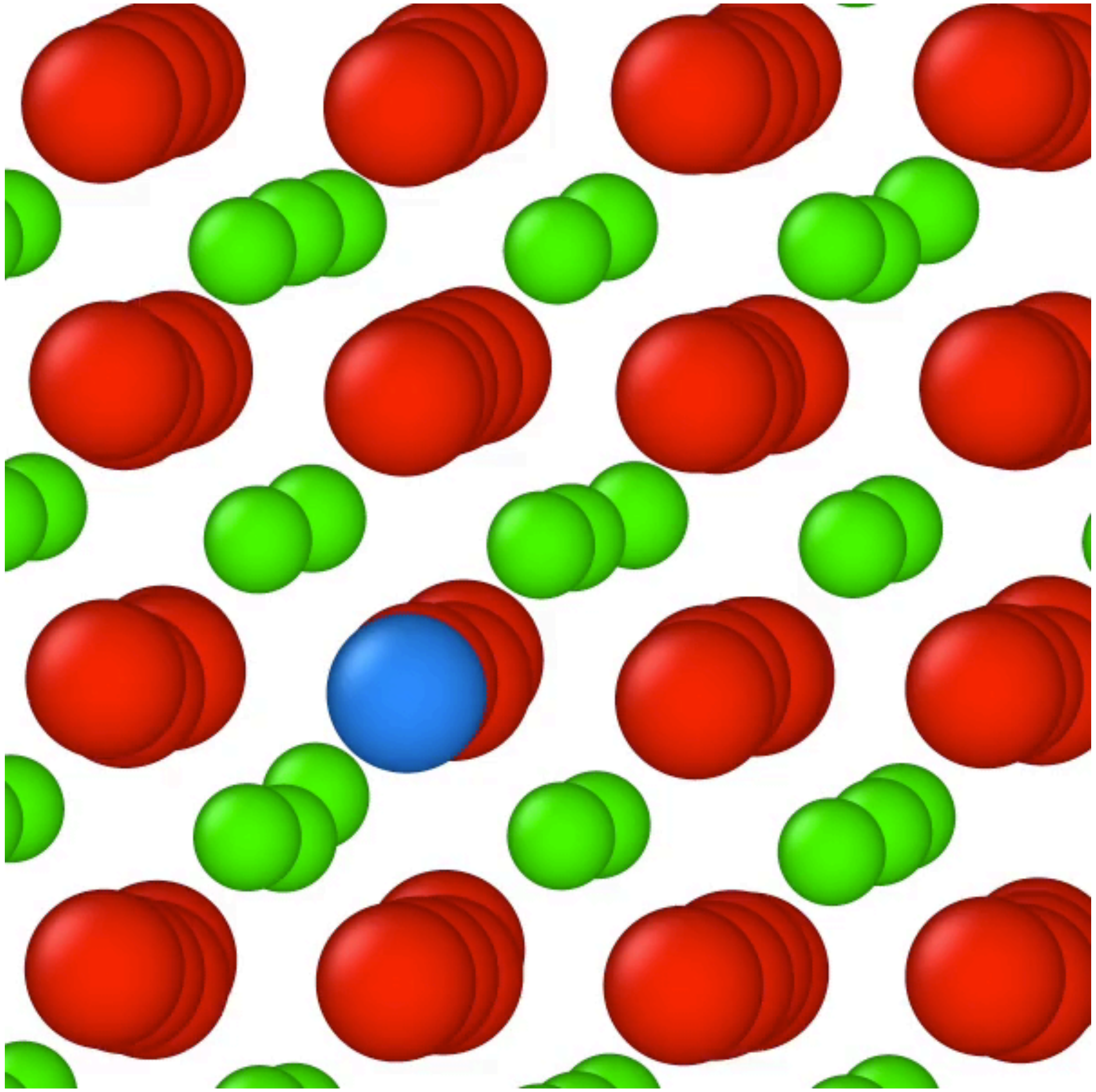
Threshold Displacement Energy (E_d)

Energy = 30eV



Threshold Displacement Energy (E_d)

Energy = 40eV

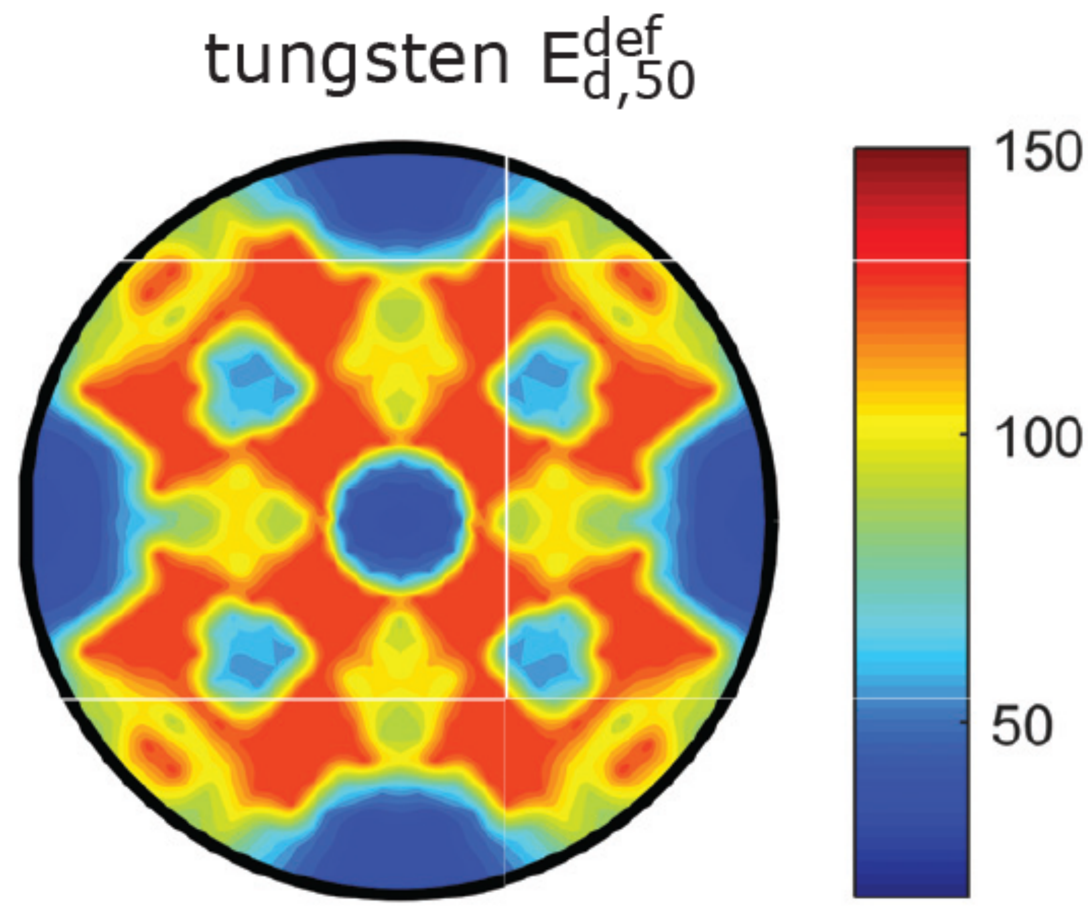


Threshold Displacement Energy (E_d)

Energy = 50eV

E_D and Crystallography

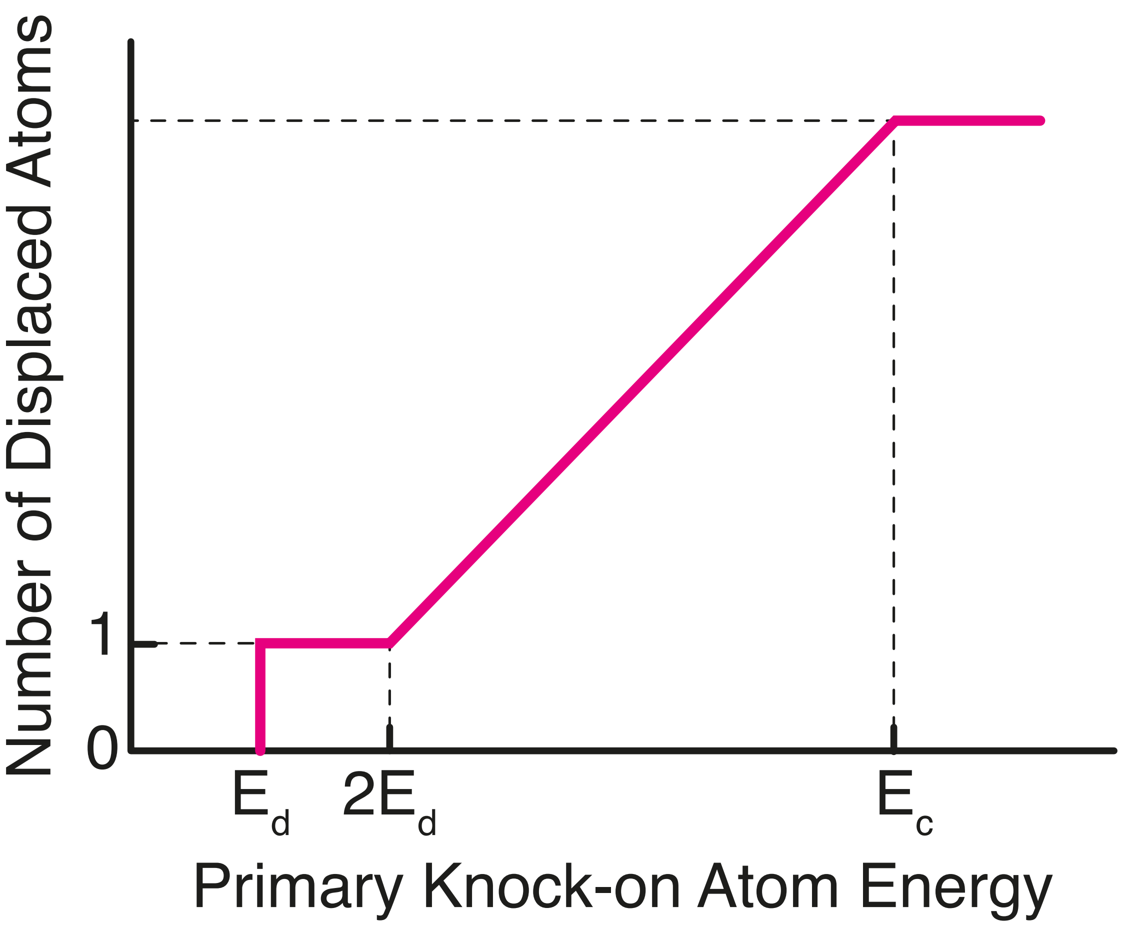
- Threshold displacement energy can vary significantly based on an atom's local environment. This can make choosing an appropriate E_D tricky.
- Figure shows stereographic projection of E_D values in tungsten for simulations where probability of displacement was 50% at given energy.
- Projection is viewed along <0001>



M.L. Jackson, "Atomistic Simulations of Materials for Nuclear Fusion", PhD Thesis, Imperial College, 2017.

The Kinchin-Pease Model

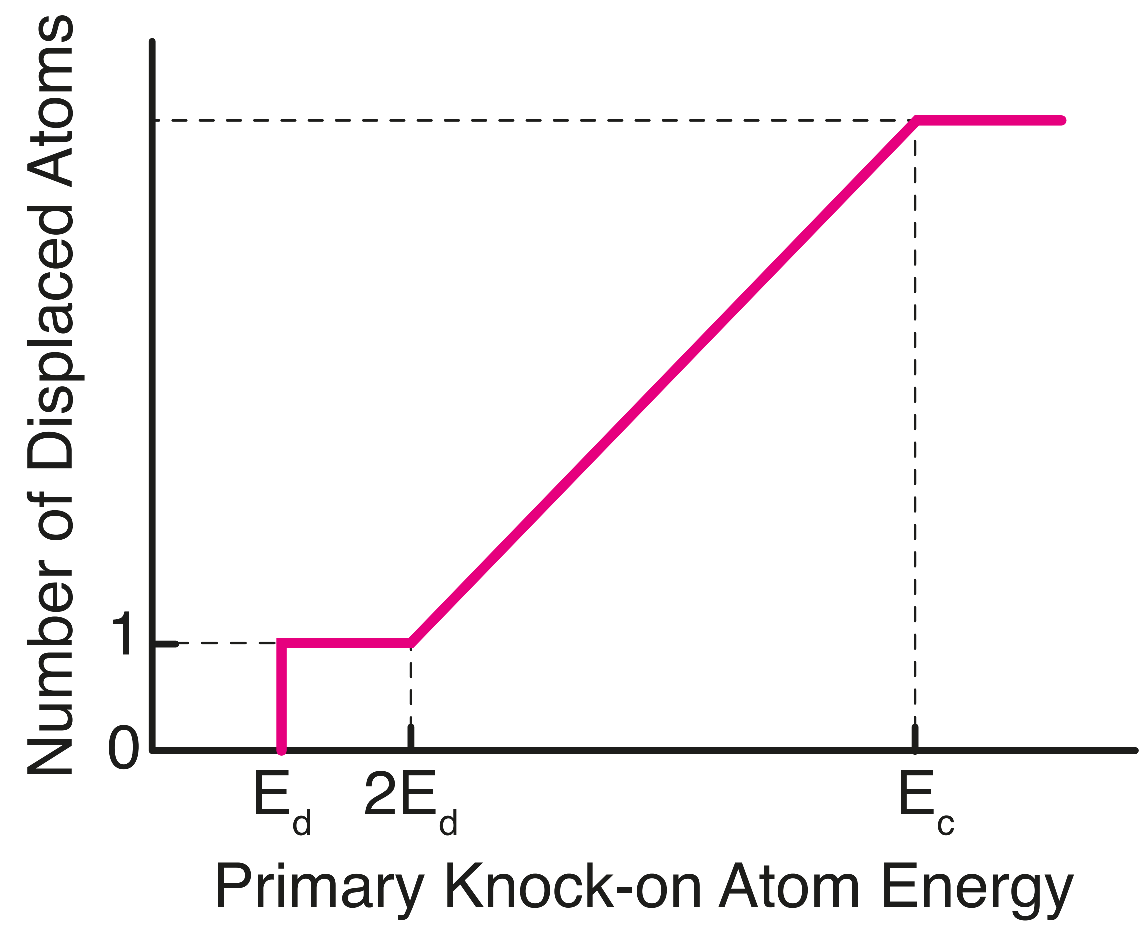
- The Kinchin-Pease Model relates the energy of an incident atom to the number of defects (Frenkel pairs) produced.



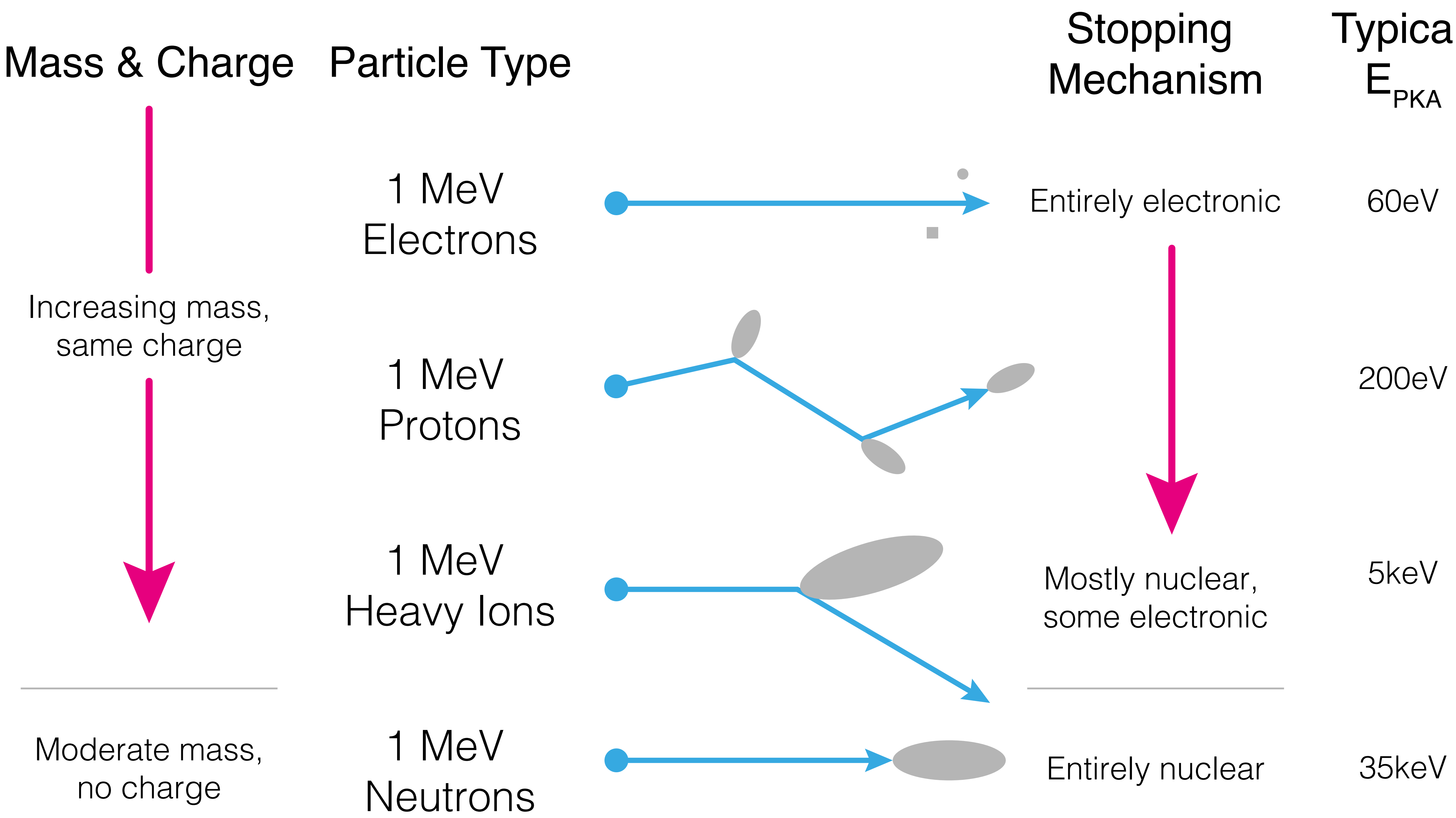
E_d = threshold displacement energy
E_c = cutoff energy

The Kinchin-Pease Model

Energy Range	Description
$E < E_d$	No defect production
$E_d < E < 2E_d$	Single Frenkel Pair
$2E_d < E < E_c$	Defect production proportional to incident energy
$E > E_c$	Defect production stops Electronic stopping

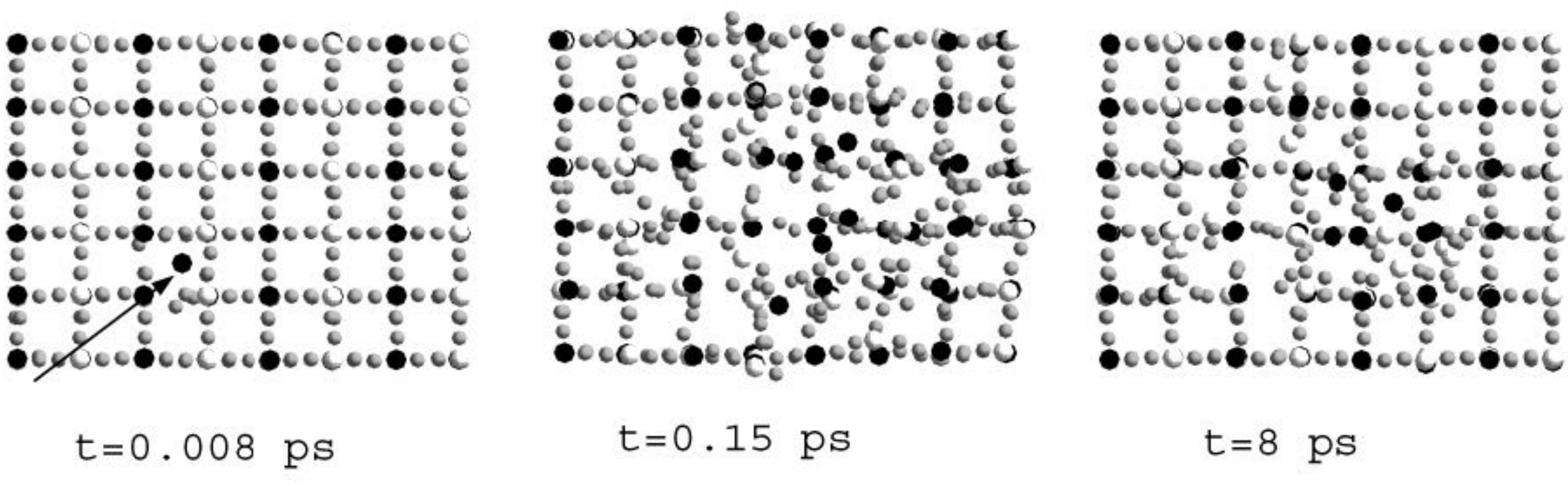


E_d = threshold displacement energy
E_c = cutoff energy



How Does Type of Incident Radiation Affect Damage?

An Example Collision Cascade: Zircon 1keV PKA



1. Ballistic Phase **2. “Themal Spike”** **2. Quench**

View along <001> Kostya O Trachenko et al 2001 *J. Phys.: Condens. Matter* **13** 1947

Recovery

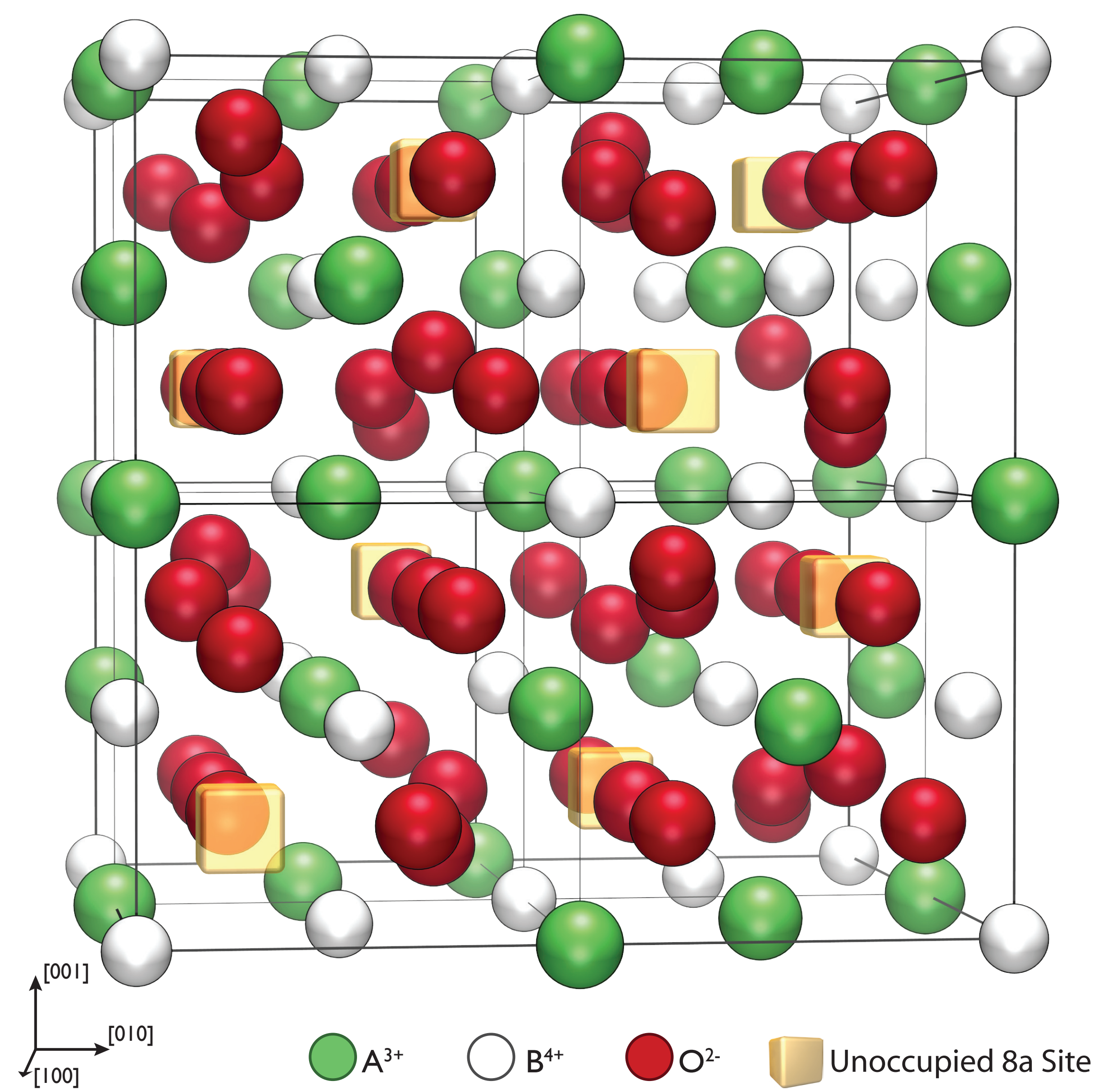
- The previous slide showed that at the core of a damage cascade a significant number of defects are likely to be formed.
- Following the cascade a large number of these defects will recombine (e.g. Frenkel pair recombination).
- To a large degree damage will be annealed away and the lattice will recover.
- The damage retained minutes, days, weeks or years after the damage depends on:
    - the degree of initial damage,
    - defect chemistry: some defects are thermodynamically favourable,
    - kinetics: the ability of the system to reach its low energy condition (e.g. diffusion).

It’s not just about Frenkel pairs

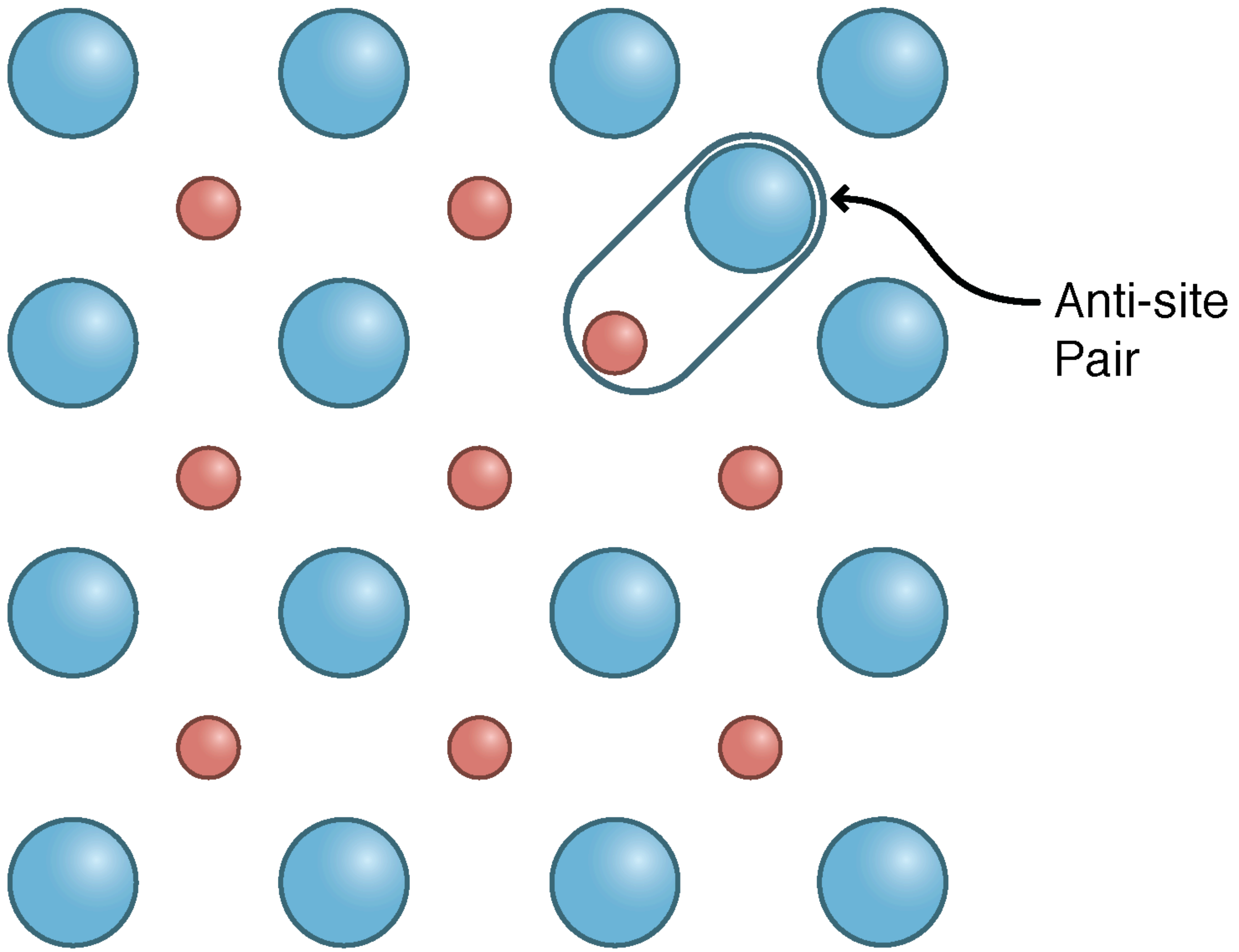
- **Point defects:** missing atoms (vacancies), displaced atoms (interstitials), inappropriate atoms (dopants).
    - May occur as isolated defects or as clusters containing multiple species.
- **Line defects:** dislocations extend through crystal a a line.
    - Dislocation core contains atoms displaced well away from usual sites in crystal.
- **Planar defects:**
    - Grain boundaries
    - Surface
    - Stacking faults, inversion domains and twins.
- **Precipitates, Bubbles:** large clusters of atoms that are too large to be considered as point defects.
- **Electronic Defects:** missing electrons, trapped electrons, excited states

Case Study: Radiation Tolerance in Pyrochlore Oxides

- $A_2B_2O_7$ pyrochlore oxides are being studied as hosts for the disposal of high level nuclear waste.
- As a result they require good tolerance self-irradiation from the nuclides they contain.
- A very wide range of compositions exhibit this structure.
    - A^{3+}: La to Lu,
    - B^{4+}: Ti to Pb.
- How can we narrow this down to a smaller number of materials for further study?

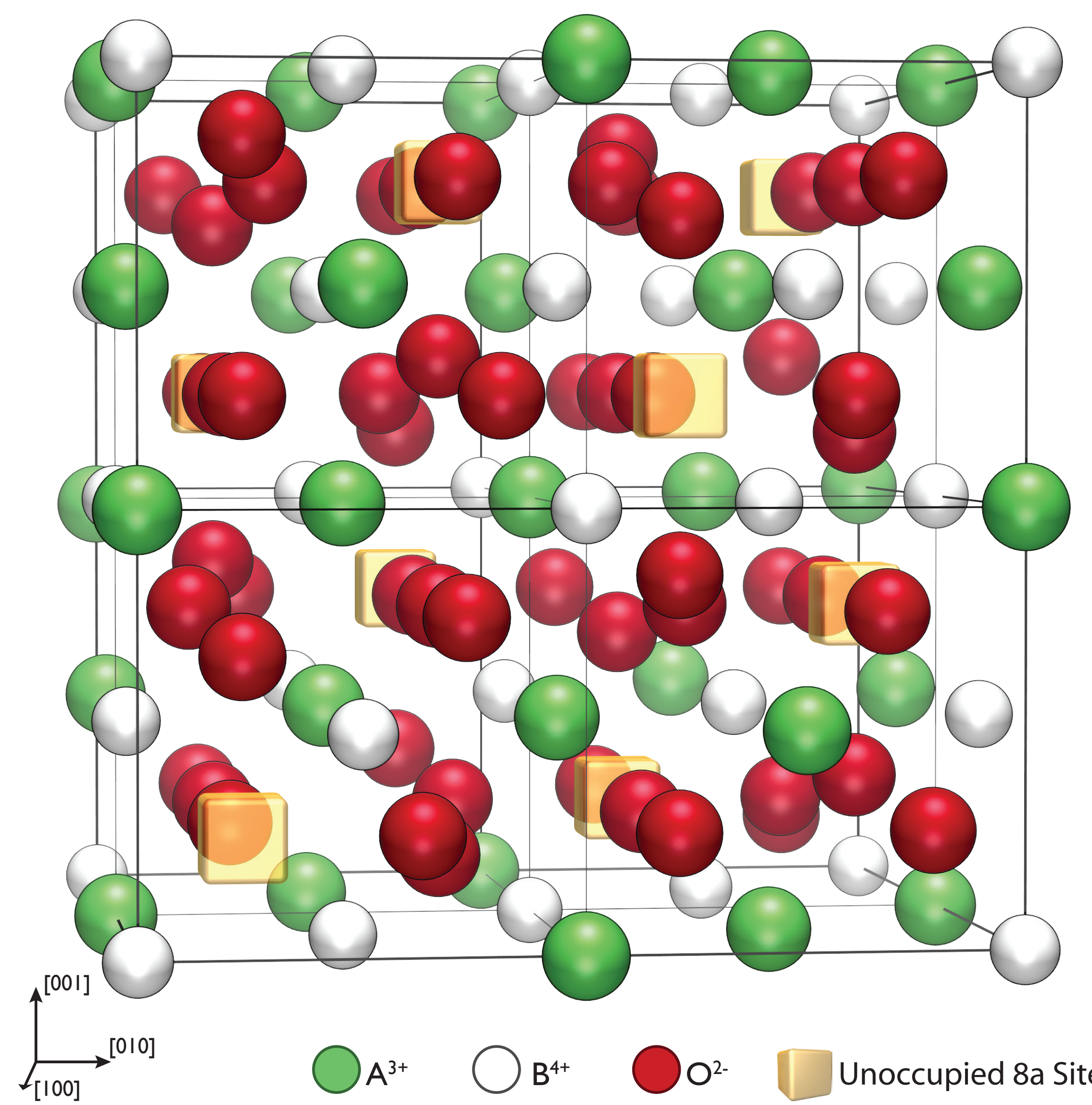


Intrinsic Defect Processes: The Anti-site Reaction



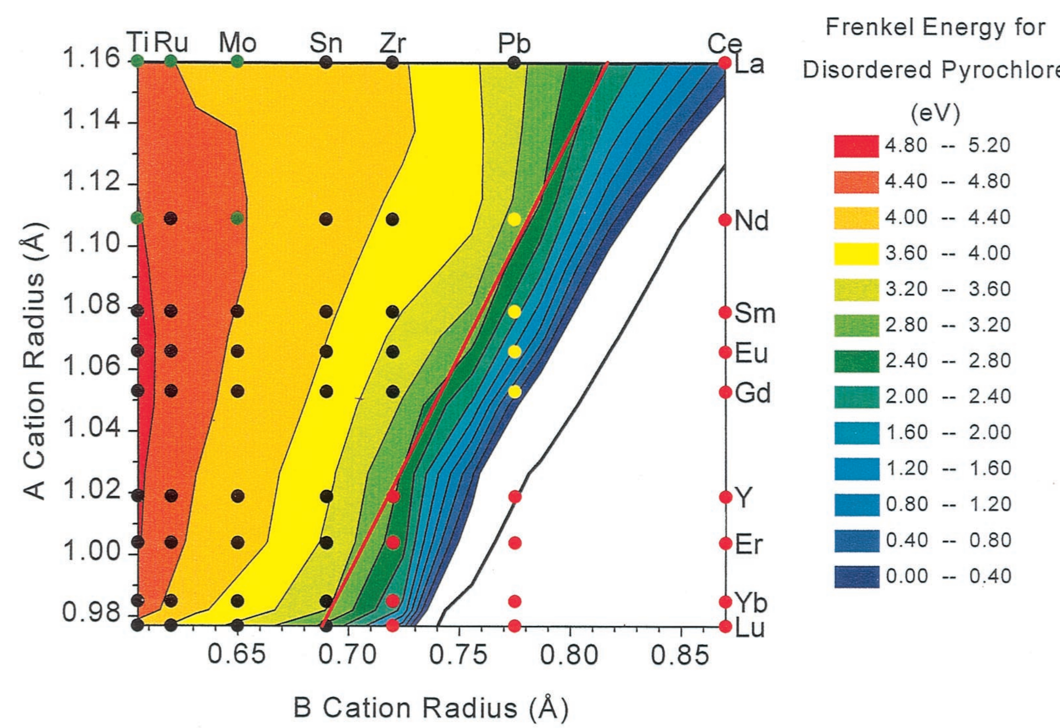
Case Study: Radiation Tolerance in Pyrochlore Oxides

- Radiation tolerance of these materials could be linked to the energy required to incorporate a cluster of defects containing the following into the lattice:
  - A_B, B_A antisite pair.
  - Oxygen Frenkel pair adjacent to antisite



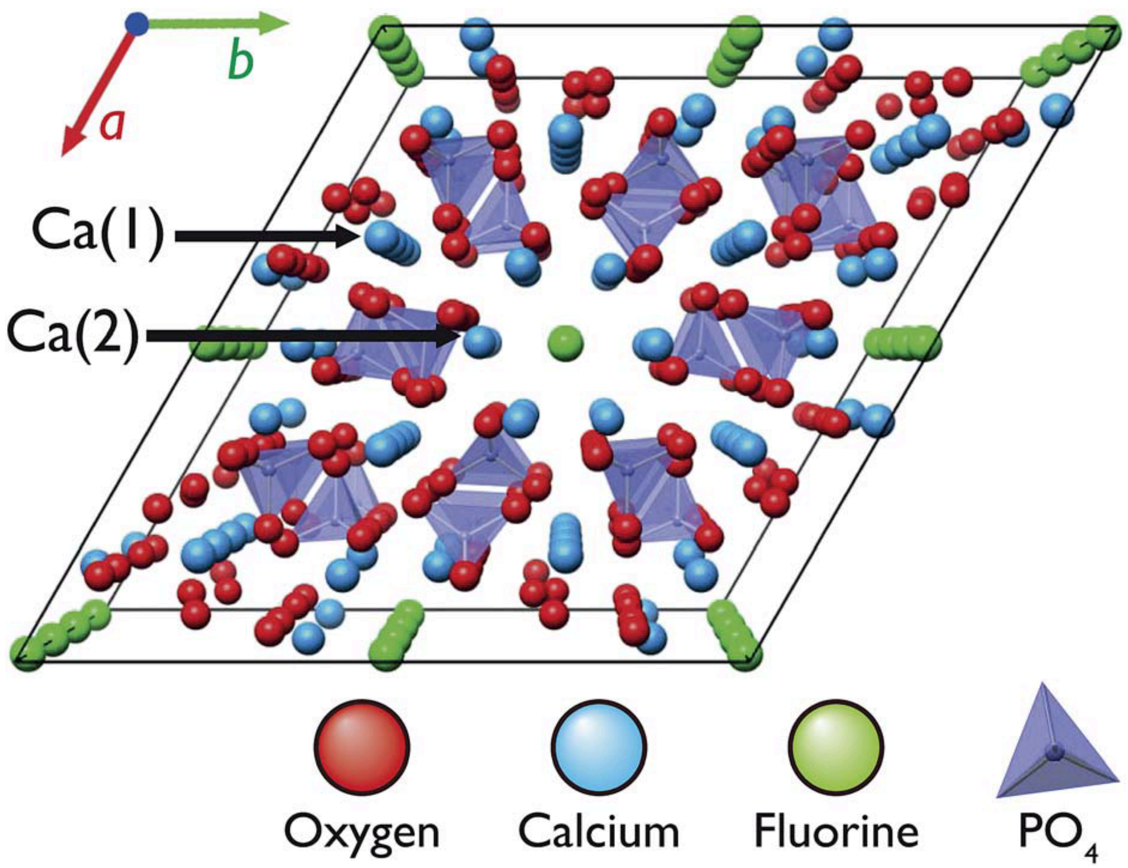
Case Study: Radiation Tolerance in Pyrochlore Oxides

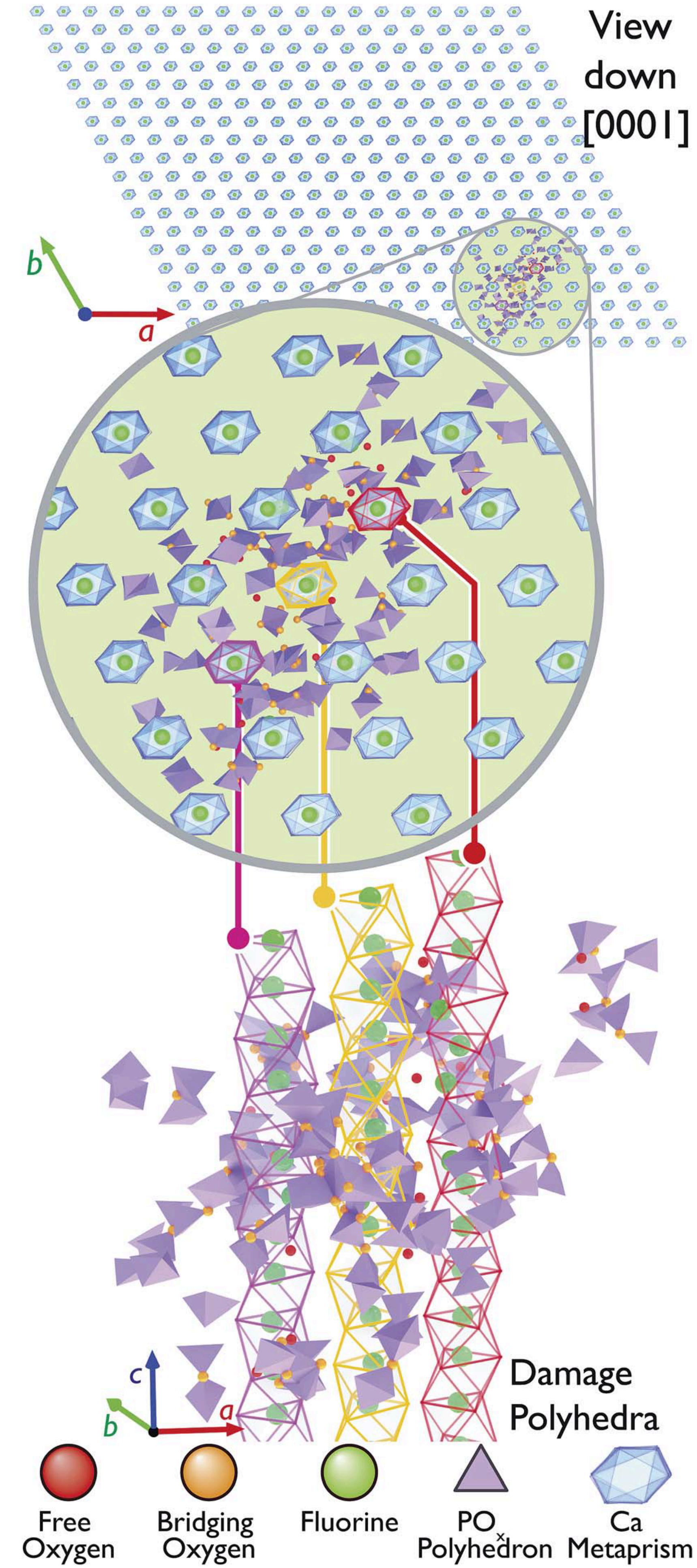
- Computer simulations performed to calculate defect energies used in contour plot to the right.
- Interestingly, compositions exhibiting low defect energies correspond with those which readily transform to a defect fluorite.



Minervini, L., Grimes, R.W., Sickafus, K.E.: Disorder in Pyrochlore Oxides. *J. Am. Ceram. Soc.* **83**, 1873–1878 (2004).

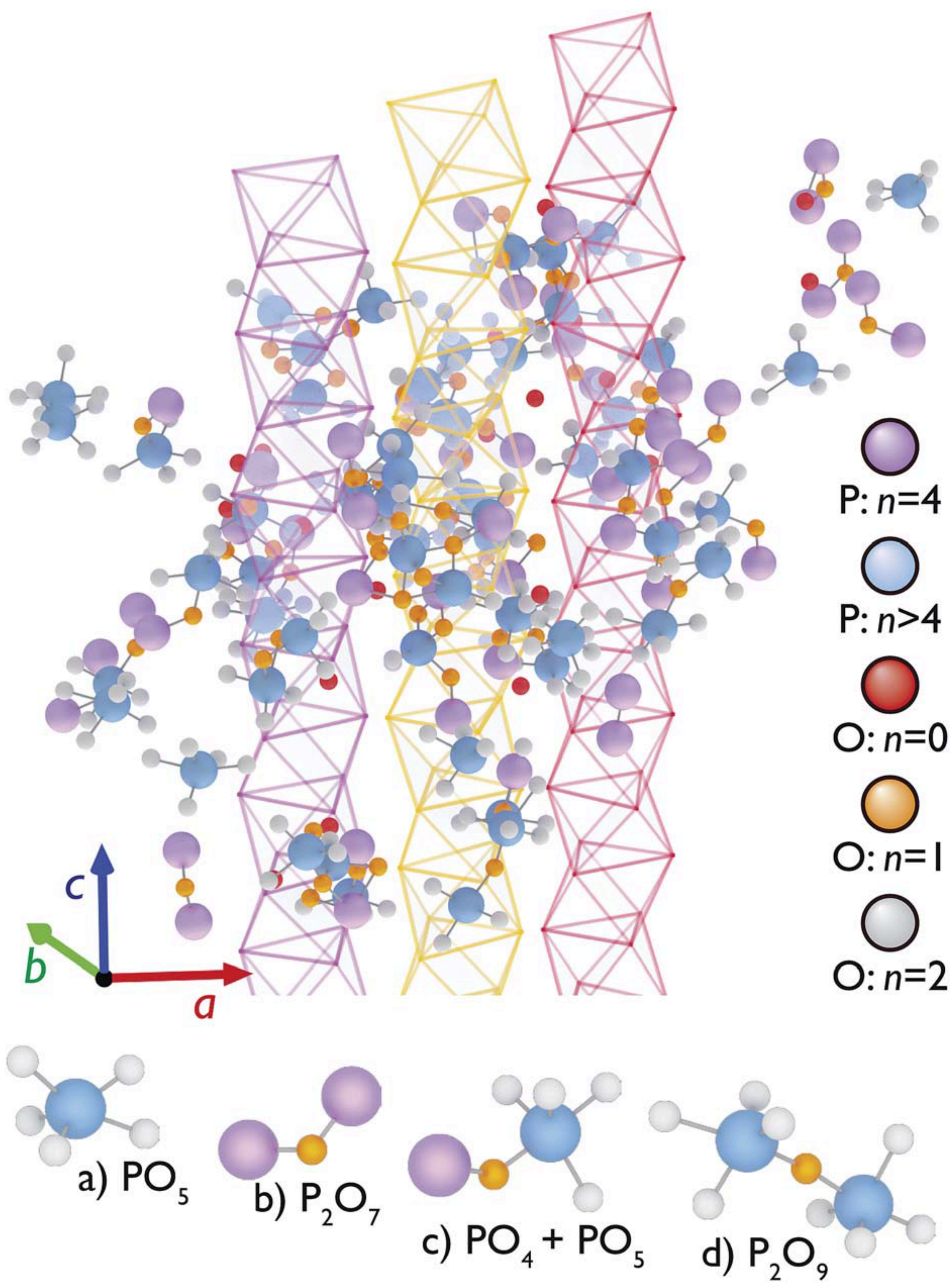
Case Study: Fluorapatite

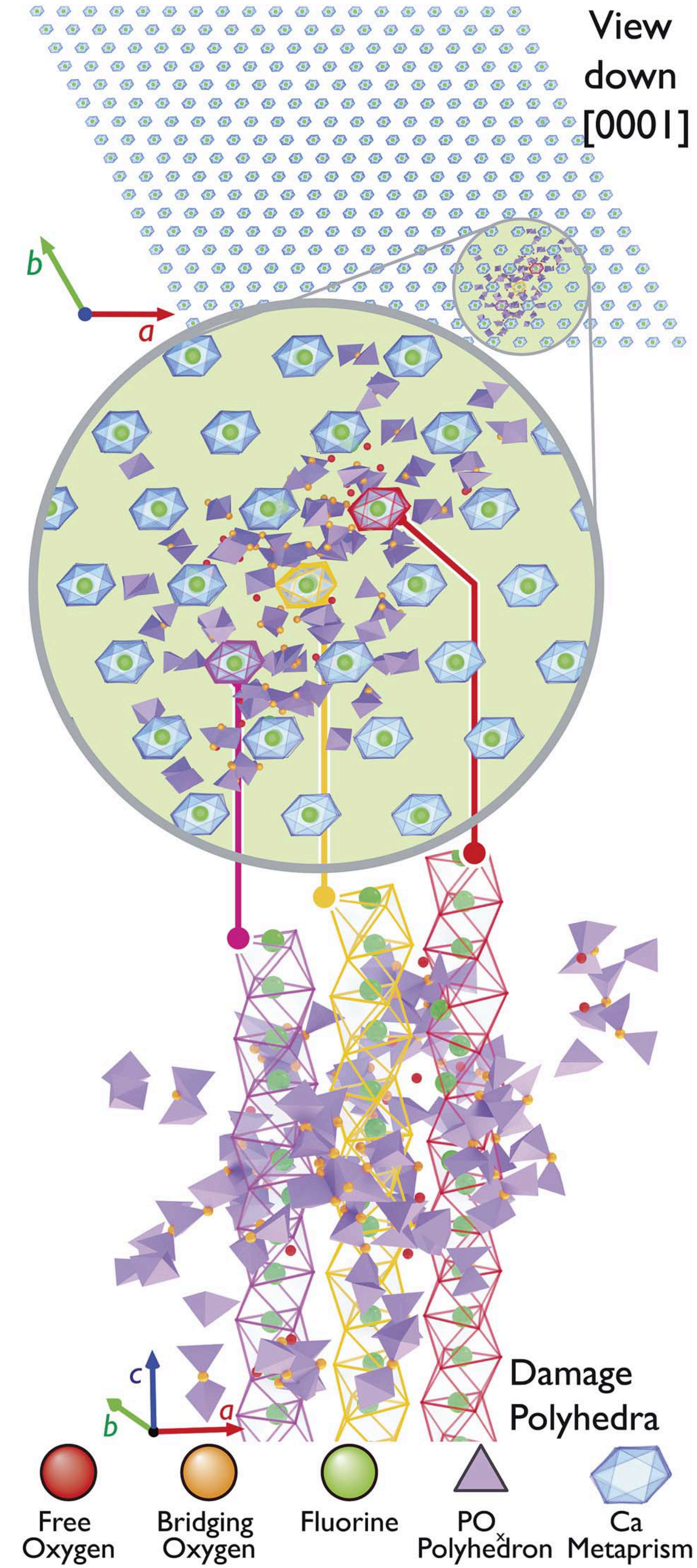




Jay, E.E., Fossati, P.M., Rushton, M.J.D., Grimes, R.W.: Prediction and Characterisation of Radiation Damage in Fluorapatite. *J. Mater. Chem. A.* ***3*** (2014) 1164.

Case Study: Fluorapatite





Recovery and Defect Sinks

- Radiation induced defects can interact with existing lattice defects.
- Some of these can act as “sinks” for this damage aiding recovery.
- Research is ongoing into “defect engineering” strategies to deliberately seed a material with sink sites and improve radiation tolerance.

Conclusions

- Radiation damage can lead to large changes in material properties.
- It must be considered when designing materials for nuclear applications if their long term behaviour is to be predicted.
- Modelling and experiment are highly complementary.
- There is still plenty of research to be done incorporating more detail into models (microstructural, chemical, electronic).

Recommended Reading

- J.F. Ziegler, J.P. Biersack, M.D. Ziegler, “SRIM Textbook”.
- G.S. Was, “Fundamentals of Radiation Materials Science”, Springer (2010).
- S.E. Donnelly, J.H. Evans (eds), “Fundamental Aspects of Inert Gases in Solids”, Nato Science Series B vol 279, Springer (1991).
- W. J. Weber, R. C. Ewing, C. R. A. Catlow, T. D. de la Rubia, et al. “Radiation effects in crystalline ceramics for the immobilization of high-level nuclear waste and plutonium”, *J. Mater. Res.* **13** (1998) 1434.