Ion-Irradiation-Induced Defects in Solids

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Outline: Ion-Irradiation-Induced Defects in Solids

- Sources of ion irradiation
- Ion stopping: nuclear stopping, displacements and defect formation
- Microstructural control of defect retention
  - nanolayered materials
  - amorphous materials
- Summary

Sources of Ion Irradiation

- Ion Accelerators
- Nuclear Environments (fusion, fission)

Ion Implantation System with Mass Separation

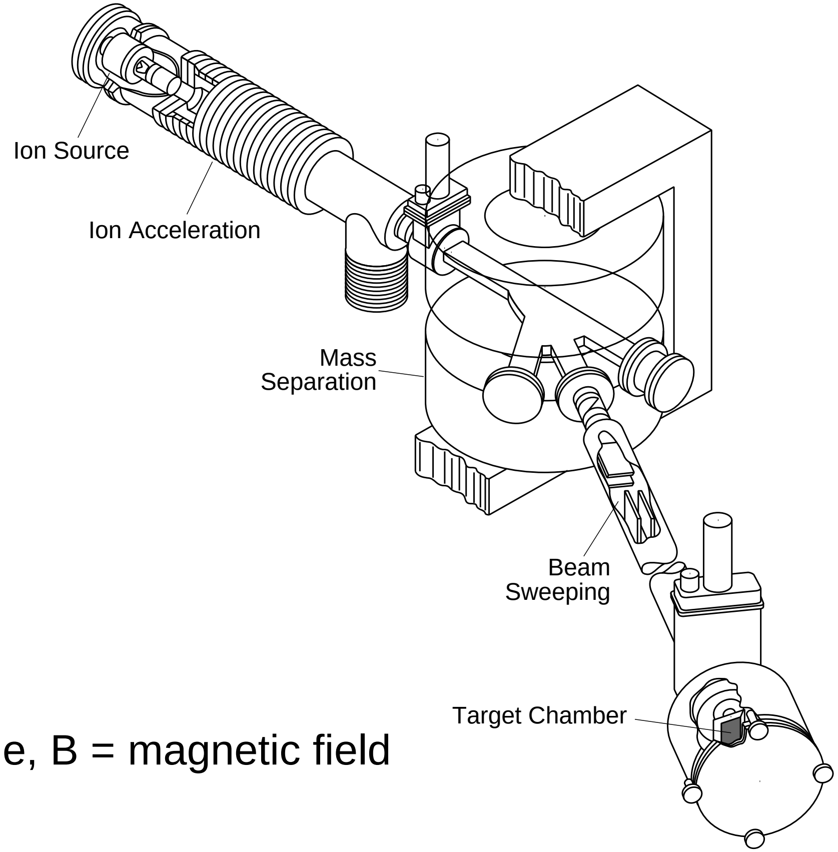
Schematic drawing of an ion implantation system. A mass-separating magnet is used to select the ion species (elements and isotopes) of interest. Beam-sweeping facilities are required for large-area uniform implantations

Magnetostatic field can not change the kinetic energy (K.E.) of the particle, only change the direction of its velocity.

K.E. = $0.5\ mv^2 = eV$

The radius (R) of the circular path is proportional to the velocity of the particle.

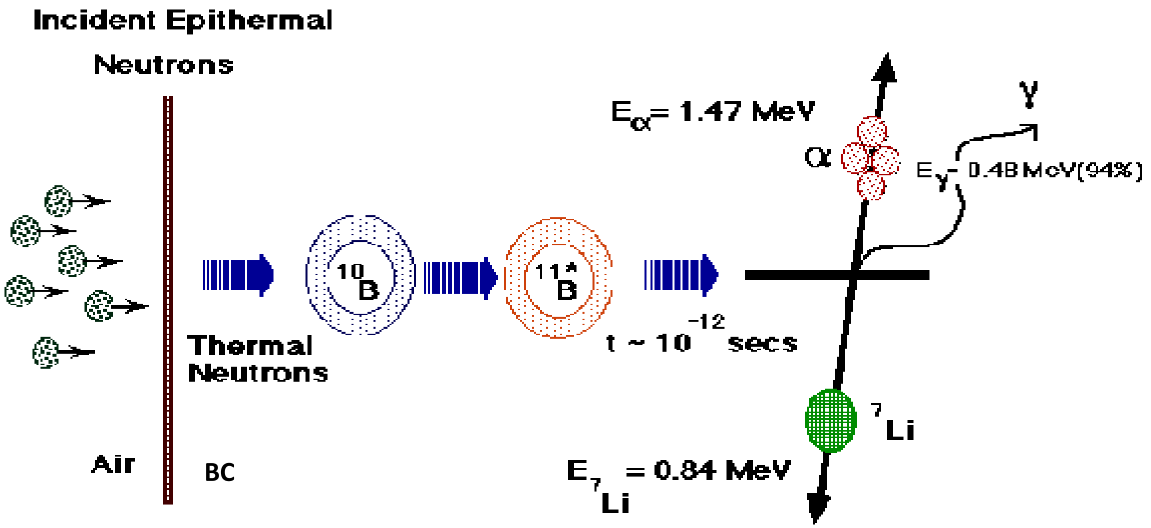
$R = m\ v\ /\ q\ B$



m = ion mass, v = ion's velocity, q = charge, B = magnetic field

Neutron Sources of Ion Irradiation

^{10}B neutron capture and boron disintegration



$^{10}B + n \rightarrow {}^{7}Li$ (0.84 MeV) + ^{4}He (1.47 MeV) + γ (0.48 MeV) (94%)
$^{10}B + n \rightarrow {}^{7}Li$ (0.84 MeV) + ^{4}He (1.78 MeV) (6%)

The energetic He and Li ions give rise to radiation damage

This damage process can be simulated with energetic ions produced by an ion accelerator

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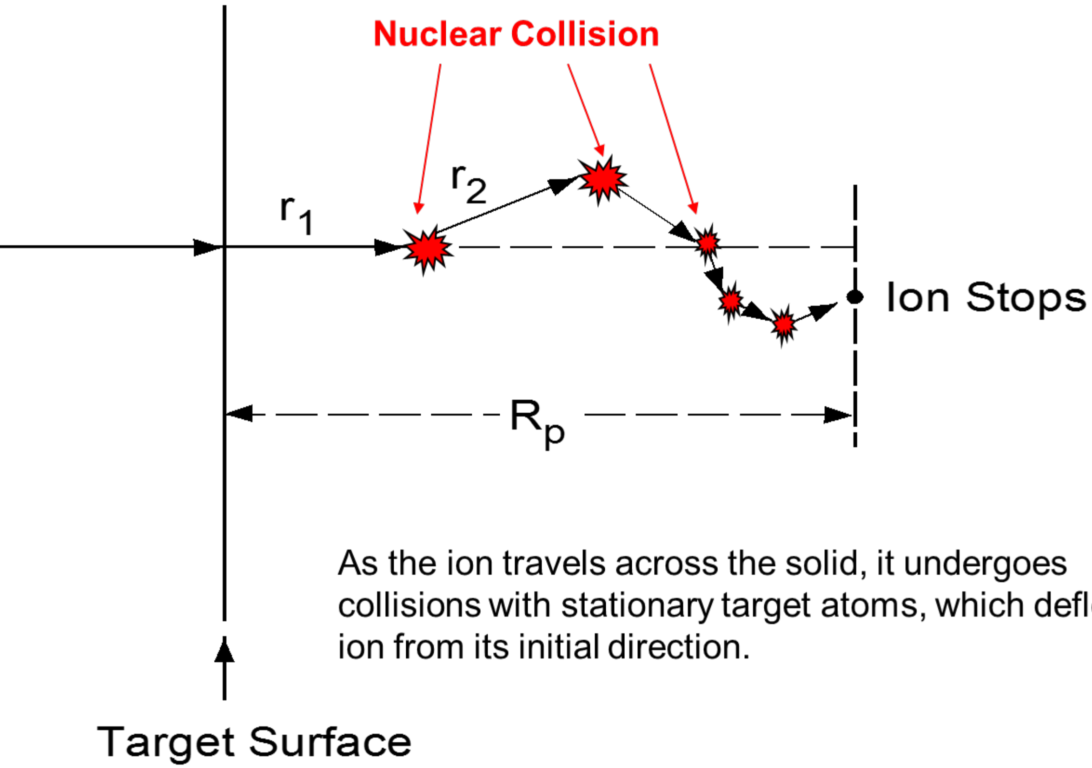
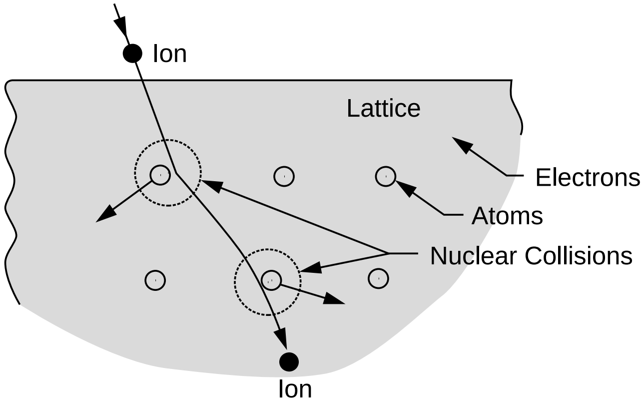
Ion Stopping: The Process of Slowing Down the Ion



As the ion travels across the solid, it undergoes collisions with stationary target atoms, which deflect the ion from its initial direction.

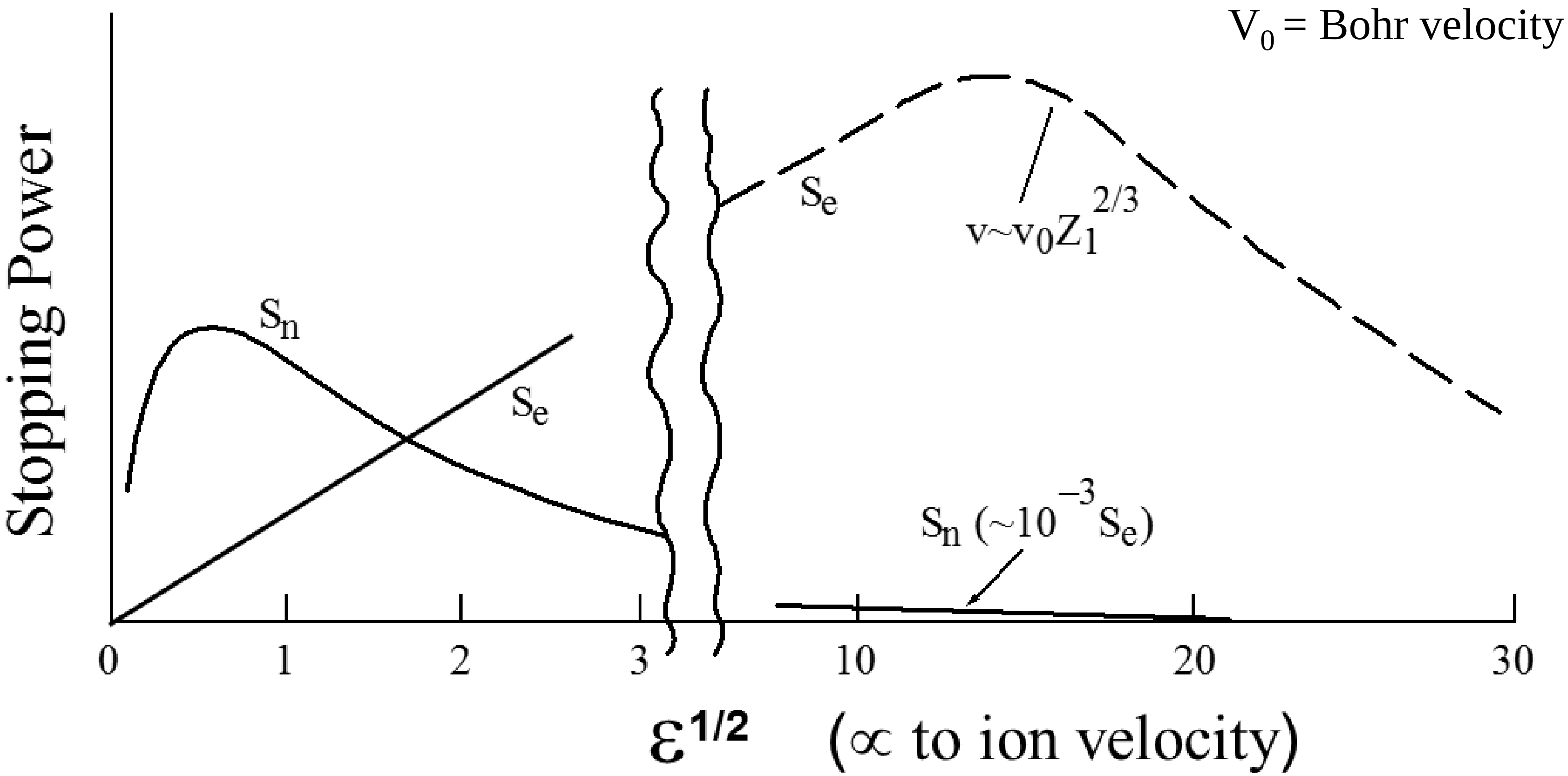
The passage of an energetic ion in a solid during an ion implantation. As the ion travels across the solid, it undergoes collisions with stationary target atoms, which deflect the ion from its initial direction (**nuclear stopping**).

The ion also collides with electrons in the solid and loses energy in these collisions (**electronic stopping**).



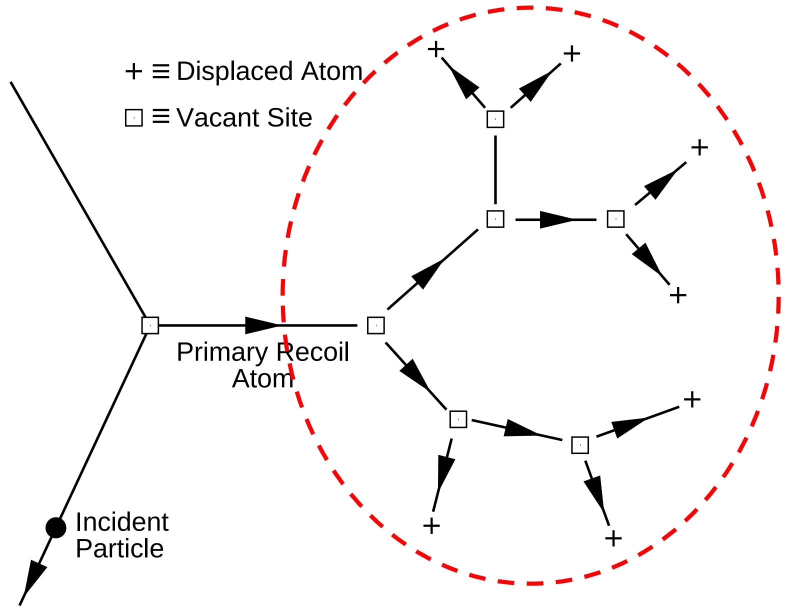
The major changes in the ion's flight direction are due to the ion's collision with individual lattice atoms (nuclear collisions).

Energy loss: Ion Stopping

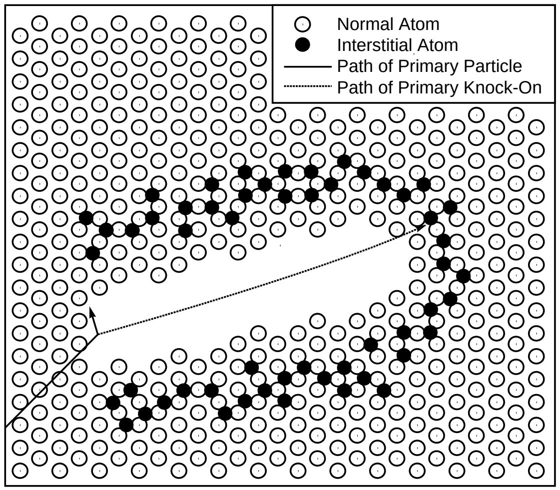


Nuclear stopping dominates as the ion slows down

Nuclear Stopping, Displacements and Defect Formation



Collisions between ions and target atoms result in the slowing down of the ion. In these collisions, sufficient energy may be transferred from the ion to displace an atom from its original site



Atoms that are displaced by incident ions are called ***primary knock-on atoms*** or **PKAs**.

The PKAs can in turn displace other atoms, i.e., secondary knock-on atoms, tertiary knock-ons, etc., thus creating a ***cascade of atomic collisions***.

This leads to a distribution of defects, vacancies, interstitial atoms and other types of lattice disorder, in the region around the ion track.

Damage Production and dpa

A commonly used measure of irradiation damage is ***displacements per atom*** (dpa).

dpa = Number of displacements per unit volume/atomic density

A unit of 1 dpa means that, on average, every atom in the irradiated volume has been displaced from its equilibrium lattice site one time.

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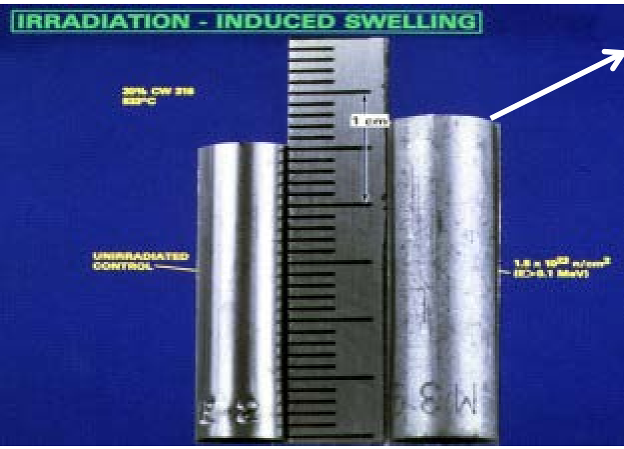
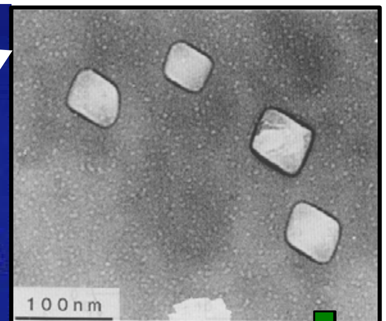
Microstructural Control of Defect Retention

- Why are Defects a Problem?
- How to Reduce or Avoid Defect Formation?

Why are defects a problem?

Crystalline structural materials are prone to radiation damage: *void swelling and embrittlement*

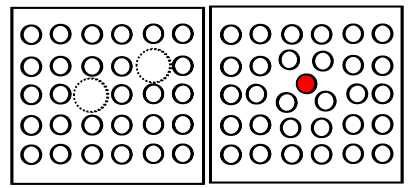




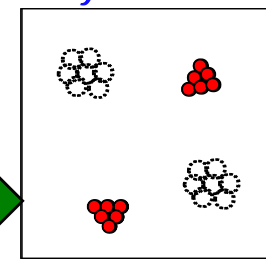
Atomic defects produced by irradiation

Vacancies

Interstitials



Defects that do NOT recombine aggregate into vacancy or interstitial clusters



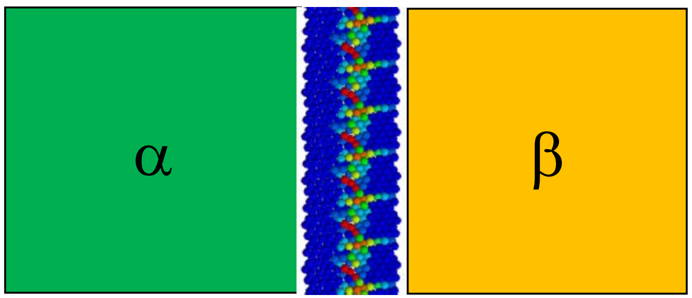
Embrittlement

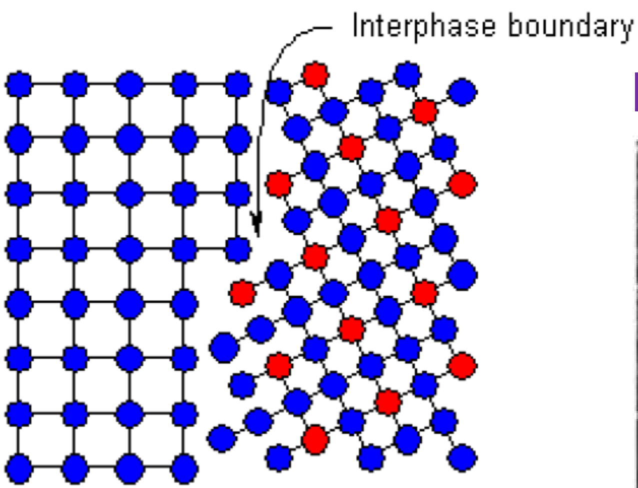
D.L. Porter and F. A. Garner, J. Nuclear Materials, **159**, (1988) 114
D.J. Bacon and Y.N. Osetsky, Int. Mater. Rev., **47**, (2002). 233
H. Trinkaus and B.N. Singh, J. Nuclear Materials, **323**, (2003) 229 .

How to Reduce or Avoid Defect Formation?

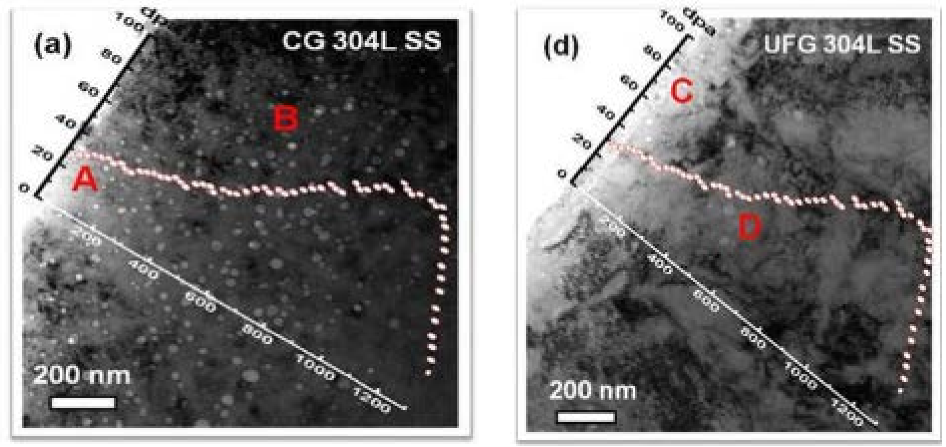
Radiation Tolerance: Approach to Finding the Solution in Crystalline Materials

Defect sinks

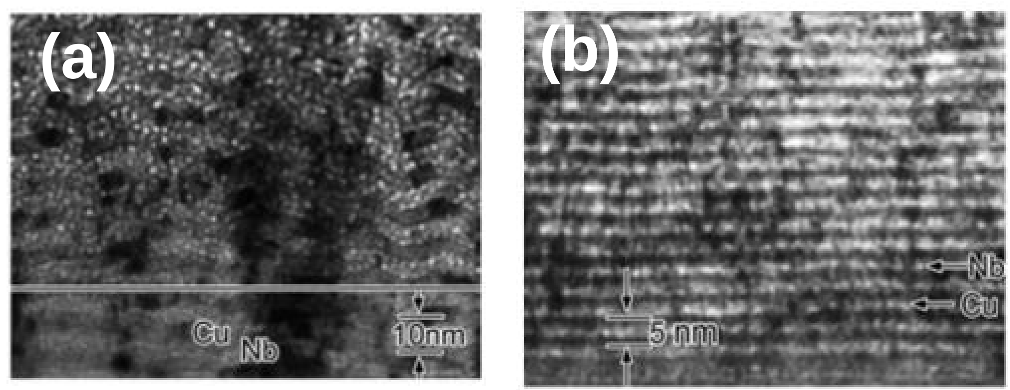




Grain boundary



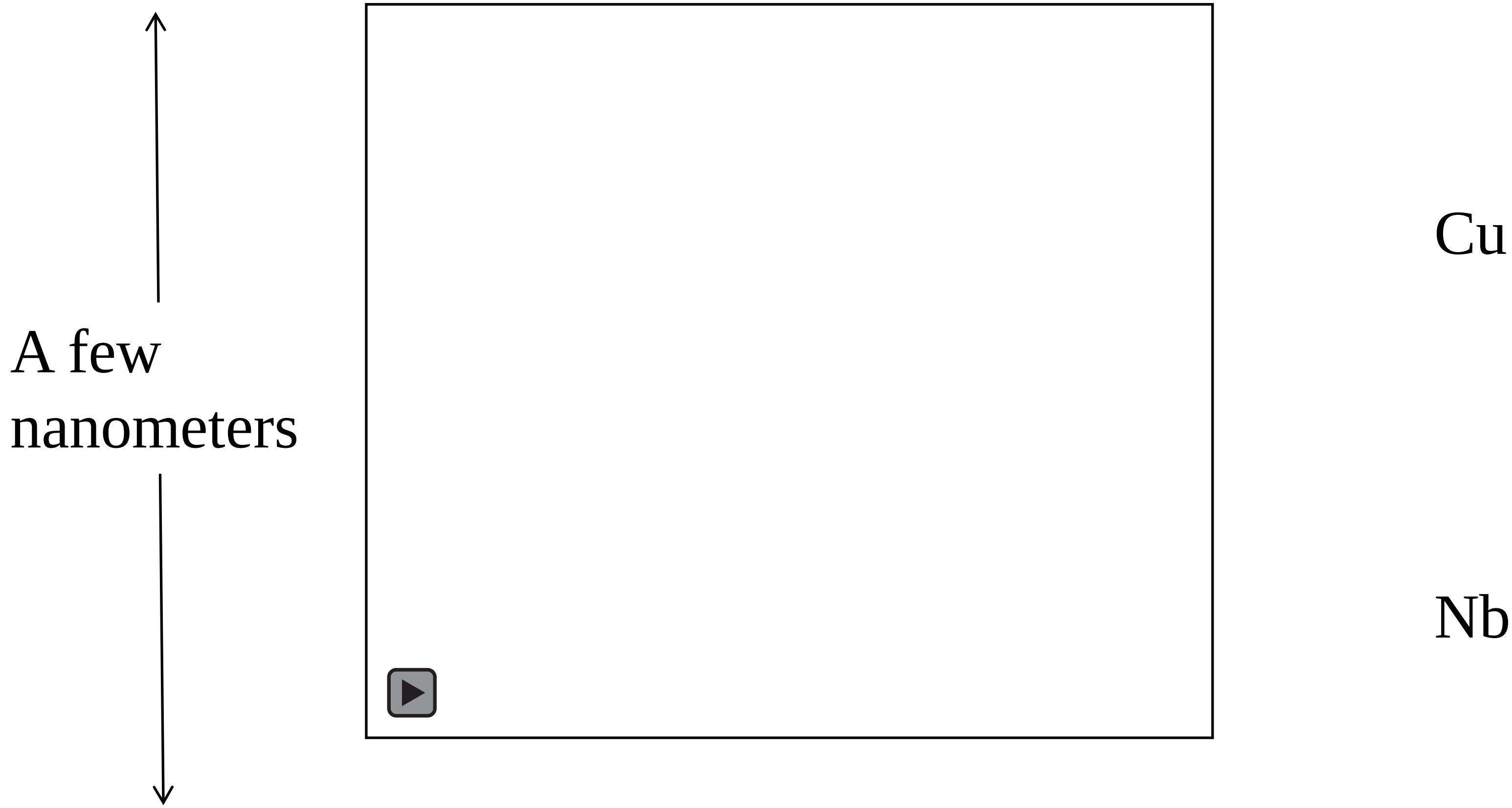
Incoherent interface



1. A. Misra, *et al*., JOM, 59 (2007) 62.
2. 2. C. Sun *et al*., Sci. Rep., 5, (2014) 7801.

Grain and interphase boundaries are known to be defect sinks

B.N. Singh, J. Nucl. Mater., **46** (1973) 99; Phil. Mag. 28 (1973) 1409.
B.N. Singh, S.J. Zinkle, J. Nucl. Mater., (1993).



M.J. Demkowicz, R.G. Hoagland, J.P. Hirth,
Physical Review Letters, **100**, 136102 (2008).

Lattice misfit gives rise to multiple interface atomic configurations

Each in the Kurdjumov-Sachs orientation relation with nearly degenerate energies

KS_1

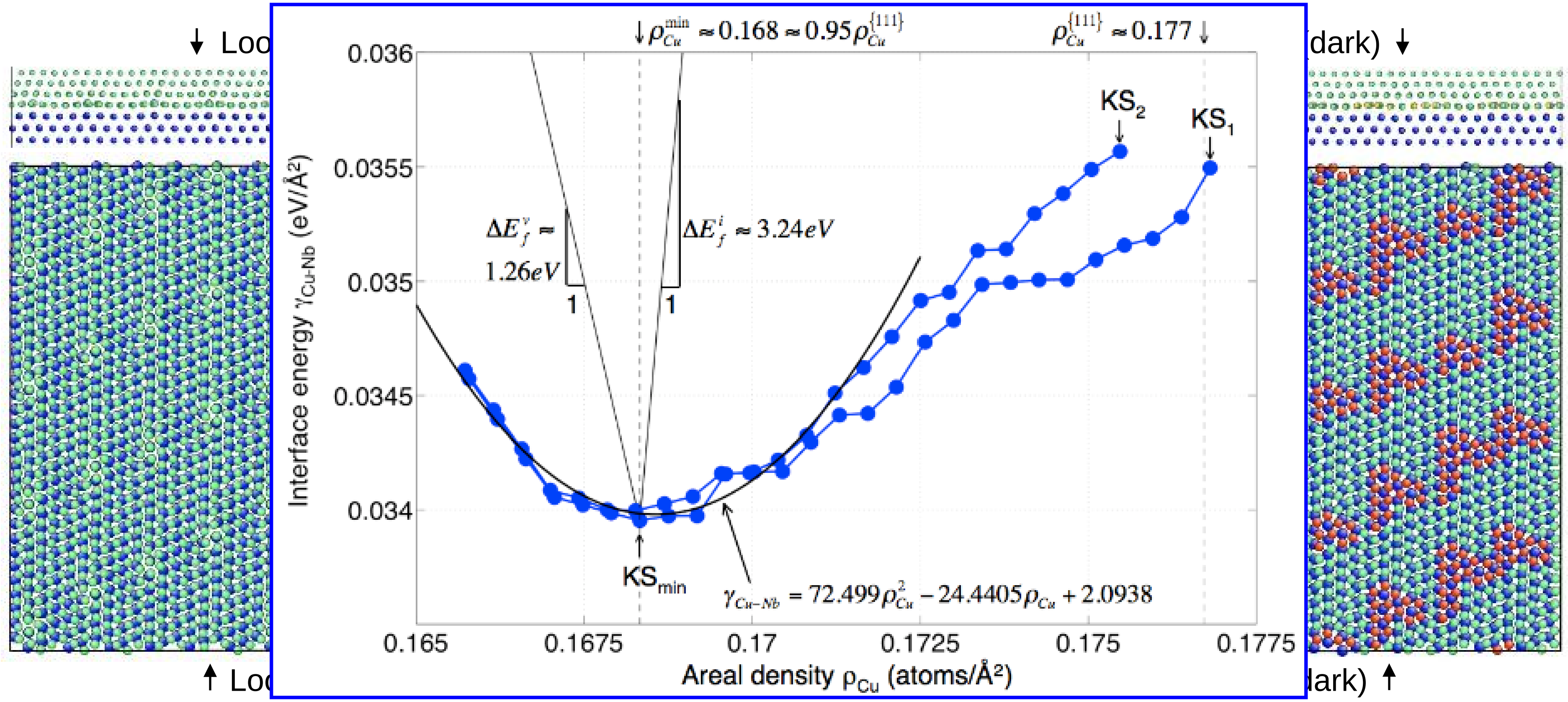
- Created by simply joining Cu and Nb in the KS OR
- Quasiperiodic pattern of low coordination sites

KS_2

- Interfacial Cu atom layer homogeneously strained with respect to Cu (111)
- No low coordination sites

KS_{min}

- Contains a 5% atomic vacancy concentration
- Lowest energy configuration at T=0K



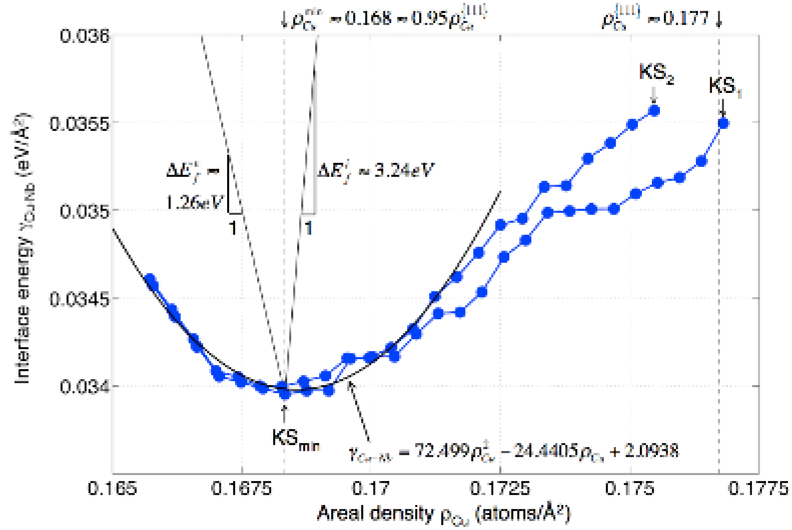
Consequences of lattice misfit

- Low defect formation energy

Effective formation energy for a concentration "c" of defects in KS_{min}:

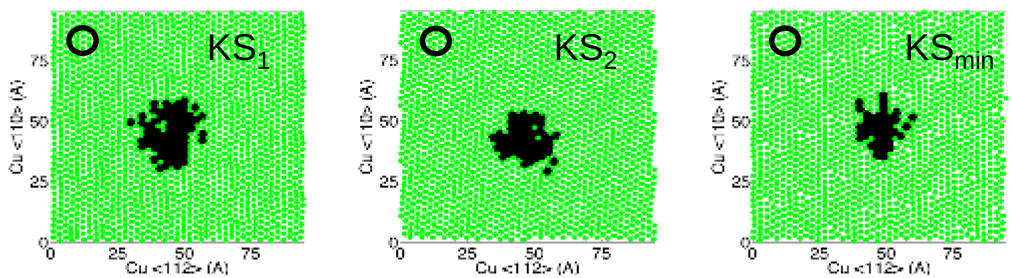
$$\Delta E_{eff} = \frac{\Delta\gamma_{Cu-Nb}}{|\Delta\rho_{Cu}|} = \frac{\gamma_{Cu-Nb}\left((1+c)\rho_{Cu}^{min}\right) - \gamma_{Cu-Nb}\left(\rho_{Cu}^{min}\right)}{c\rho_{Cu}^{min}} \approx 12|c|\left[\frac{eV}{defect}\right]$$

For example, if c=0.01 then **$\Delta E_{eff} \approx 0.12$ eV/defect**



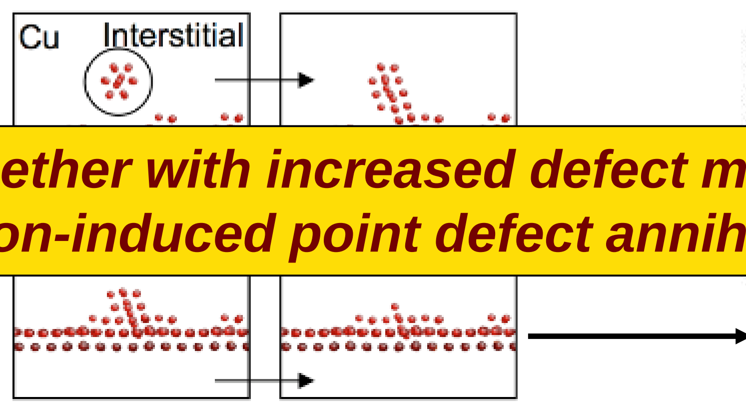
- Large defect core sizes

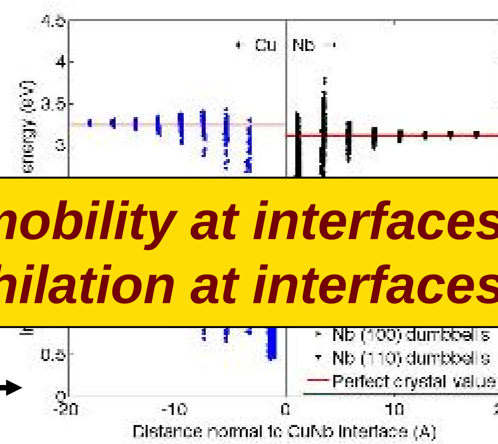
Effective interfacial defect recombination radius ≥ 0.75nm



- Athermal migration of defects to interfaces

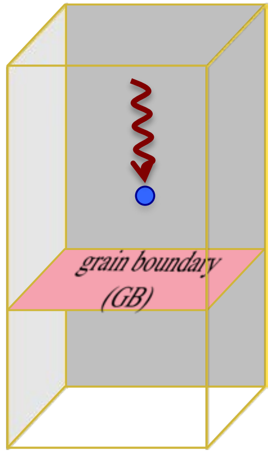
Energy barriers for point defect migration



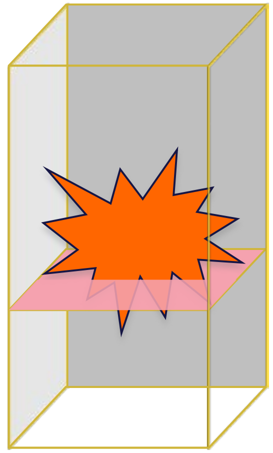


These properties, together with increased defect mobility at interfaces, favor radiation-induced point defect annihilation at interfaces.

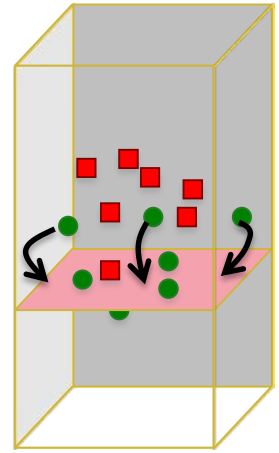
Recovery of vacancies and interstitials at grain boundaries



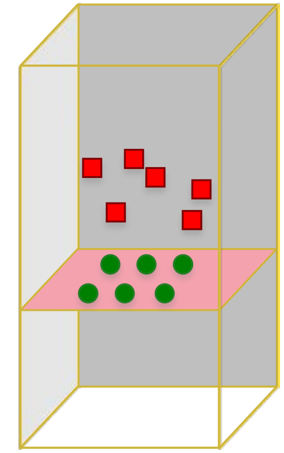
An energetic particle, such as a neutron, hits an atom ● in the material, giving it a large amount of kinetic energy.



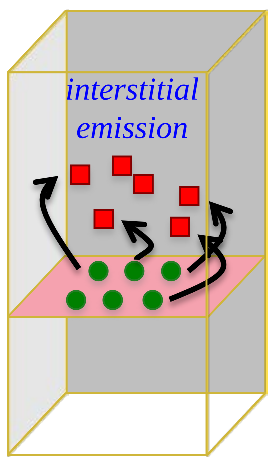
This atom displaces many other atoms in its path, creating a collision cascade, which overlaps with the grain boundary (GB).



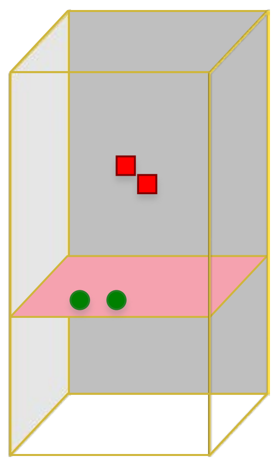
After the cascade settles, point defects -- interstitials ● and vacancies ■ -- remain. The interstitials quickly diffuse to the GB.



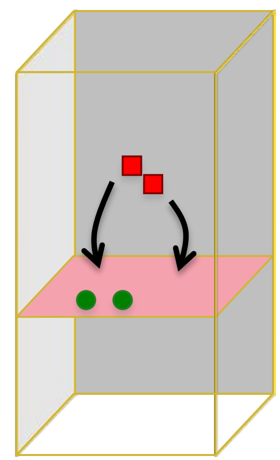
At this point, vacancies remain in the bulk and interstitials are trapped at the GB.



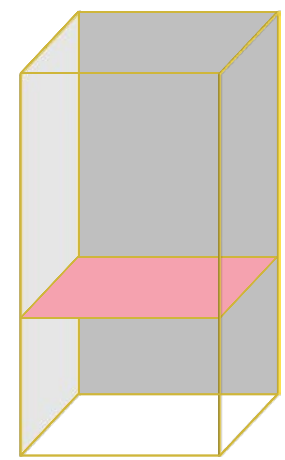
Surprisingly, these trapped interstitials can re-emit from the GB into the bulk, annihilating the vacancies on time scales much faster than vacancy diffusion.



After the interstitial emission events have occurred, some vacancies that were out of reach persist. The system is now in a relatively static situation.



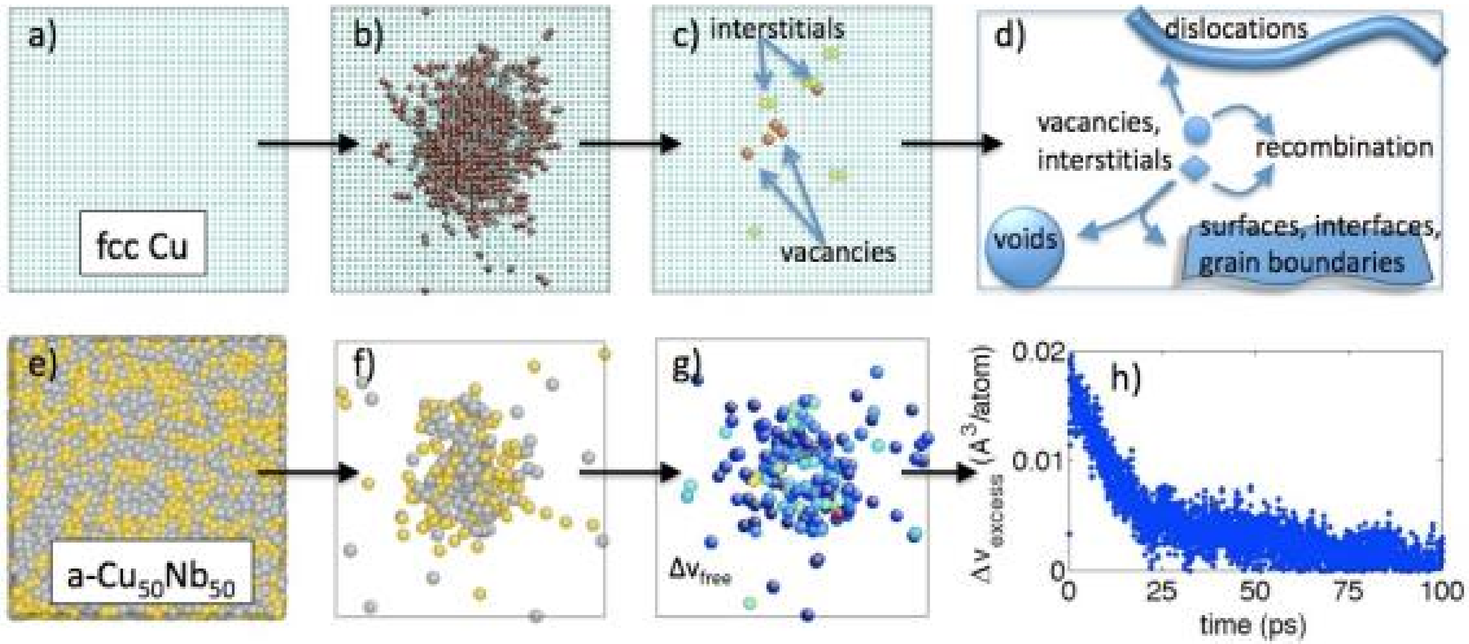
On much longer time scales, the remaining vacancies can diffuse to the GB, completing the healing of the material. At low temperatures this diffusion is exceedingly slow.



In the ideal case, the system returns to a pristine GB. At low temperatures, the only hope for reaching such a state is via the newly discovered interstitial emission mechanism.

X.-M. Bai, A. F. Voter, R. G. Hoagland, M. Nastasi, and B. P. Uberuaga, *Science* ***327,*** 1631 (2010).

Amorphous Materials: Eliminate the Root Source of Defects



Questions:

- **How do amorphous materials respond to radiation damage?**

R. E. Baumer *et al*., Materials Research Letters, 2, (2014) 221.

Proposed amorphous material: SiOC

WHY?

This class of amorphous ceramics has been shown to have crystallization temperatures in excess of 1300 ^{o}C and good oxidation and creep resistance

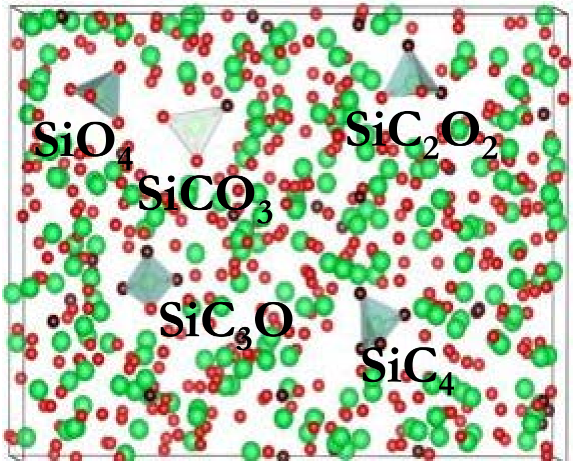
Even higher temperature crystallization temperatures with silicoboron carbonitride ceramic, stable to 2,000 ^{o}C!

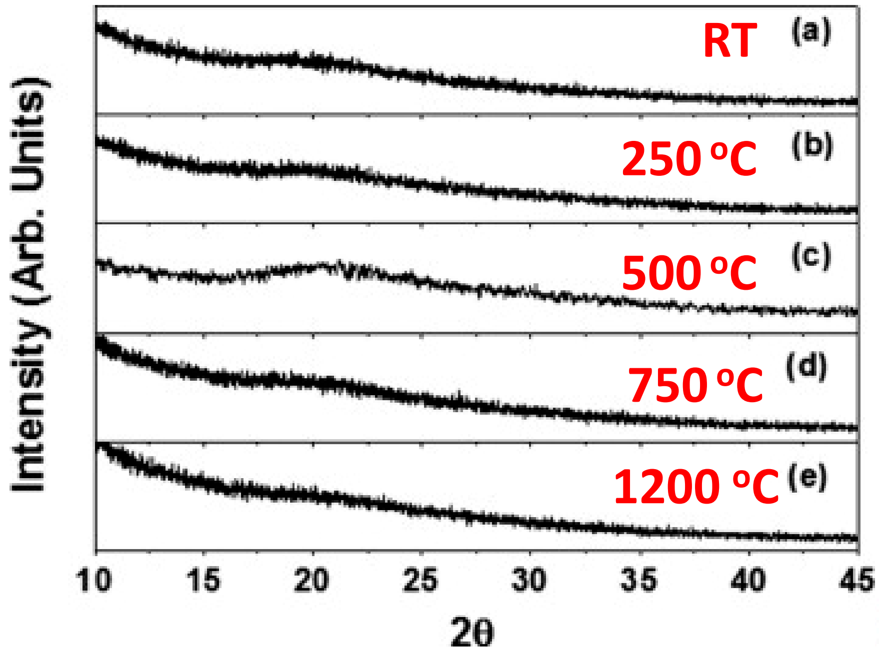
R. Riedel, H-J. Kleebe, V. Schonfelder and F. Aldinger. Nature, 374, pp. 526–528, (1995).

Hypothesis: High crystallization temperature amorphous alloys will be stable under irradiation at elevated temperatures

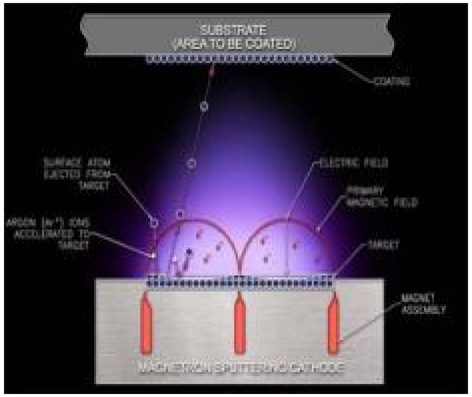
Thermal stability of amorphous SiOC

SiO_xC_{4-x} *(x=0,1,2, 3,4)*





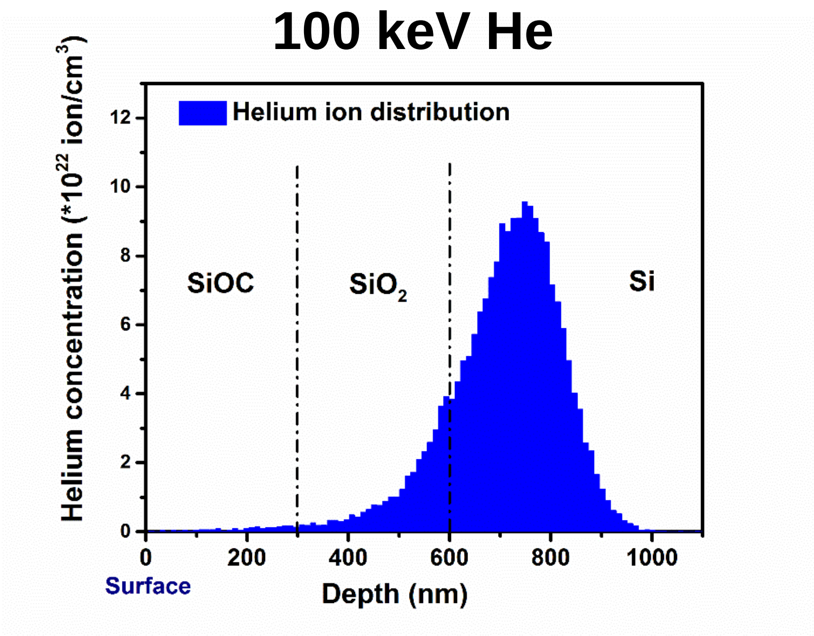
RF Sputtering

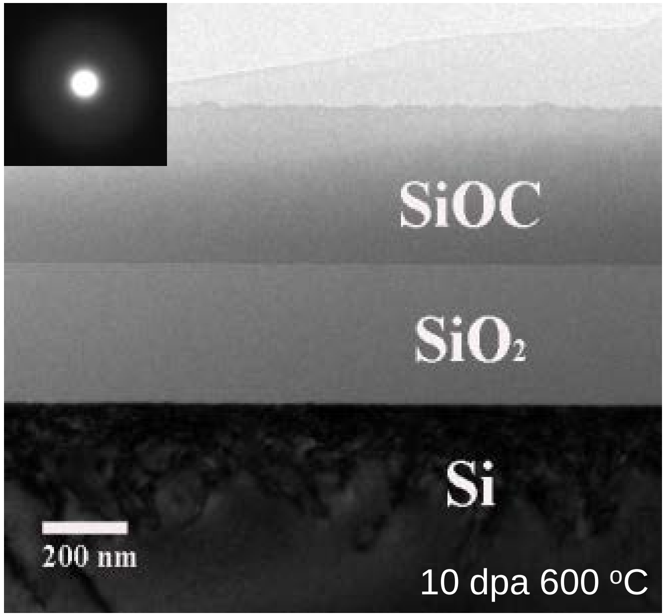


Amorphous SiOC is stable >1200 °C.

J.A. Colón Santana, *et al.*, Nucl. Inst. Methods Phys. Res. B, 350 (2015) 6.
H. Ding, *et al.*, Scientific Report, 5 (2015) 13051.

Ion irradiation of SiOC



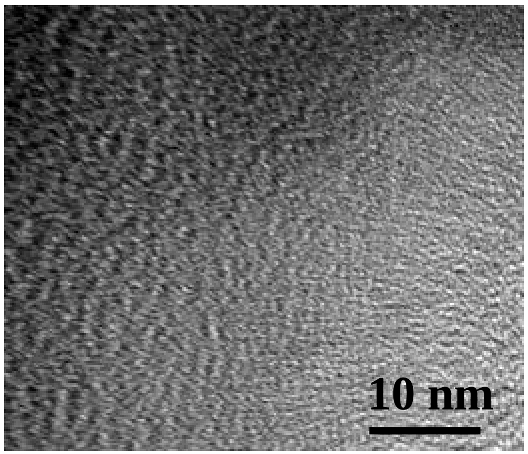


(Little to no helium implantation)

M. Nastasi, Q. Su, *et al.*, J. Nucl. Mater., 461 (2015) 200.

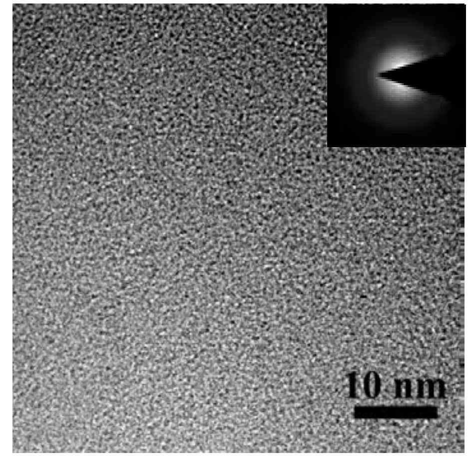
Irradiation Stability of Amorphous SiOC

As-prepared



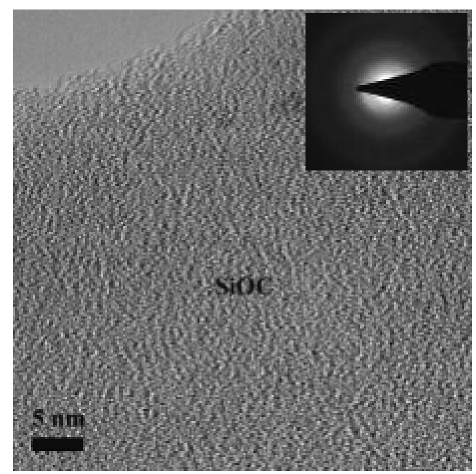
600 ^oC irradiation

100 keV He, 20 dpa



300 ^oC irradiation

1 MeV Kr, 5 dpa



SiOC remains amorphous, no void formation or element segregation!!!

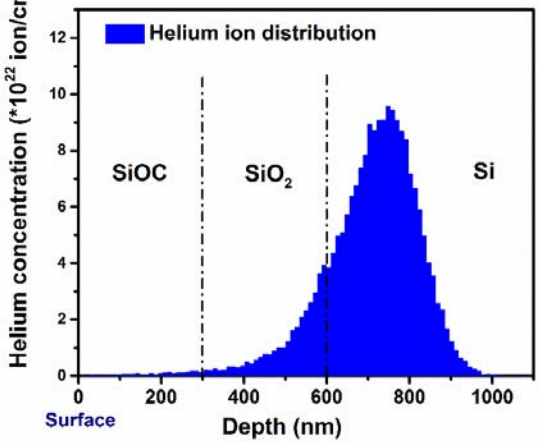
M. Nastasi, *et al.*, J. Nucl. Mater., 461 (2015) 200.
Q. Su, *et al.*, Phil. Mag. Lett., Vol. 96 , Iss. 2, 2016

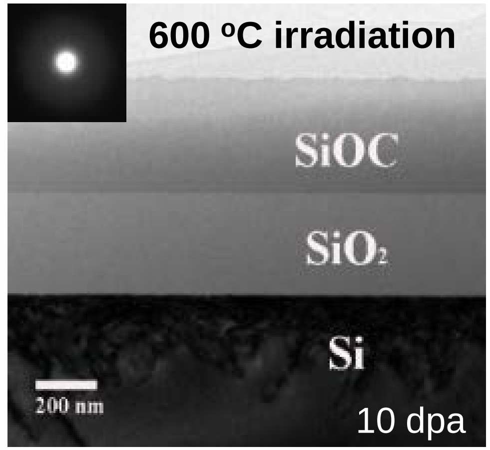
Summary of Kr and He irradiation of SiOC

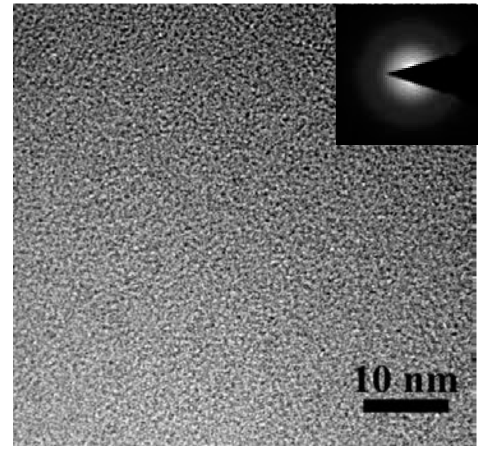
Ion species	Kr	He
Acceleration voltage	1 MeV	100 keV
Cascade type	Dense Cascades	Dilute cascades
Irradiation temperature	RT to 300 ° C	RT to 600 ° C
Dose	Up to 5 dpa	Up to 20 dpa
Crystallization	No	No
Void formation	No	No
Element segregation	No	No

Amorphous SiOC is a unique radiation tolerant material with ultra-high helium diffusivity!!!

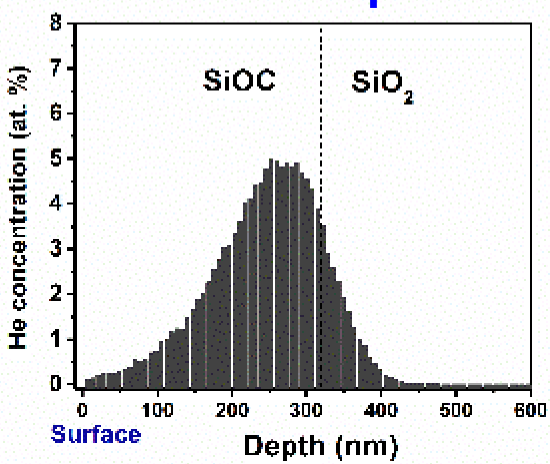
100 keV He irradiation



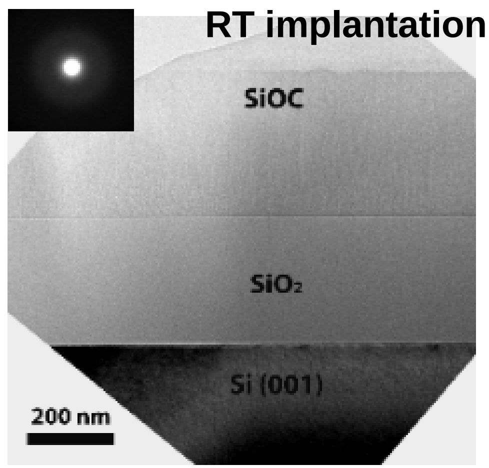




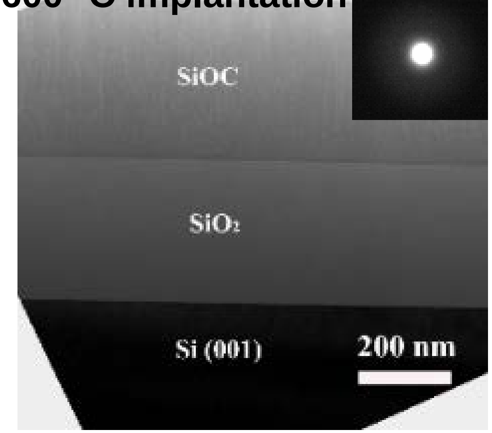
50 keV He implantation



10 at% He



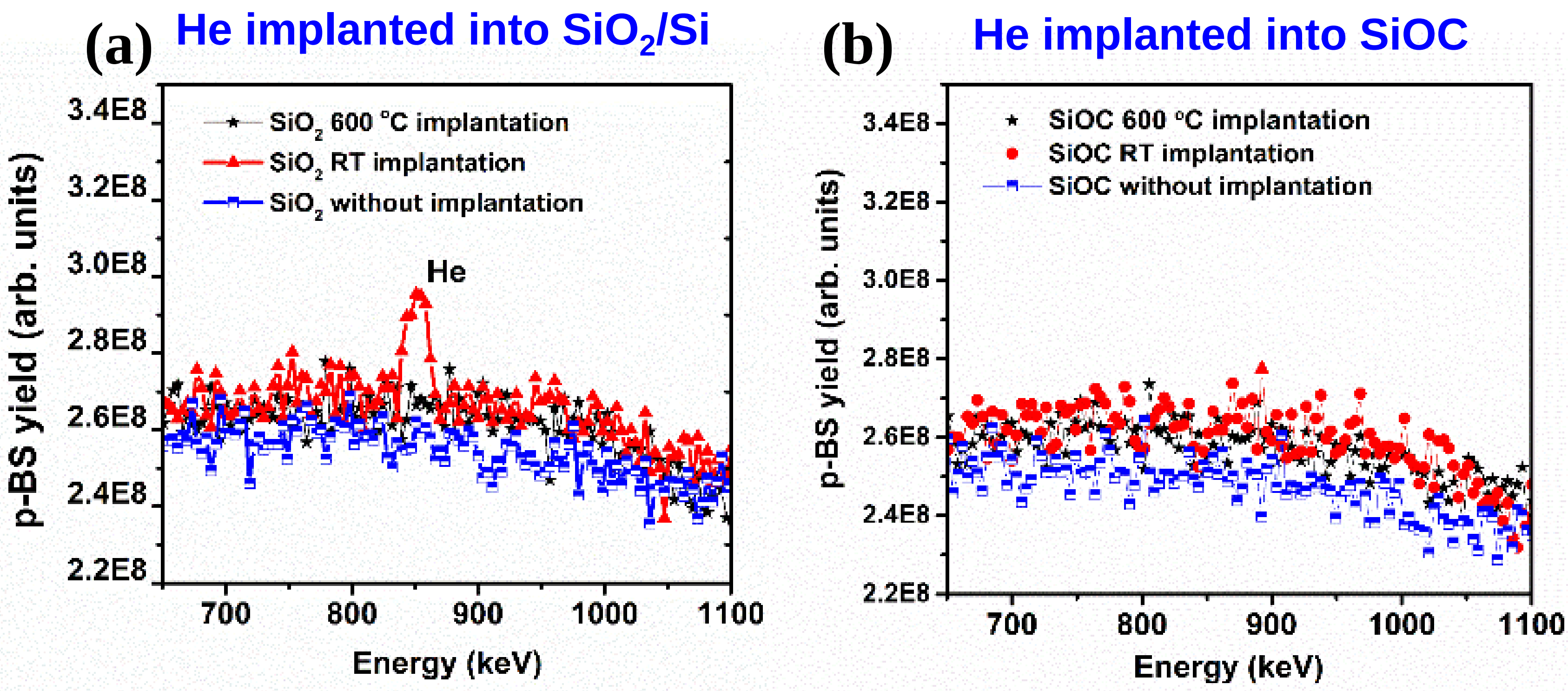
600 ºC implantation



No signs of He bubble formation!

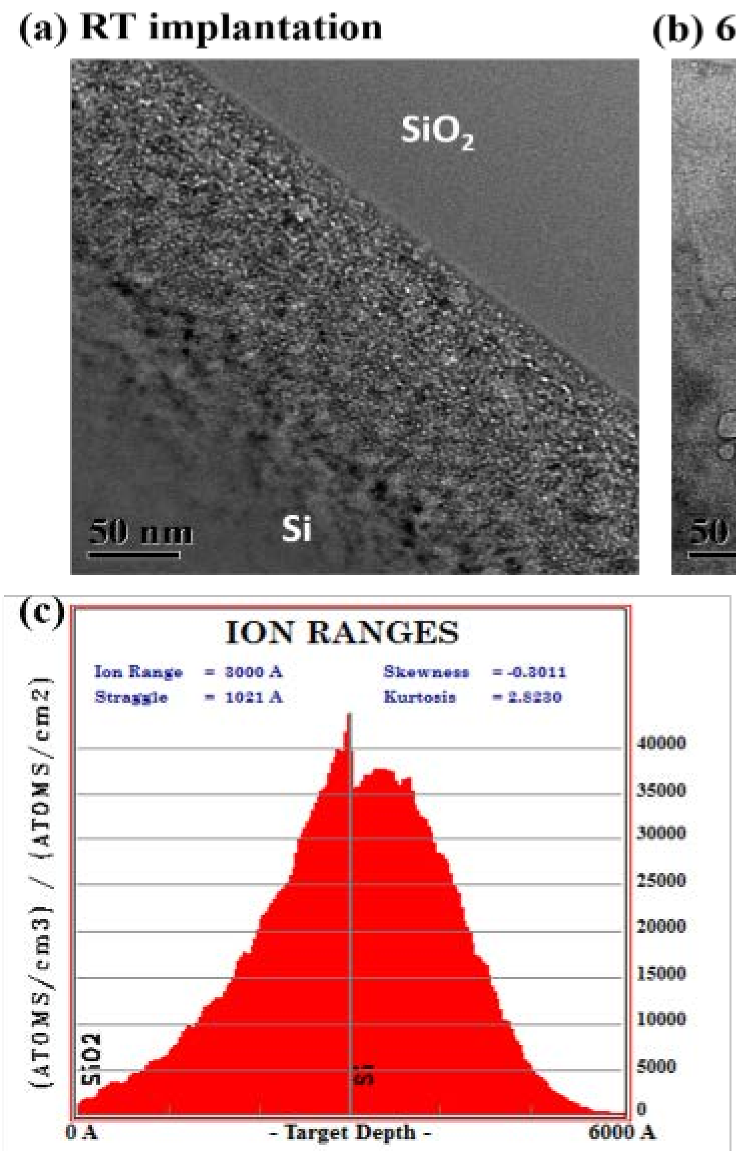
M. Nastasi, Q. Su, *et al.*, J. Nucl. Mater., 461 (2015) 200.

Proton non-Rutherford backscattering (10 at%)



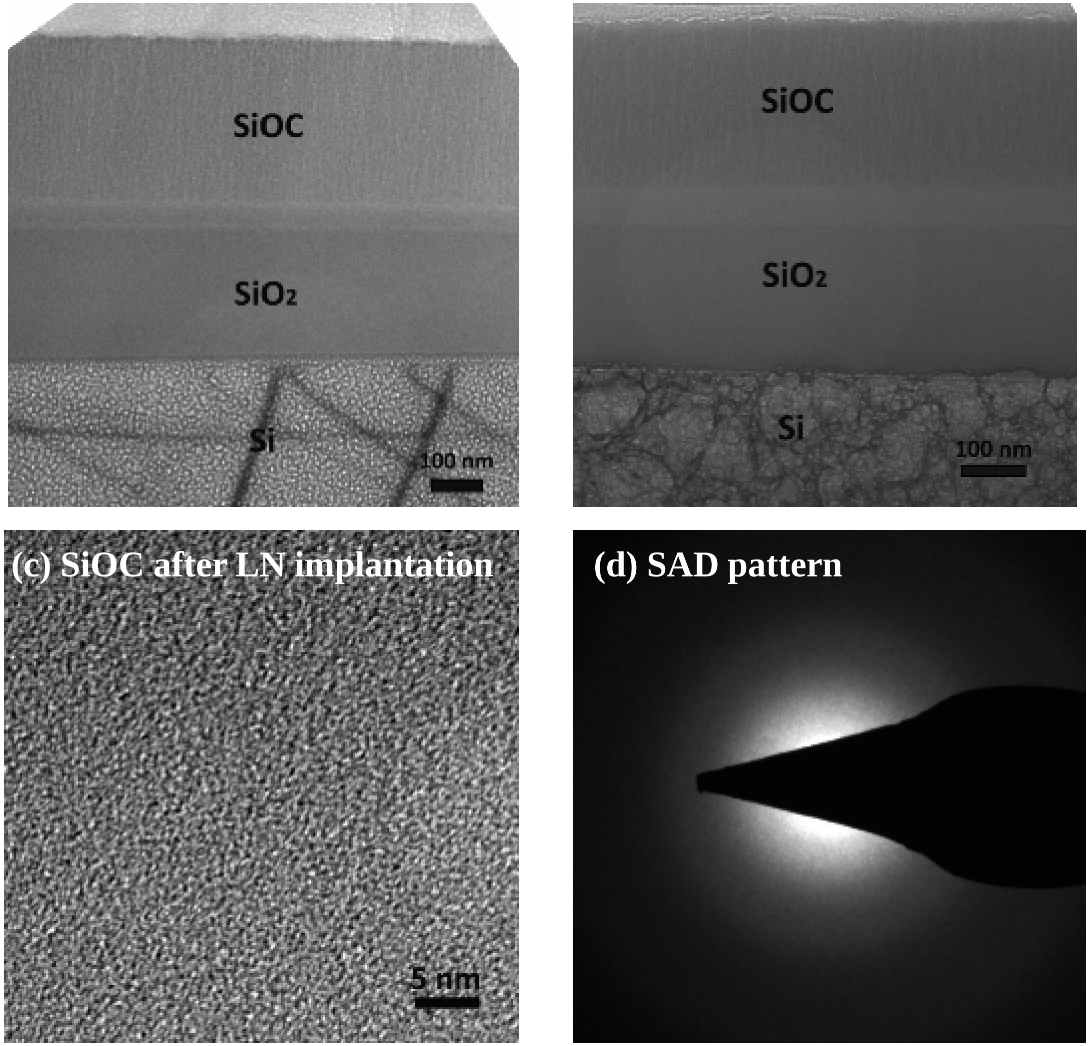
Helium atoms diffuse out of SiOC matrix even at RT!!!

He implanted into SiO_2/Si
Helium RBS signal is from helium bubbles in Si substrate.



In-Situ TEM 3 keV He Implantation Experiment

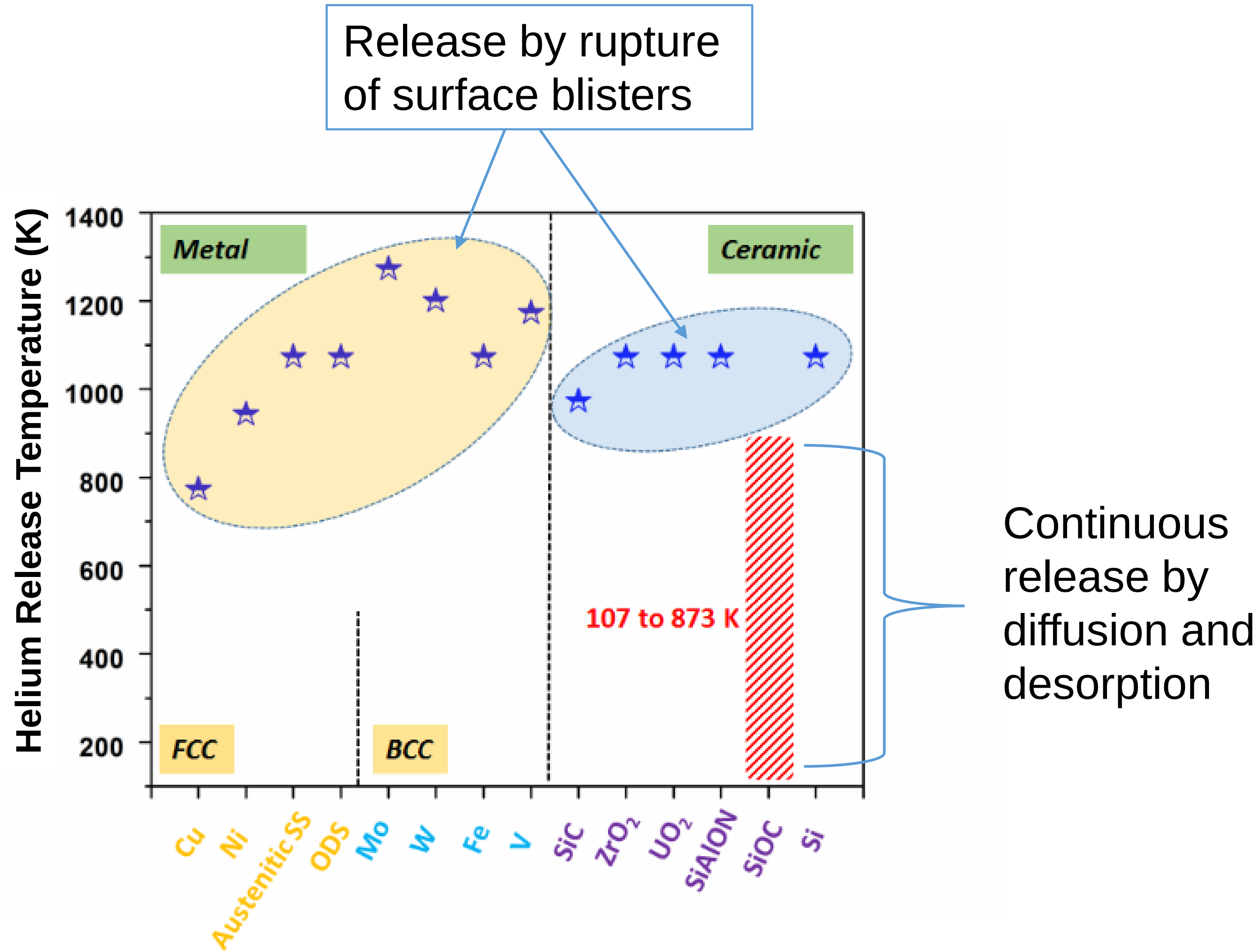
(a) LN implantation (50 at% He) **(b) RT implantation (50 at% He)**



In collaboration with Steve Donnelly at the University of Huddersfield, UK

- **No helium bubbles are observed in SiOC and SiO_2 layers and large amount of helium bubbles are shown in Si substrates.**
- **The size of helium bubbles in Si after RT implantation is much bigger than that after LN2 implantation.**
- **Amorphous SiOC retains its amorphous structure after LN2 implantation.**

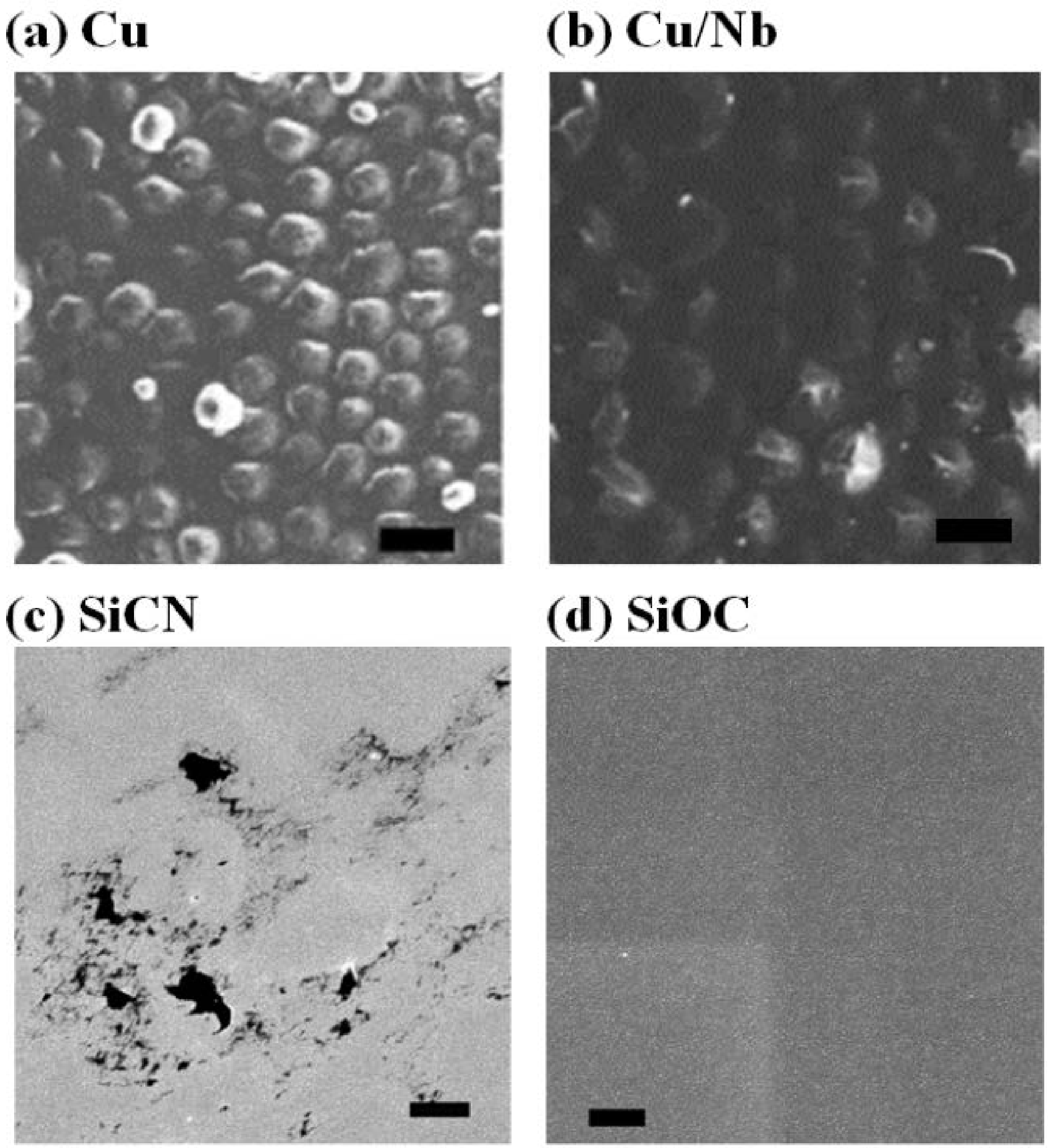
Helium Release in Materials



Surface blisters and damage following He implantation

1.5x10^{17} He ions/cm^2
600 C anneal

1.5x10^{17} He ions/cm^2
600 C anneal



7.4x10^{17} He ions/cm^2
Room temp.

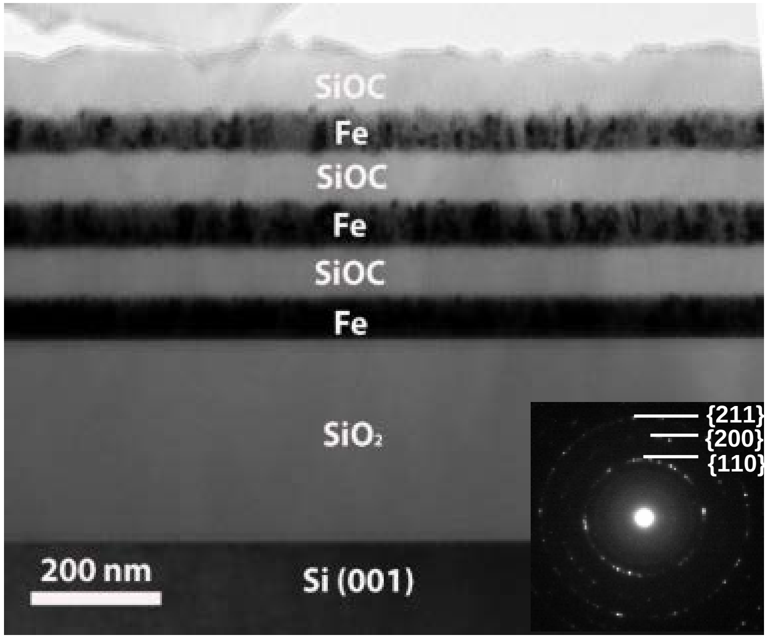
1.6x10^{17} He ions/cm^2
Room temp.

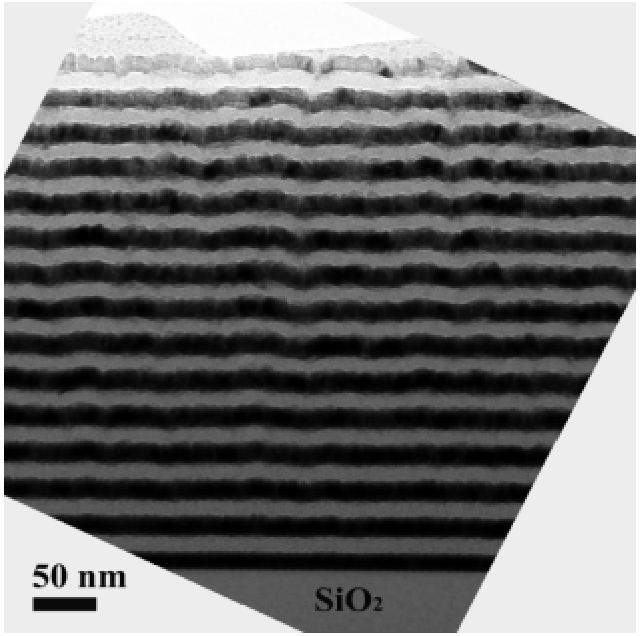
a) and b), Hochbauer, *et al*, J Appl Phys **98**, (2005).

The Role of Interfaces

Radiation tolerance of Fe/SiOC nanocomposites

Goal: To possess good mechanical and thermal properties, be capable of operation at temperatures greater than 500 ^{o}C, and have extreme radiation tolerance.

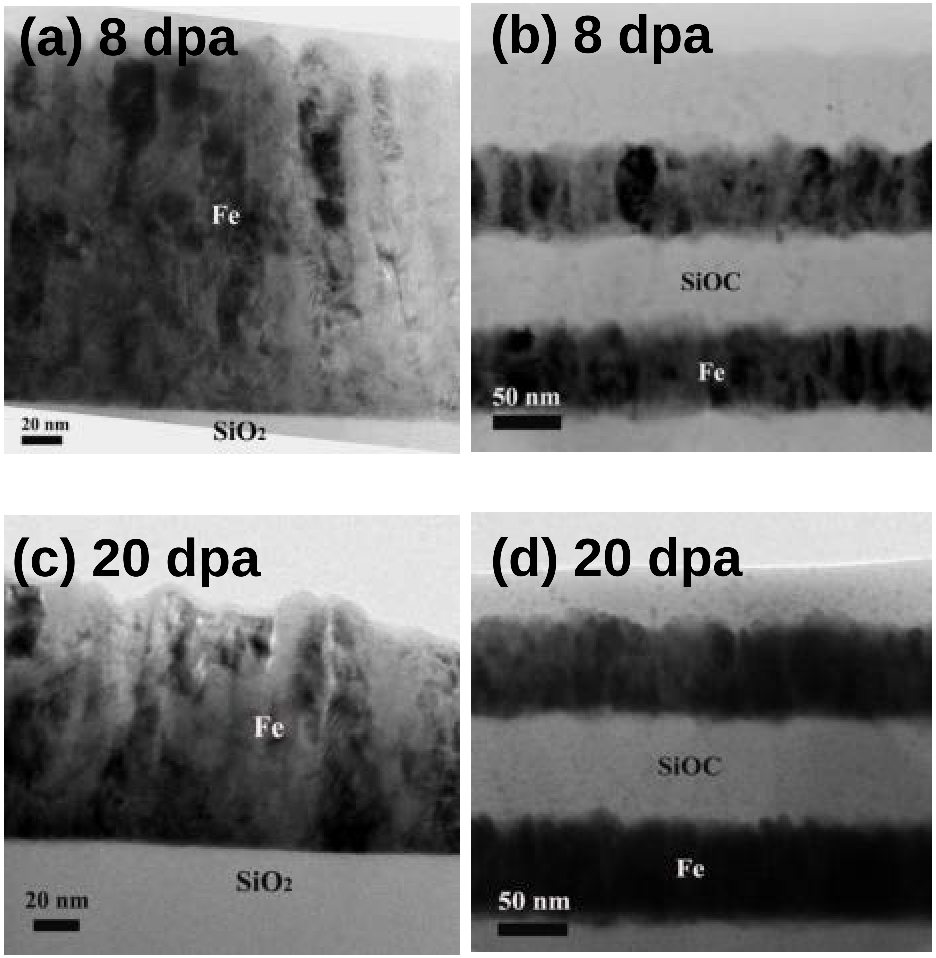


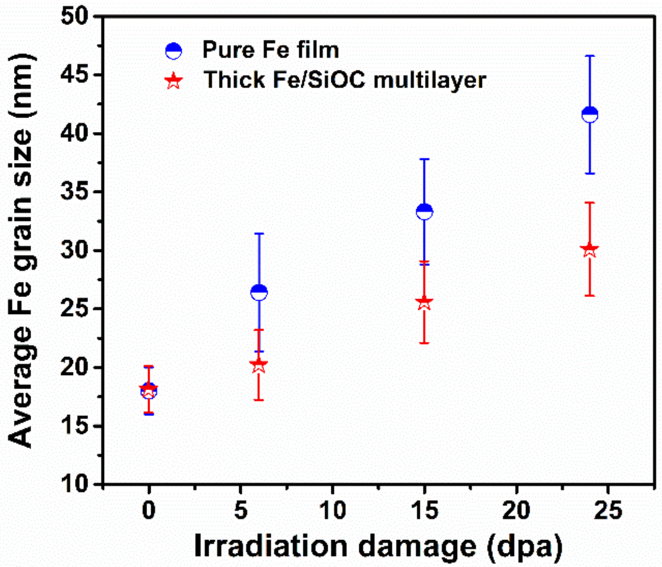


Multilayer films with different length scale consisted of BCC polycrystalline Fe with amorphous SiOC.

Q. Su, *et al.*, Materials Letters, 155 (2015) 138.

Room temperature He irradiation: Effect on Fe grain size



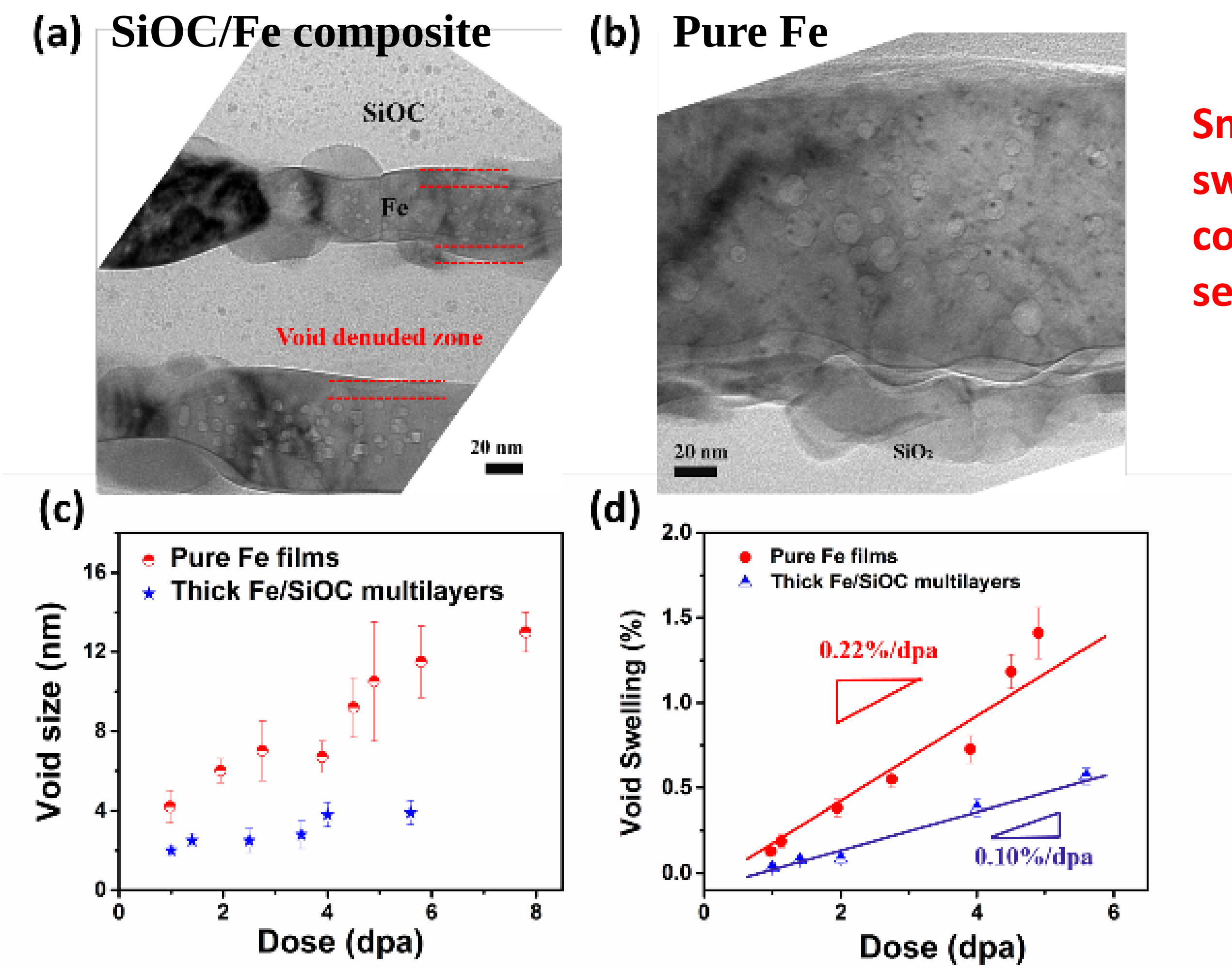


Fe/SiOC crystalline/amorphous interfaces act as efficient defect sinks, benefiting overall structure stability.

NCESR

Q. Su, *et al.*, Materials Research Letters, (2015) 1103796.

Improved swelling resistance of Amorphous SiOC/Crystalline Fe Nanocomposite (600 ºC He Irradiation)

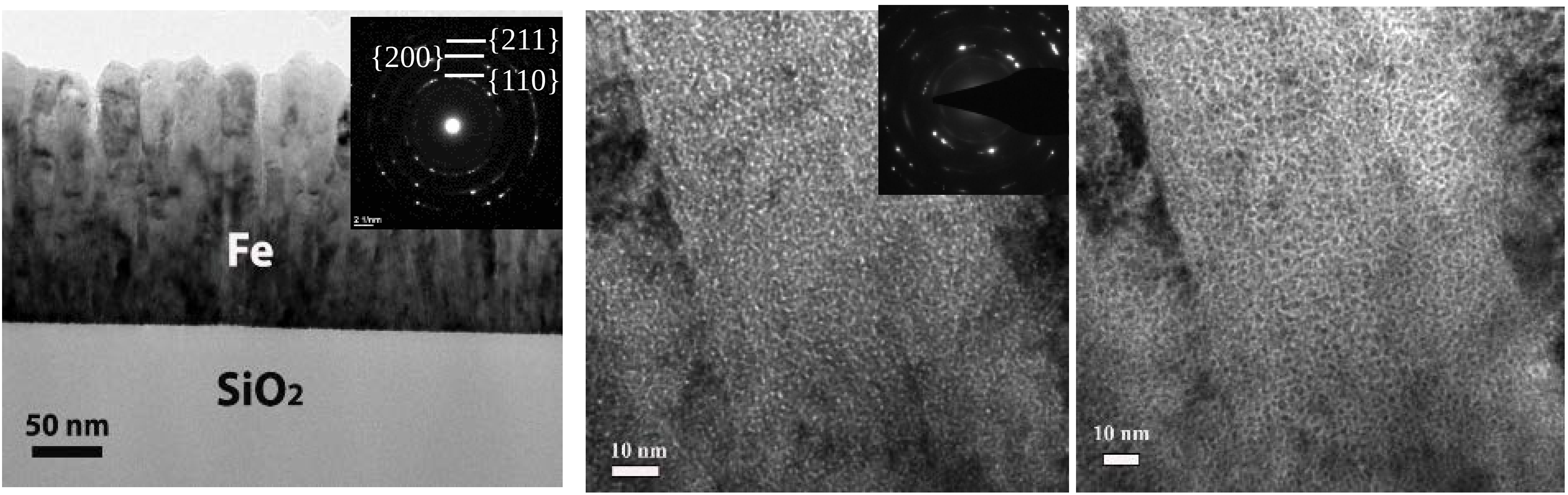


Smaller voids & Less swelling in SiOC/Fe composite. Interface serves as a sink.

Q. Su, *et al.*, Journal of Nuclear Materials, 479 (2016) 411.

Pure Fe films after 5 at% helium implantation exhibit presence of high density of helium bubbles (cavities).

(a) As-deposited (b) 5 at% He (under-focus) (c) 5 at% He (over-focus)

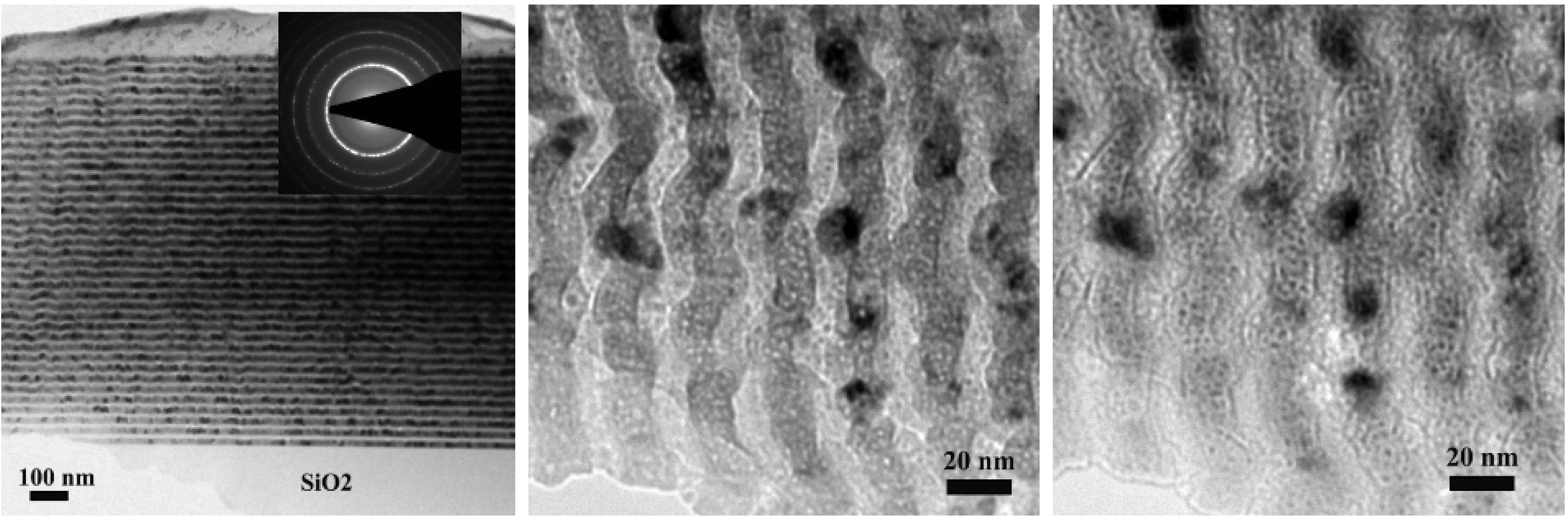


Bright dots with dark fringe. Dark dots with bright fringe.

Fresnel fringe suggests formation of helium bubbles (cavities) in Fe films after 5 at% He implantation.

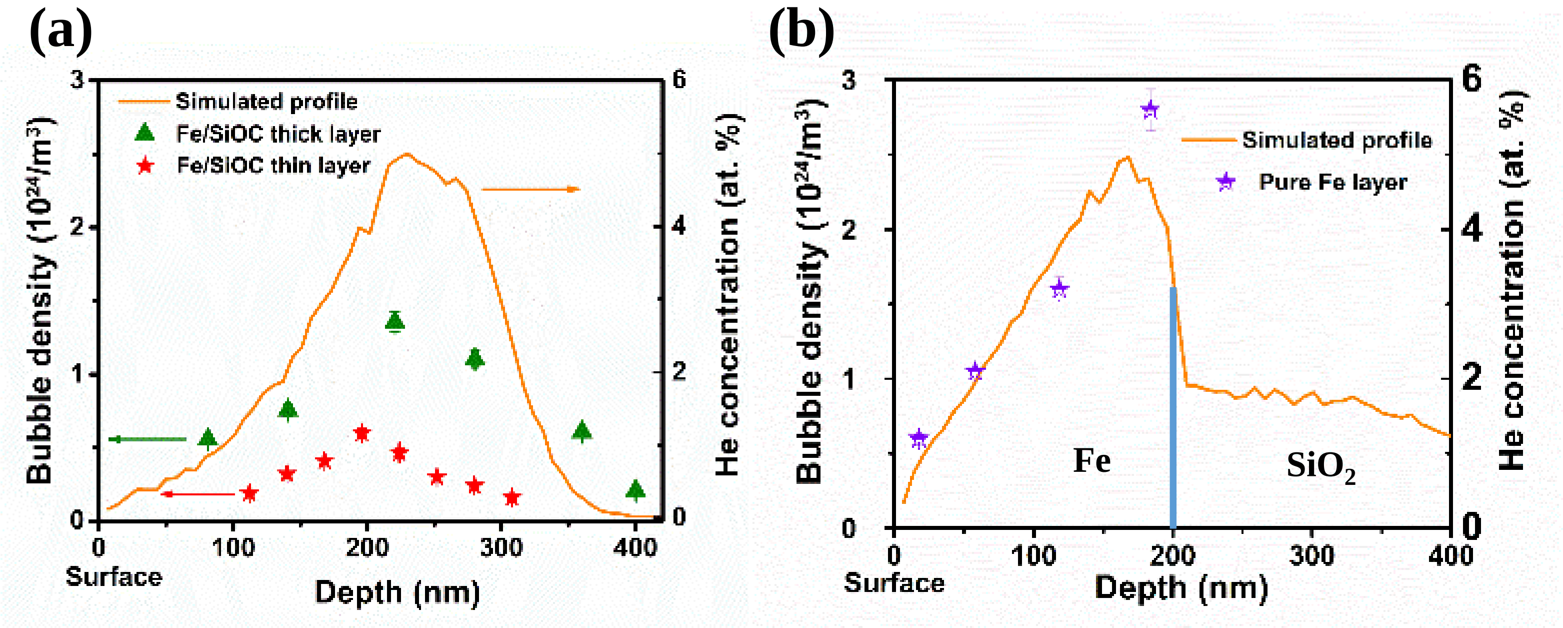
The presence of helium bubbles in Fe layers of thick Fe/SiOC specimen after 5 at% helium implantation

(a) 5 at% implantation (b) under-focus (c) over-focus



- Fe layers retain their BCC crystal structure. Fe grain growth is observed.
- SiOC layers are still amorphous
- He bubble formation in Fe but not in SiOC

Helium bubble distribution after RT implantation



Higher Fe/SiOC interface density

- **Lower helium bubble density**
- **Smaller helium bubble distribution range**

SUMMARY/CONCLUSIONS

- **Sources of ion irradiation: ion accelerators & neutron irradiation (e.g. (n, α) reactions)**
- **For typical ion implanter energies, nuclear stopping dominates atomic displacements and defect formation**
- **Microstructure can be tailored to control defect retention and accumulation**
- **Amorphous alloys may offer unique opportunities for radiation stability**

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U.S. DEPARTMENT OF ENERGY

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University of Nebraska

Thank you for your attention!!!

➢EXTRA SLIDES

Mechanical Properties for Samples Irradiated with 3.5 MeV Fe^{+} Ions