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Title: Nanofoams Response to Radiation Damage

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Nanofoams Response to Radiation Damage

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CAARI 2012, 5-10 August 2012, Fort Worth, USA

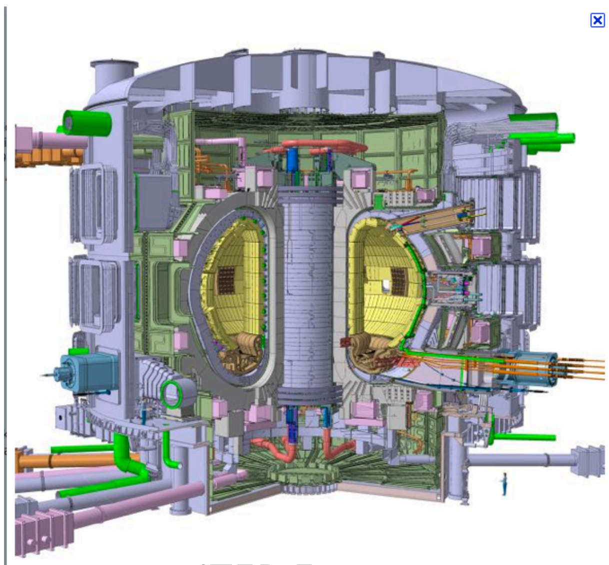
Motivation: Radiation Damage Effects in Nanoporous Materials - Fusion Reactor Designs

Radiation damage induce porous structure in tungsten in ITER

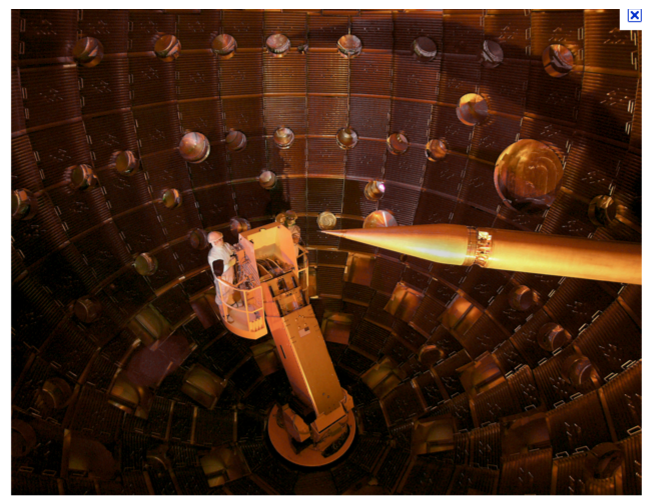
- **Nanoscale W-fuzz structure in divertor armour in ITER**
- **W armour FW plasma facing material**

Nanoporous materials for hydrodynamic stability in NIF targets

- **Foam shells in DT capsule in inertial confinement fusion design**
- **Low-density nanoporous Cu**
  - **<100 mg/cc; cel size ~100 nm**



ITER France

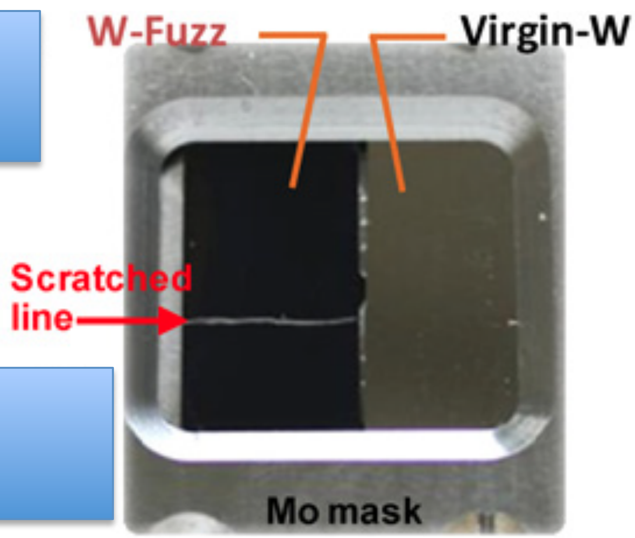


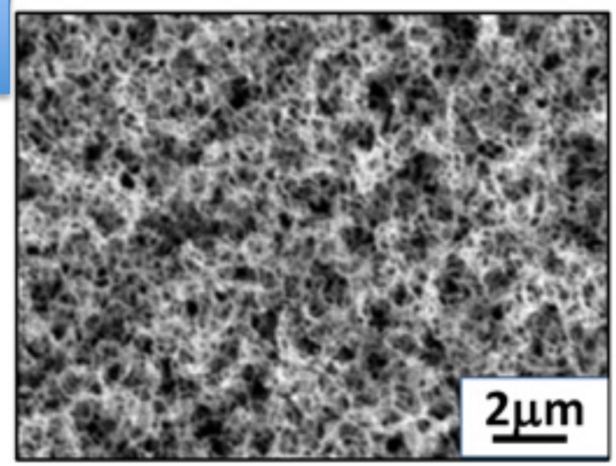
National Ignition Facility LLNL

Radiation effects induce porous structure in tungsten divertor armour in ITER plasma facing material

- **Growth of nanometre-sized fibreform structure (W-fuzz)**

Nanoporous tungsten created in the linear divertor simulator





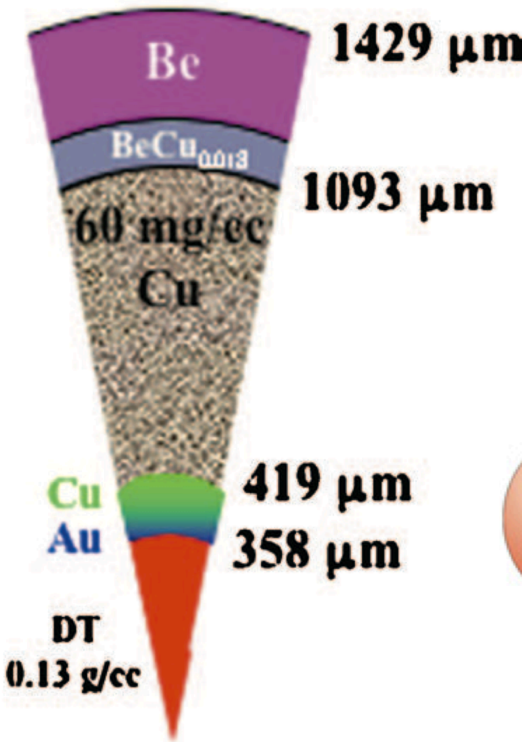
Temp.
1000-2000 K

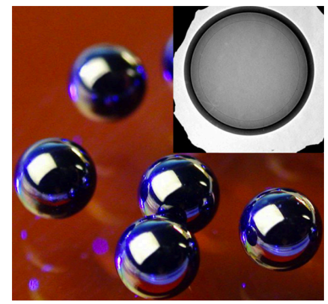
Helium ion
E > 20 eV

Tokitani, et al. Nucl. Fusion 51 (2011) 102001

Radiation effects might affect Cu foams performance during laser ablation and shell collision in NIF target

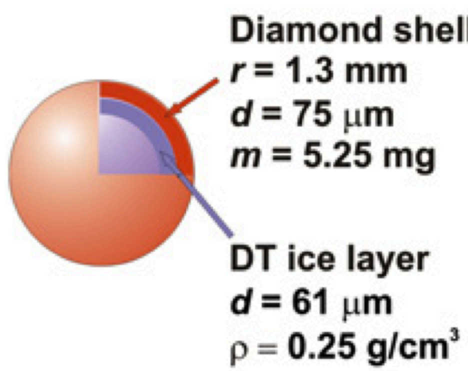
- **Non-cryogenic NIF ignition target**
- **DT at RT**





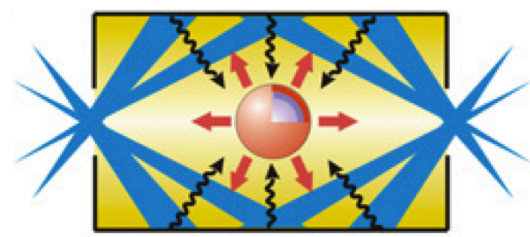
Digital x-radiograph Diamond target with inner layer of frozen DT fuel

Capsule inner diam. ~ 2 mm
Fuel layer ~50 μm thick



Amendt et al. Phys. Plasmas 14 (2007) 056312

Amendt et al., Phys. Plasmas 9 (2002) 2221

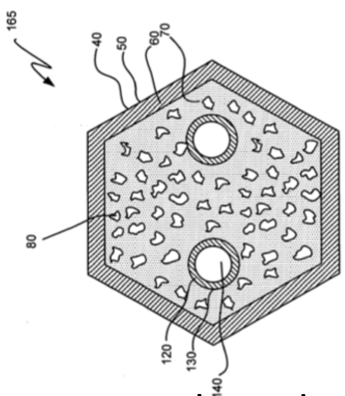


Indirect-drive Configuration

Biener et al, Nucl. Fusion 49 (2009) 112001

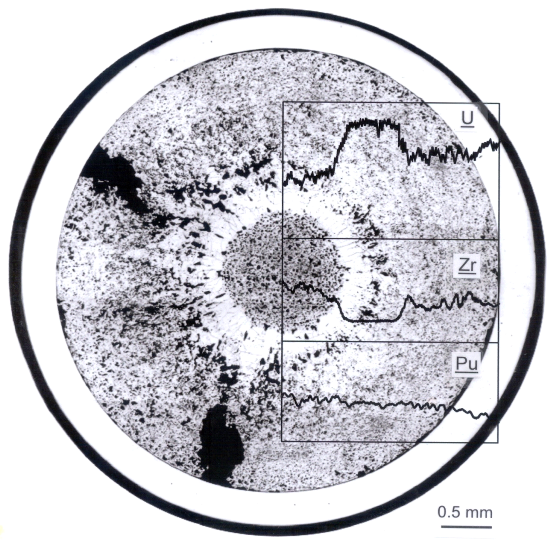
Radiation Damage Effects in Nanoporous Materials Fuel in Fission Reactors & Nuclear Space Propulsion

- **Refractory Open-Cell Foam Fuel for High Energy Nuclear Space Propulsion**
- **Nuclear fuel foam patent for fission reactor fuel assembly**
- **Radiation damage in nuclear fuels produce foam-like structure**

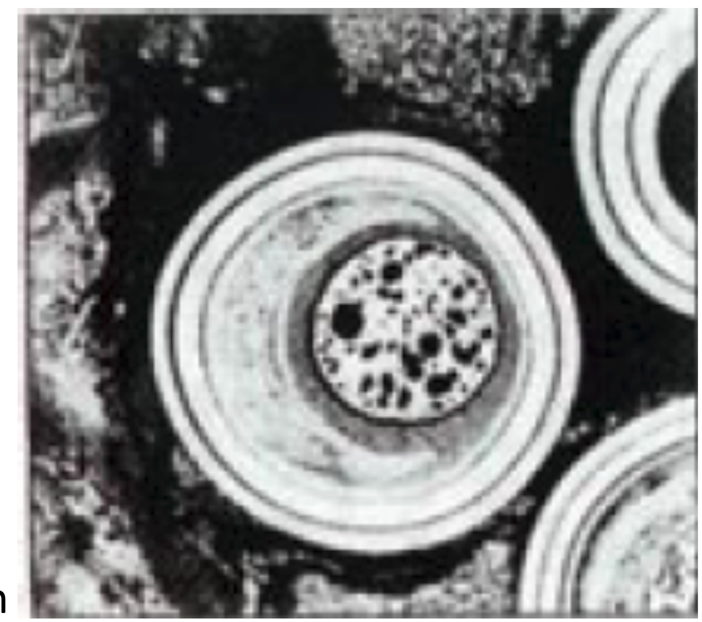


US. Patent on Foam based Fission reactor fuel assembly

Fast Reactor Fuel Pin



TRISO Particle in Pebble Bed Reactors

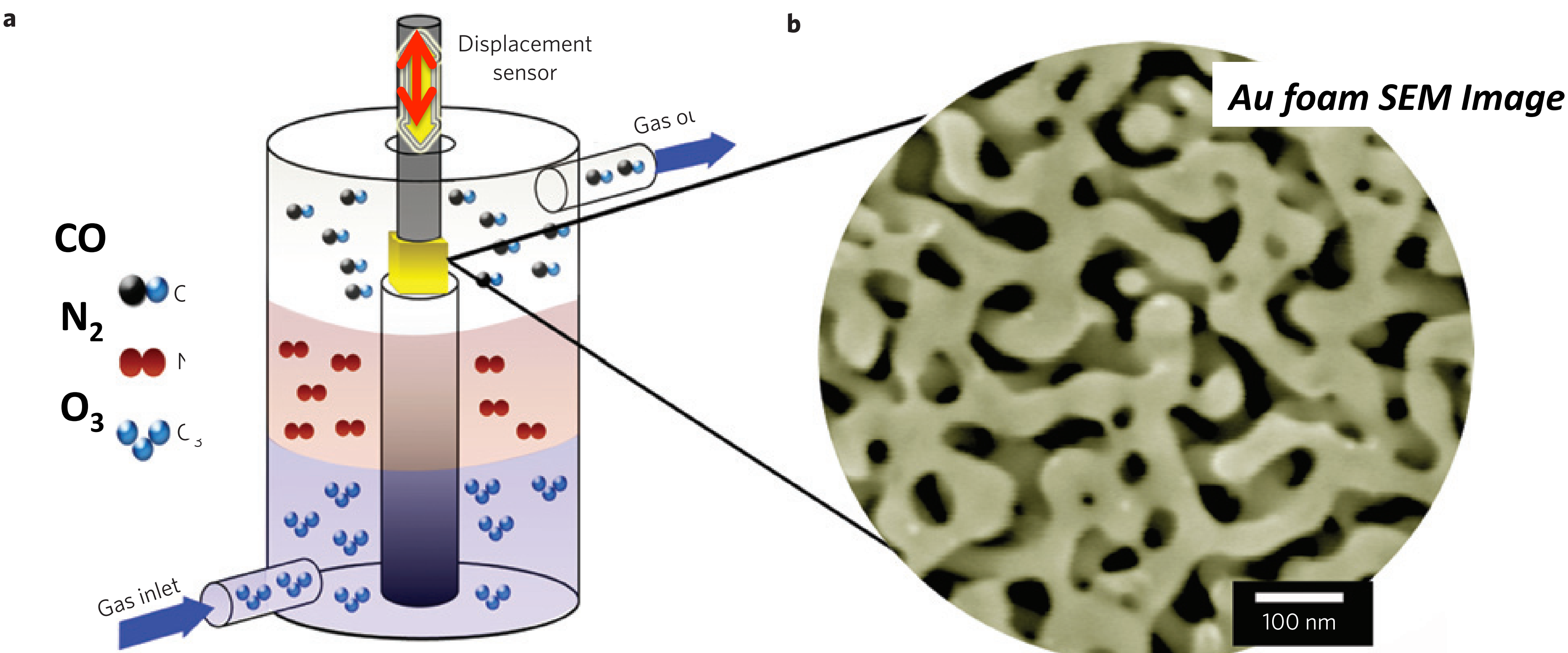


Nuclear fuel foam: Foam expands due to heat generation and fission gas release

Other Applications
Nanoporous Gold Materials as Actuators

Actuator: A motor that converts a source of energy to motion

Alternating exposure to O_3 and CO induces contraction and expansion of Au foams



Sample is purged with N_2 between exposures

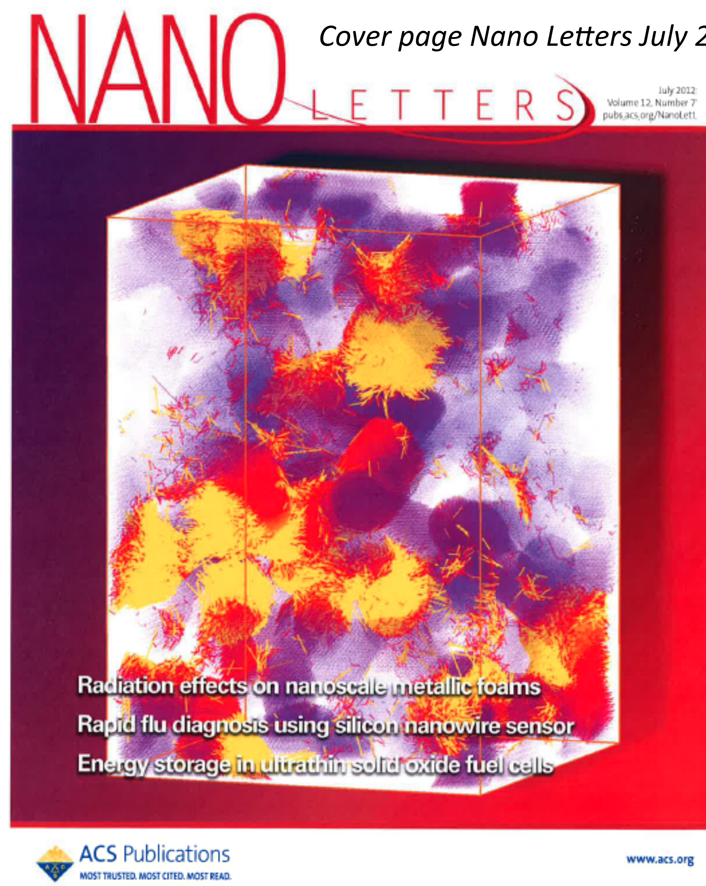
Biener et al. NMAT 8 (2009) 47

Radiation effects on nanoscale metallic foams

- **<u>Objective:</u> Explore the physics of surface-driven bulk physical behavior, such radiation tolerance**

- **<u>Opportunity</u>**:
- **Nanoporous materials could become a new class of extremely radiation tolerant materials**
  - **Nanoporous materials offer a large amount of free surfaces**
  - **Free surfaces act as sinks providing opportunity for irradiation induced defects to annihilate through diffusion**

Cover page Nano Letters July 2012



Atomic displacements larger than a_0 are shown
Red indicates more than 4 a_0

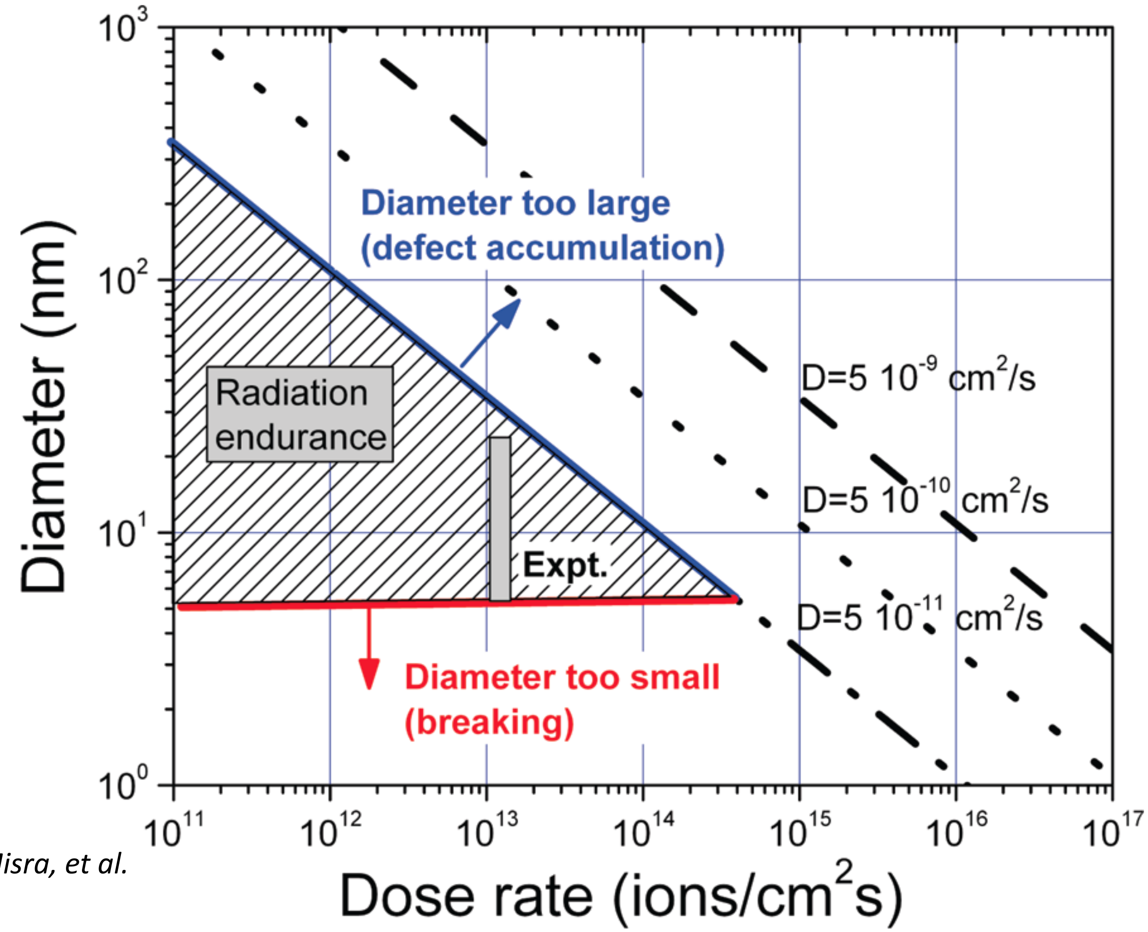
E. M. Bringa, J. D. Monk, A. Caro, A. Misra, et al. Nano Letters (2011) DOI: 10.1021/nl201383u.

The model: Window of Radiation Endurance

There exists a window in the parameter space where nanoporous material show radiation resistance

- **There exists an optimum ligament size at which radiation resistance occurs**

- **Defect migration to the ligament surface happens faster than the time between cascades, ensuring radiation resistance for a given dose rate**

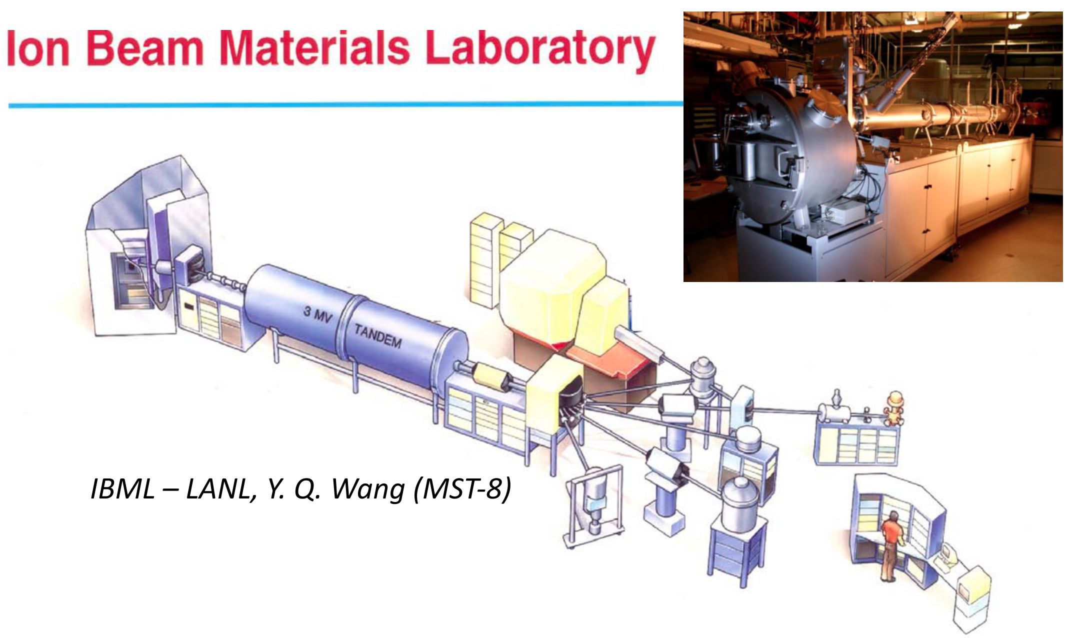


E. M. Bringa, J. D. Monk, A. Caro, A. Misra, et al. Nano Letters (2011)
DOI: 10.1021/nl201383u.

Irradiation experiments and computer simulation of nanoporous gold (np-Au) foams

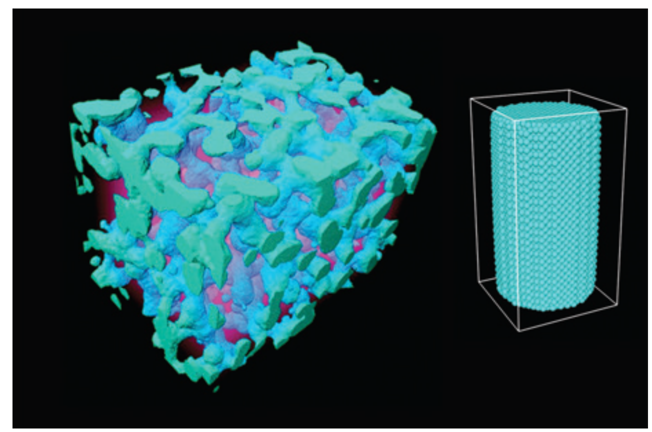
- **Develop a basic understanding of nanofoams radiation resistance**
- **Determine the evolution of the foam under irradiation:**
  - **Different dose-rates and irradiation temperatures**

Experiments performed in IBML
Danfysik 200 kV Research Ion Implanter



IBML – LANL, Y. Q. Wang (MST-8)

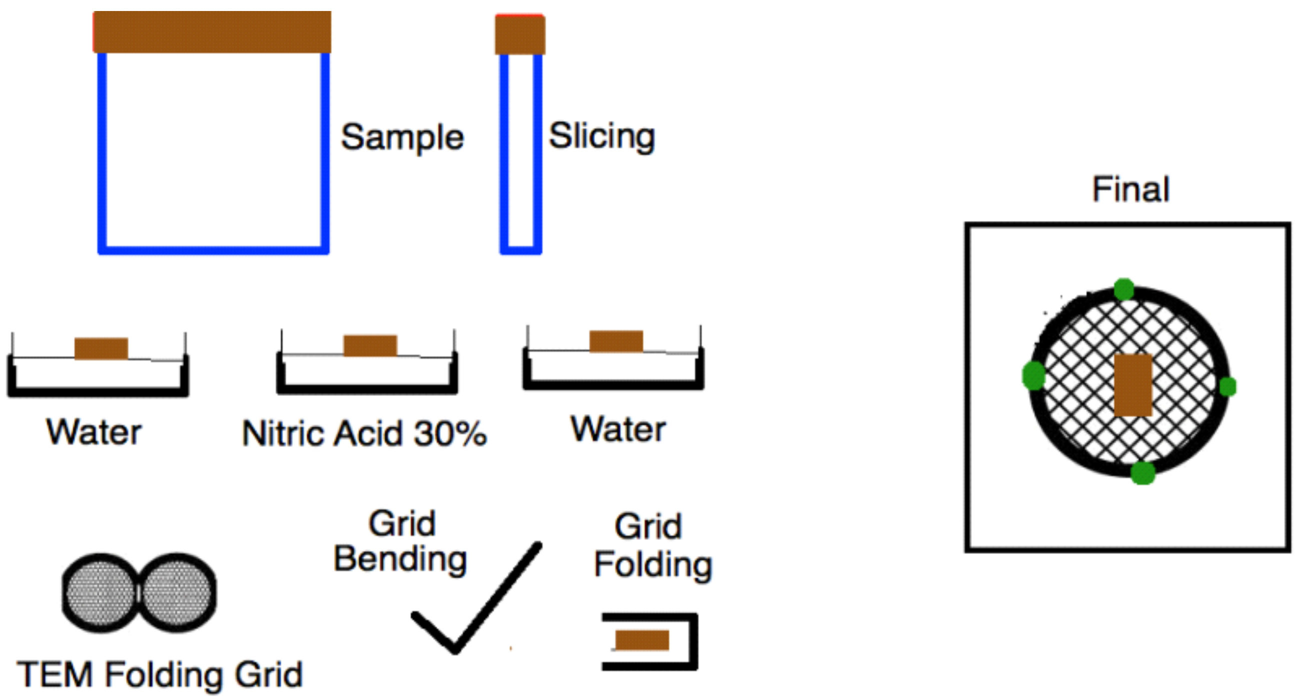
Computer simulation of np-Au foams and Au ligament



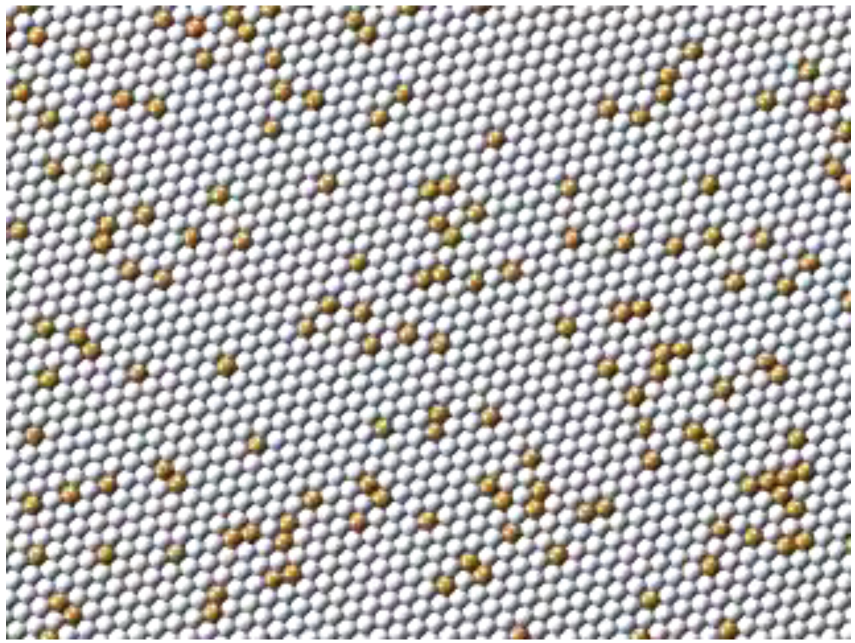
L. Zepeda-Ruiz in reference:
Biener et al. NMAT 8 (2009) 47

Schematics of np-Au specimen preparation for irradiation

- **Synthesis of Au-Ag films:** E-beam evaporation (RT) on NaCl substrates
- **Synthesis of np-Au foams:** Dealloying Au-Ag films in diluted (30%) or concentrated (70%) nitric acid (HNO_3) with varying dealloying time (1h to 18h)



Dealloying process in Au-Ag thin films



Ag atoms dissolve layer by layer and Au atoms clump together and form "clusters

Surface diffusion and dissolution events as simulated by the code MESOSIM

J. Erlebacher, Professor, Materials Science and Engineering Chemical and Biomolecular Engineering
http://p0217.projects.make-things.com/index.php/research/detail/fundamentals-of-dealloying

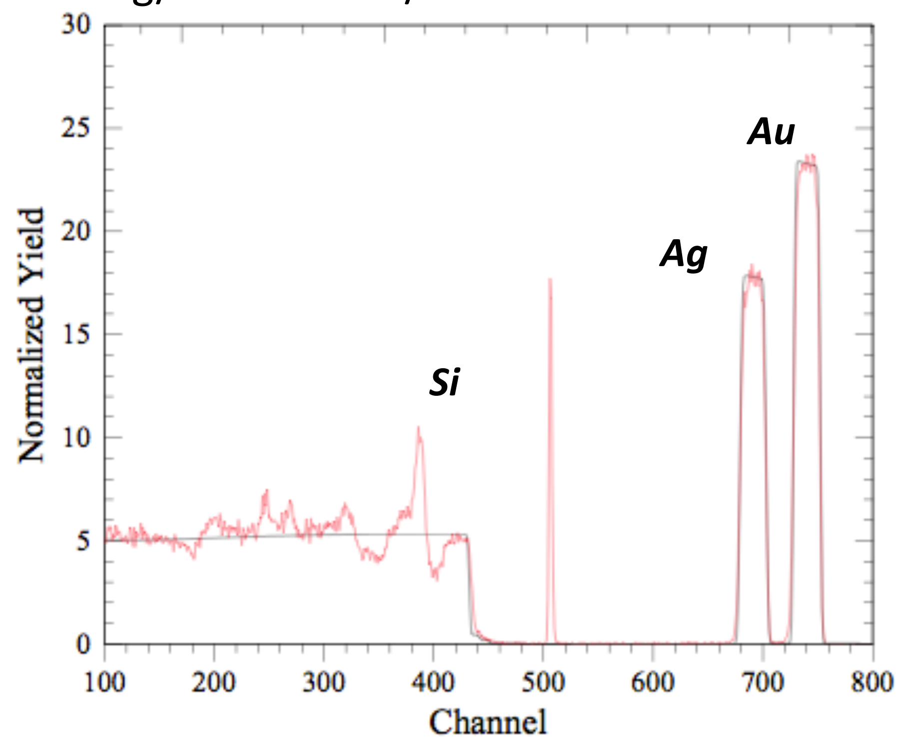
Optimizing Au-Ag Thin Film Thickness and Composition

Thin Film Fabrication

- Electron beam evaporation
  - Ag/Au co-deposition on single crystal NaCl (001) substrates
- Evaporation Conditions
  - Deposition Rate:
  - Ag 0.35 nm/s, Au 0.15 nm/s
  - RT, high vacuum < 5 x 10^{-8} torr
  - High purity Ag and Au > 99.9 %
- Variable thickness and Au%
  - 90 – 230 nm; 17 – 33 %Au
  - RBS spectroscopy 4.5 MeV ^{4}He ions (red line)
  - Spectroscopy analysis (RUMP software) – black line

Au is uniformly distributed in thin film

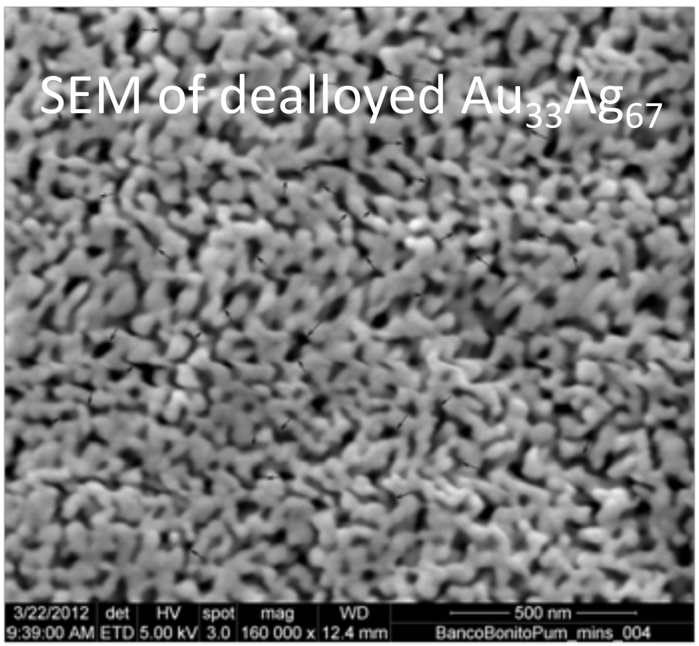
- RBS Example
- Thin Film Thickness ~ 160 nm
- Ag/Au Ratio ~ 7/3



RBS: 3.2 MV Tandem ion accelerator

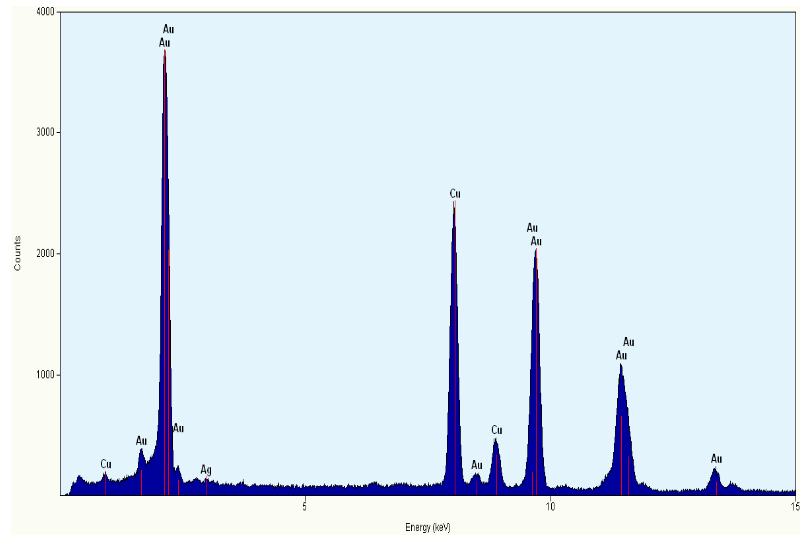
Characterization of non-irradiated np-Au foams

- **SEM image shows uniform ligament network**
- **Open porosity; Ligament size ~ 26 – 42 nm**



Thin film thickness ~ 230 nm
Etched for ~ 15 h in 40% diluted nitric acid

- **Ag dissolution occurs early in the dealloying process**
- **EDS shows no Ag left after dealloying thin film in 70% HNO_3 for 1h**



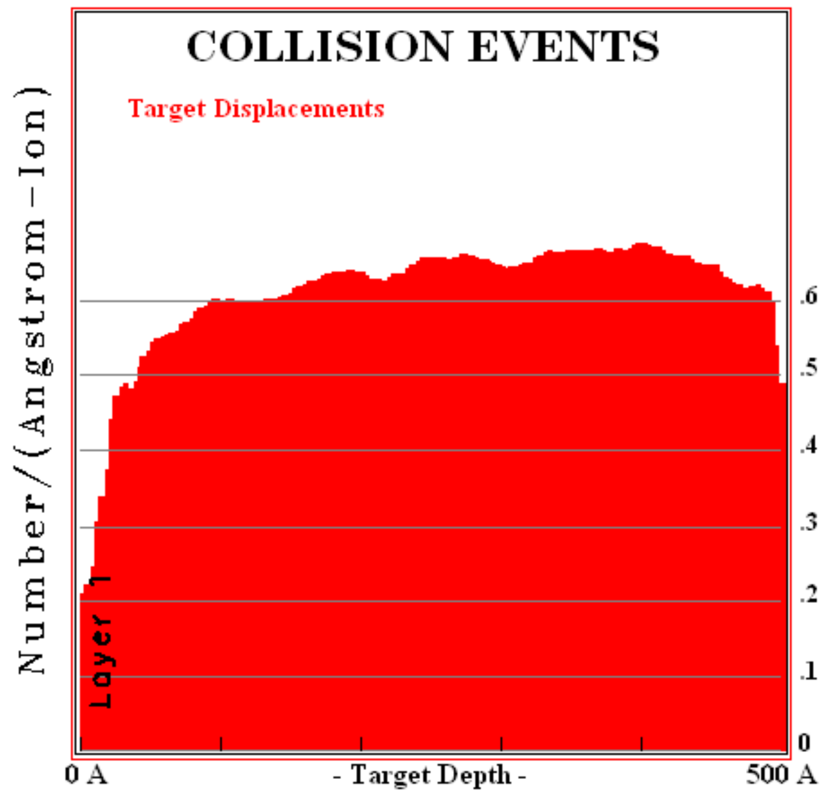
TEM Micrographs : FEI Tecnai 30, 300 kV
Oxford EDS system
SEM Micrographs: FEI Inspect F

Damage in np-Au as predicted by SRIM[1]

- **High ratio dpa/Ne concentration guarantees introduction of enough damage without forming Ne bubbles**
- **Damage:** 0.65 number/(A-ion) corresponds to ~1 dpa in the range from the sample surface to 50 nm deep
- **Ne Concentration:** < 0.03 at%
- **Simulation parameters:**

  400 keV Ne ions

  Fluence ~ 8.64×10^{14} ions/cm^2



[1] J.F. Ziegler, et al. The Stopping and Range of Ions in Solid. 1(1985)

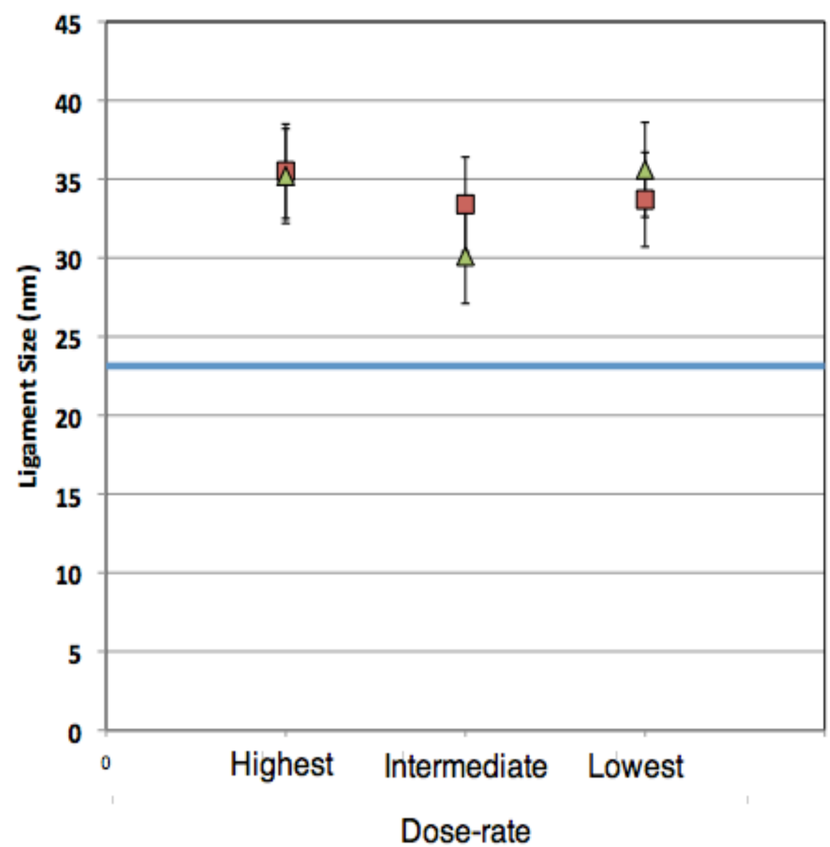
Radiation effects on np-Au morphology

Ion Irradiation Conditions

- 400 keV Ne^{++} on np-Au
  - 200 kV implanter Danfysik (IBML)
  - Total dose 8.64×10^{14} ion/cm^2
  - Dose-rates

    3×10^{12}, 3×10^{11}, 6×10^{10} ion/cm^2/s
  - RT and LN temperatures
- Net ligament diameter increase after irradiation
  - Coarsening process does not seem to exhibit any significant irradiation temperature dependence

Irradiation induces significant coarsening in np-Au ligament size

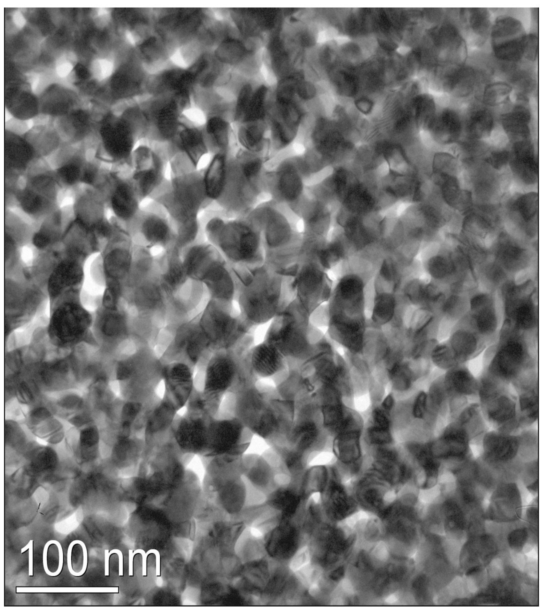


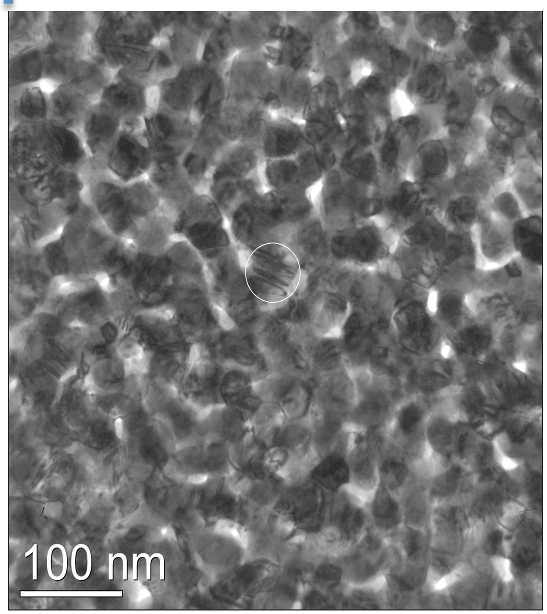
TEM before and after irradiation

- The ligament network is still present after irradiation with 400 Ne^{++} ions
- Highest dose ~ 1 dpa
- Highest dpa-rate 3 x 10^{12} ions/cm^2-s (3.5 x 10^{-3} dpa/s)

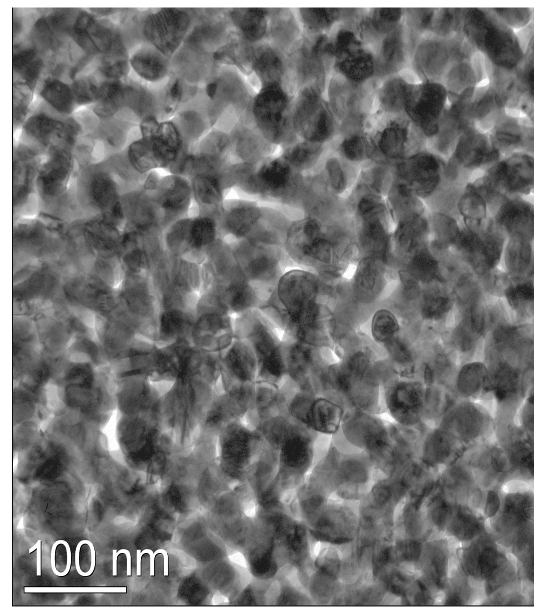
Irradiated np-Au foams

As-prep. np-Au





Liquid Nitrogen Temperature (LNT)

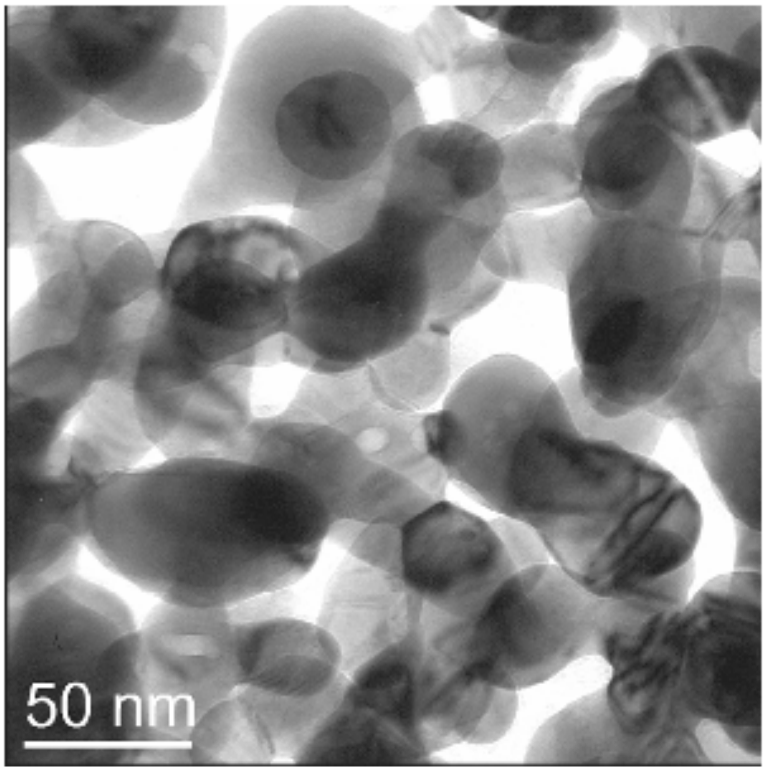


Room Temperature (RT)

Radiation effects on defect formation
np-Au irradiated at Room Temperature (RT)

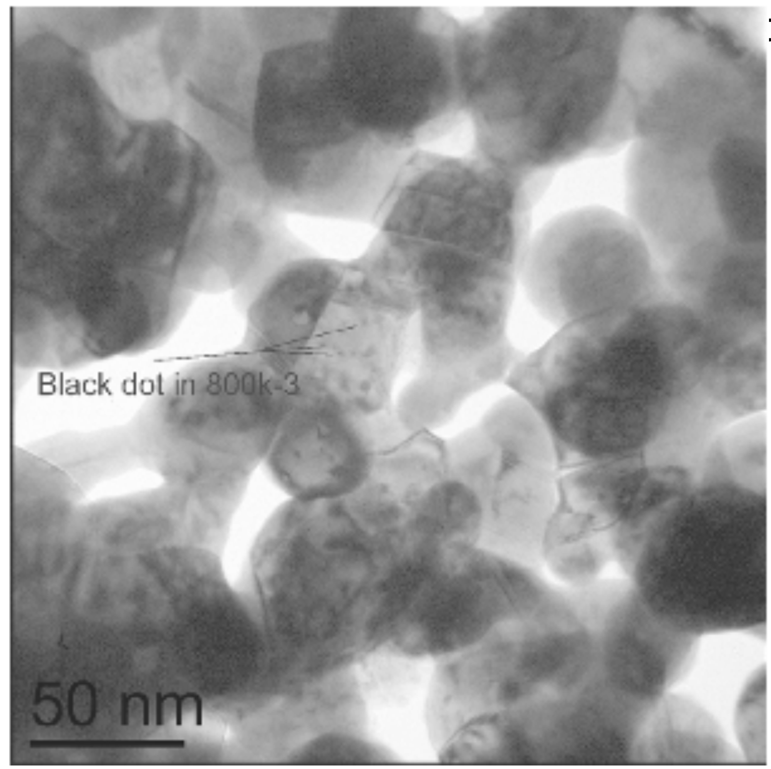
Non-irradiated Condition

- Pre-existing twins and stacking faults

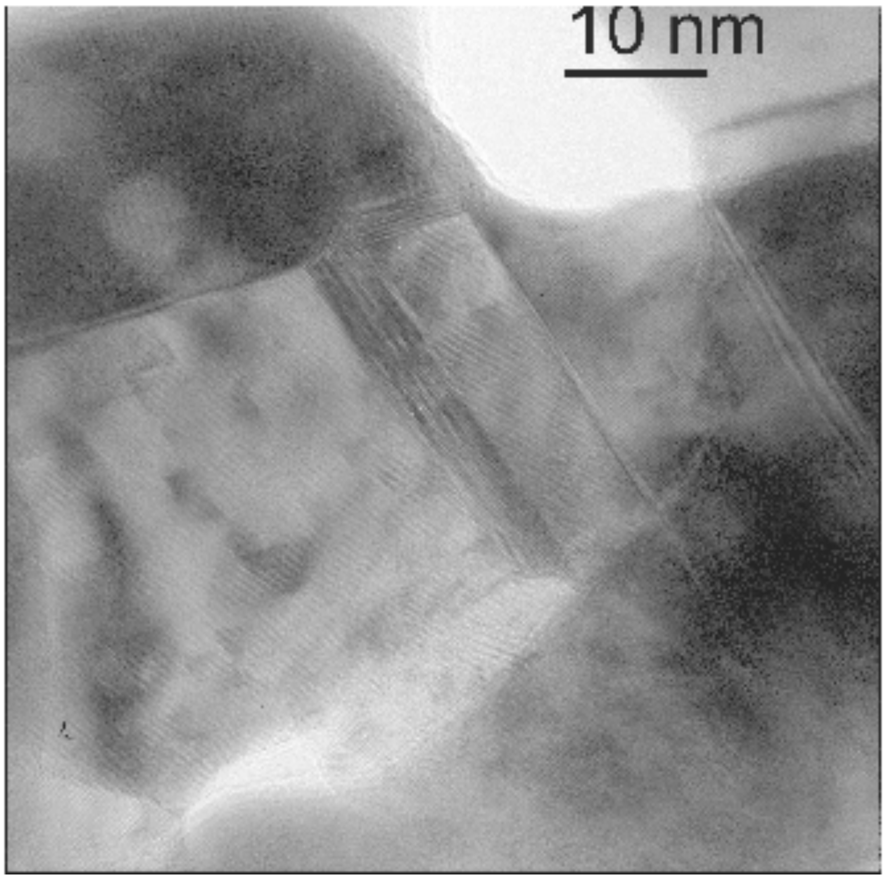


Irradiated Condition

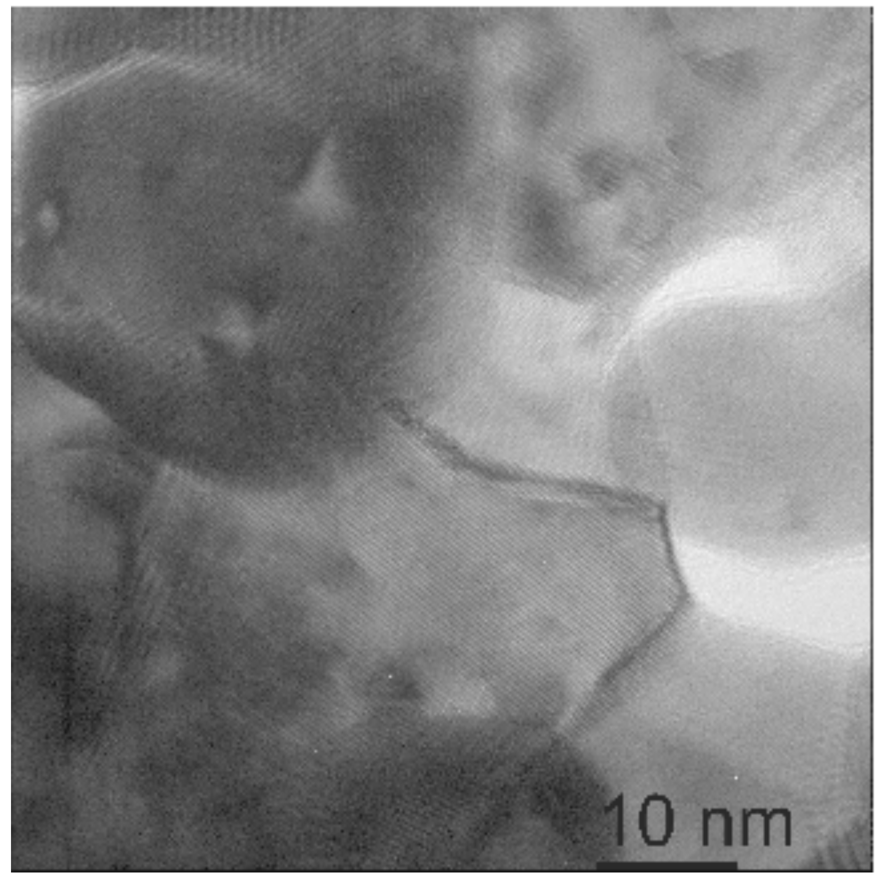
- Dark dots and dark lines appear



High-resolution image of defect formation
np-Au at RT shows the formation of Stacking Fault Tetrahedra (SFTs)



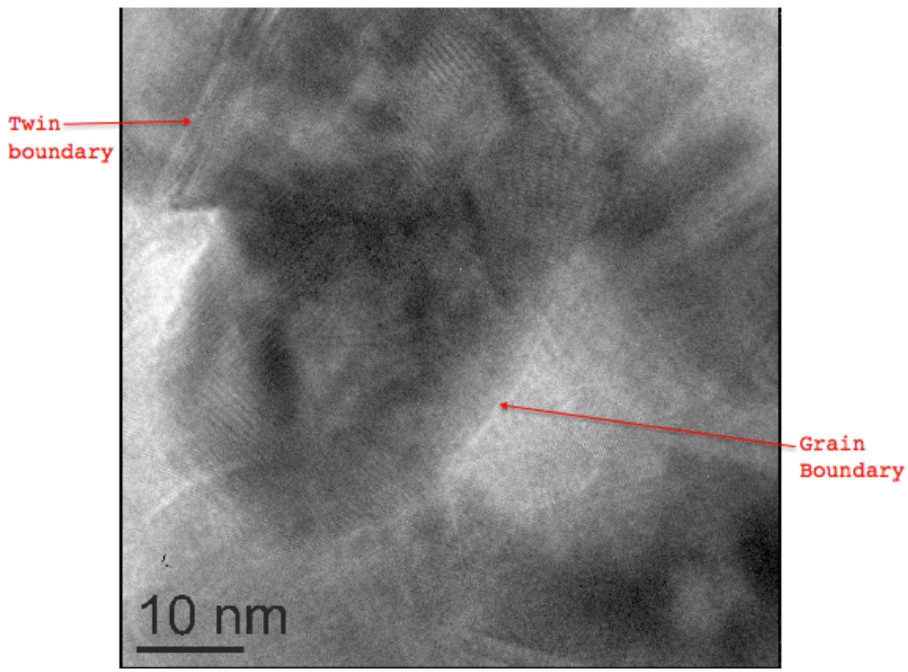
Twin Structure



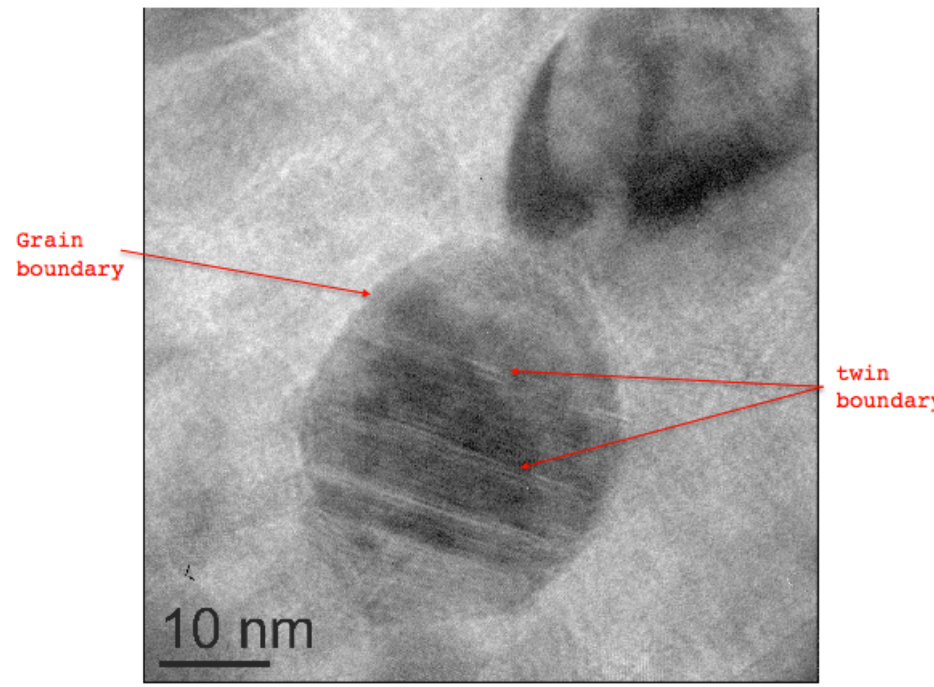
Grain Boundaries

High-resolution image of defect formation np-Au irradiated at Liquid Nitrogen temperature

Twin Structure



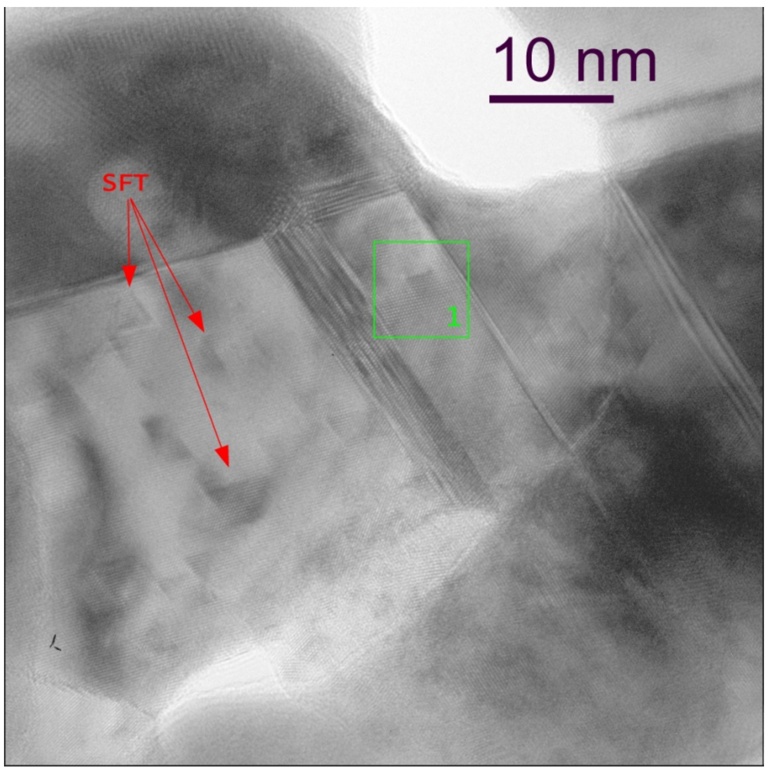
Grain Boundaries

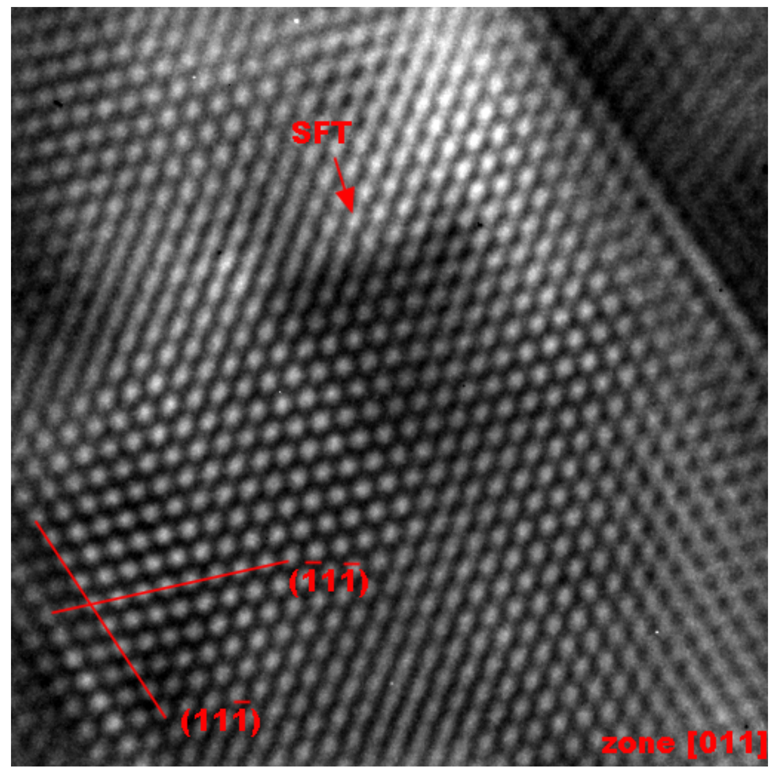


HRTEM indicates SFTs are not formed in np-Au after irradiation at LN temperature

HRTEM shows SFTs are formed in irradiated np-Au

- **SFTs are observed in np-Au foams irradiated at RT at intermediate and high flux values; i.e. flux > 6×10^{10} ions/cm^2/s**
- **No SFTs observed at low dose-rates and/or LNT irradiations**





Box 1: ~ 5 nm large SFT along (110) projection at the highest flux of 3×10^{12} /cm^2/s

Irradiation Temperature and Dose-Rates have an effect in np-Au response

- **RT allows vacancies to form SFTs**
  - At RT vacancies easily diffuse, accumulate and form SFTs
- **Vacancies were frozen at LNT**
  - Vacancies cannot diffuse to agglomerate and collapse to form SFTs
- **Low flux offer enough time interval for SIAs and vacancies to diffuse to the surface or recombine**
- **High flux allow vacancies to form SFTs before diffusing to the surface or recombine with SIAs since the rate of defect creation is high**

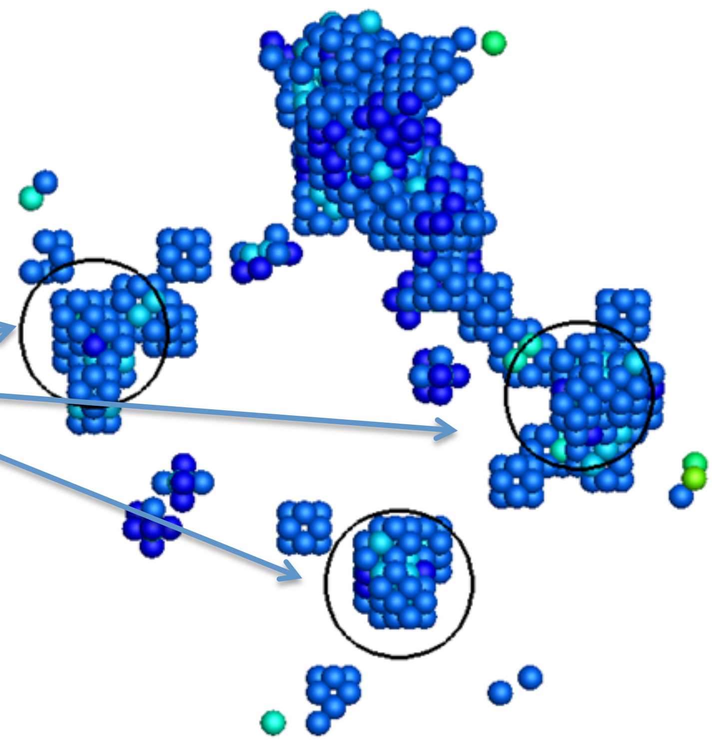
Molecular Dynamics simulations confirm damage accumulation and formation of several SFTs in Au ligaments

MD simulations of defect formation in Au ligaments under irradiation

- Steps in the simulation
  - RT equilibration (~ 2000 steps)
  - Followed by collision cascades
- Several 1.5 keV PKAs initiated at the ligament surface in the radial direction
  - Method: MD LAMMPS
  - Au cylinder 10 nm : 20 nm
    - [001]-oriented ligaments
    - 10^5 atoms

LAMMPS http://lamps.sandia.gov
EAM: Embedded atom method Foiles, Baskes, Daw PRB 33 (1986) 7983

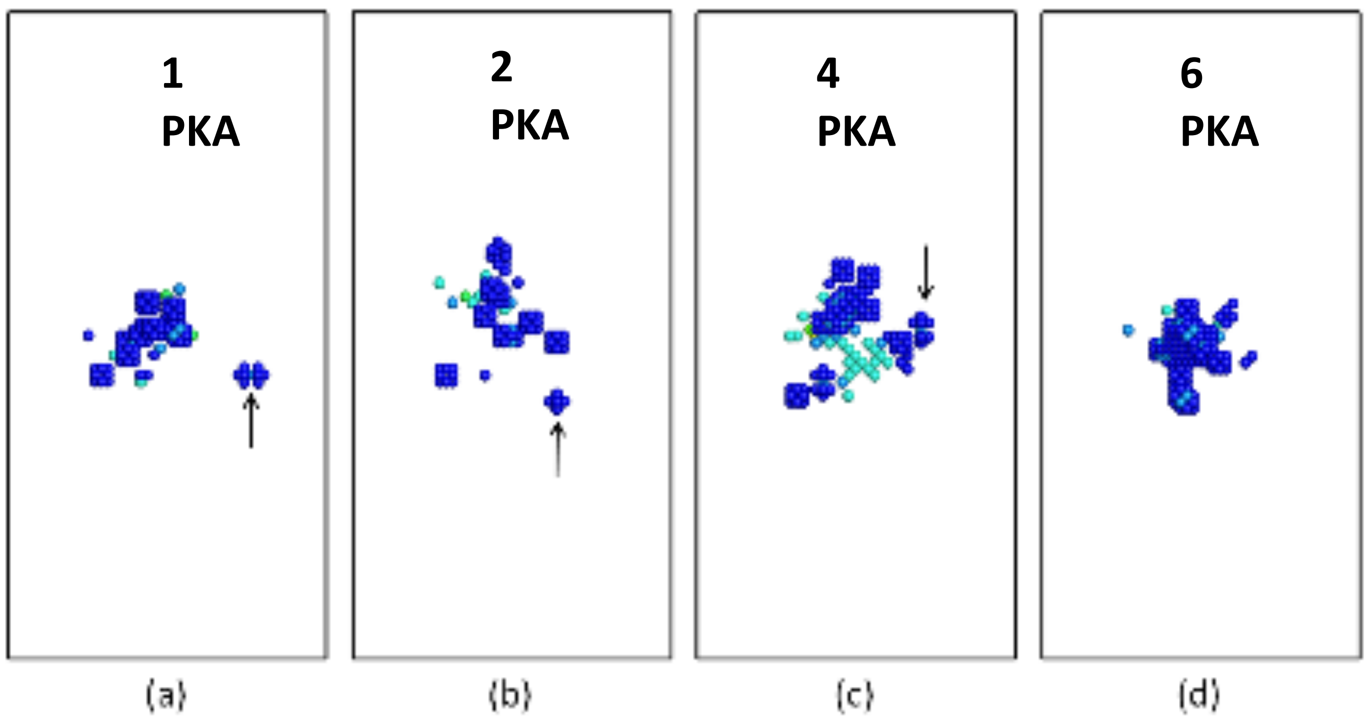
SFTs formed after 8 cascades



Defective atoms at the end of several collision cascades

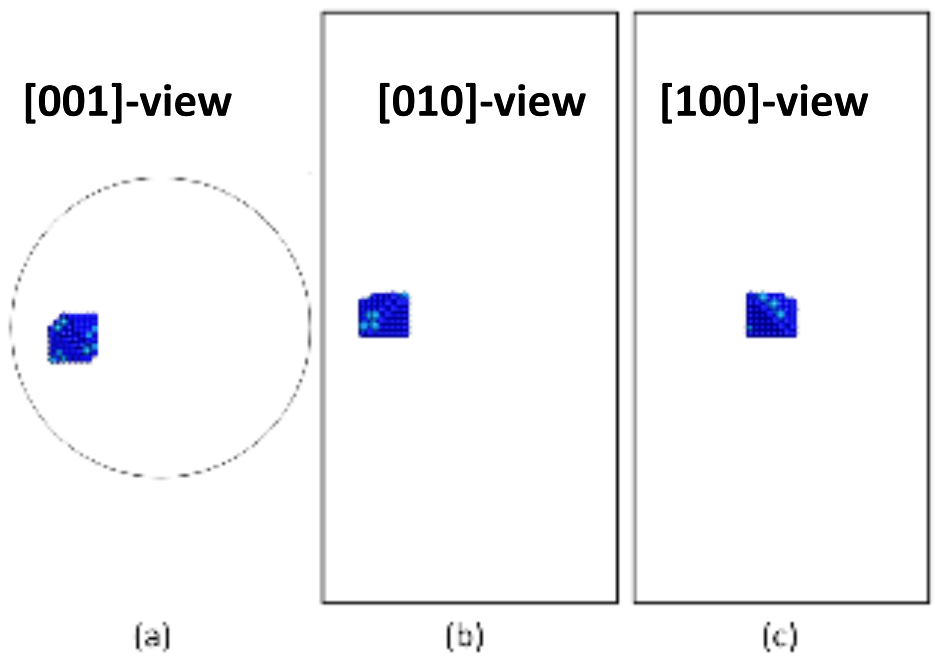
Au ligament response after accumulation of several 1.5 keV PKA induced cascades

- At the end of each collision cascade the ligament gets decorated by individual vacancies and very few self-interstitial atoms appear (SIA: indicated by black arrow)
- As the ligament cools down SIAs migrate to the surface or recombine with other vacancies; Example: SIA is present in a) b) c) but is not present in d)



Vacancy agglomeration induces vacancy cluster formation. Then cluster collapses to form an SFT

- Simulation results :
- Top and side images of Au ligament after 6 cascades generated by 1.5 keV PKAs



Vacancies surrounding the SFT were removed from the image for clarity

Conclusions

- np-Au foams were successfully synthesized by de-alloying process
- np-Au foams remain porous structure after Ne ion irradiation to 1 dpa
- SFTs were observed in irradiated np-Au foams with highest and intermediate flux, while no SFTs were observed with lowest flux
- SFTs were observed in irradiated np-Au foams at RT, whereas no SFTs were observed at LNT irradiation
- The diffusivity of vacancies in Au at RT is high enough so that the vacancies have enough time to agglomerate and thus collapse. As a result, SFTs were formed.
- The high flux created much more damage/time; vacancies don't have enough time to diffuse or recombine. As a result, SFTs were formed

Future Work

- Determine the role of surface-to-volume ratio on the irradiation response of nanoscale foams, i.e.different ligament/pore size in np-Au
- Irradiation response under irradiation with different projectiles (He, Xe)
- Dose dependence of defect evolution in np-Au
- Other np-metals (Ni-Pt alloy foams)

Acknowledgement

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on
Radiation Resistant Nanoporous Materials

CAARI 2012, 5-10 August 2012, Fort Worth, USA