This paper describes objective technical results and analysis. Any subjective views or opinions that might be expressed in the paper do not necessarily represent the views of the U.S. Department of Energy or the United States Government.

SAND2022-2291C

Sandia National Laboratories

Unravelling the Mechanisms of Irradiation Induced Phase Transformation in Nanocrystalline Gold

James E. Nathaniel II, Pranav K. Suri, Nan Li, Yongqiang Wang, Doug Medlin, Khalid Hattar, and Mitra Taheri

TMS 2022 Annual Meeting, Anaheim, California

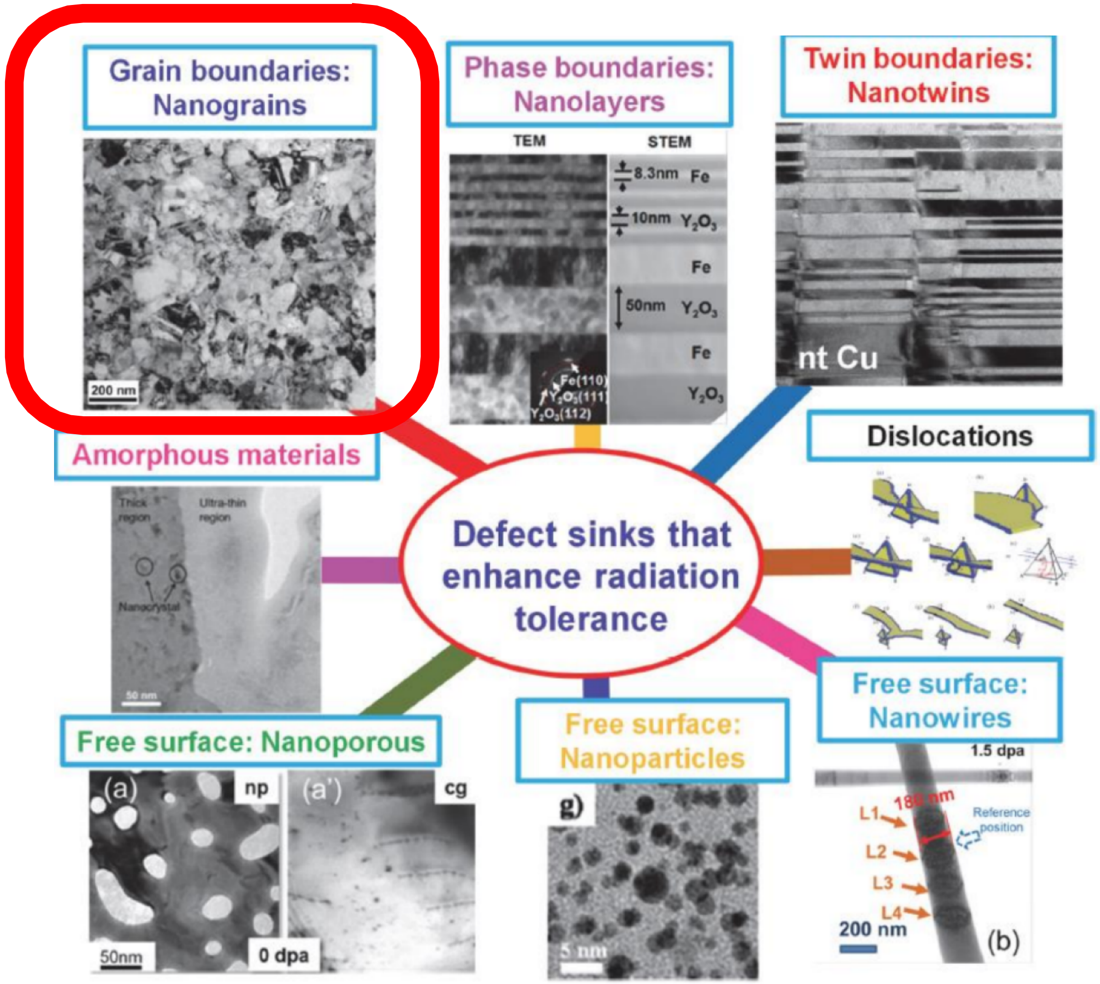
February 28th, 2022

This work was supported by the United States (U.S.) Department of Energy (DOE) Office of Basic Energy Sciences (BES), Division of Materials Science and Engineering.

U.S. DEPARTMENT OF ENERGY NNSA National Nuclear Security Administration

Sandia National Laboratories is a multimission laboratory managed and operated by National Technology & Engineering Solutions of Sandia, LLC, a wholly owned subsidiary of Honeywell International Inc., for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA0003525.

How do nanomaterials respond to irradiation?



Zhang et al., 2017

- Nanostructured metals provide the potential for radiation tolerant design
- Thermal, mechanical, and radiation properties are related to microstructure
- Non-equilibrium nanograined metals can exhibit very unique and unexpected microstructures/defect structures
- **Change in phase/crystal structure can result from irradiation exposure**

FCC to HCP Transformations in Nanostructured Materials

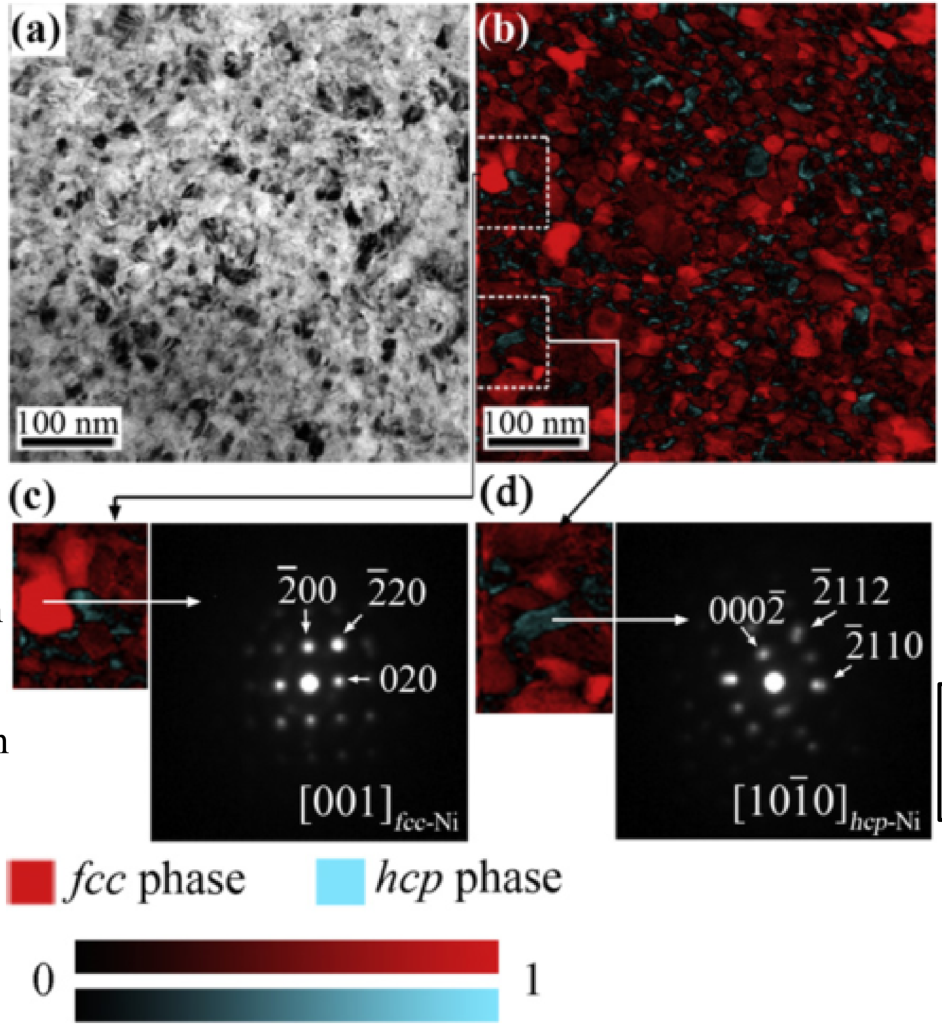
HCP in electro deposited NC Ni

(a) Bright-field TEM image of a 50 nm thick film

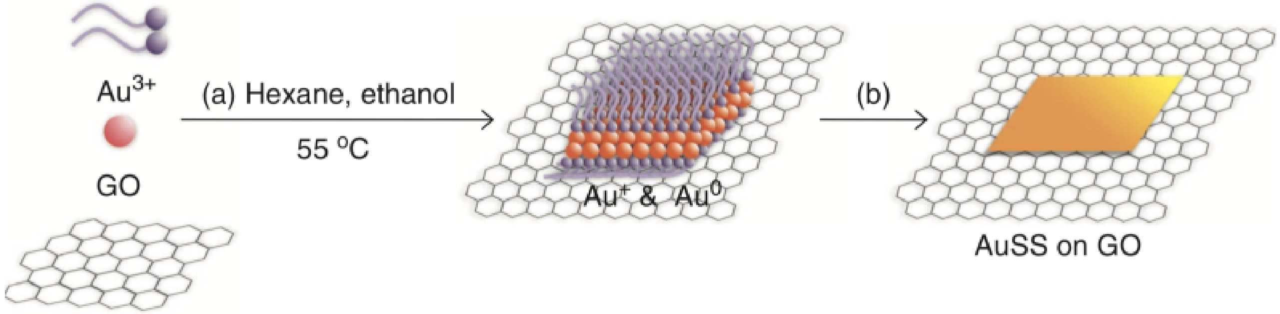
(b) Combined phase and reliability maps

(c) SAED pattern for fcc grain

(d) SAED pattern for hcp grain



HCP Au formed by nanostructure synthesis



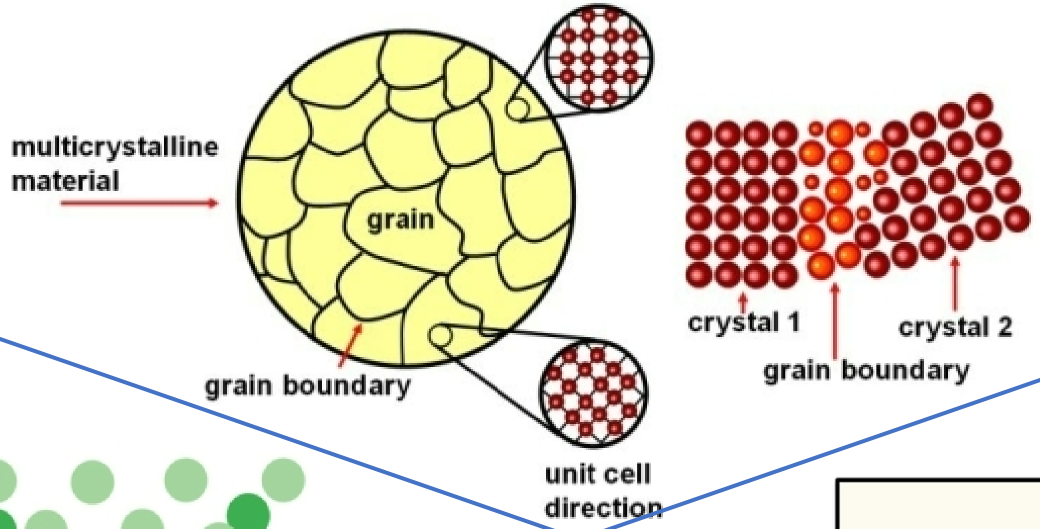
- HCP found in as deposited NC Ni thin film
- HCP Au found in nanostructures & made under high pressure

Li, et al. Acta Materialia (2016)

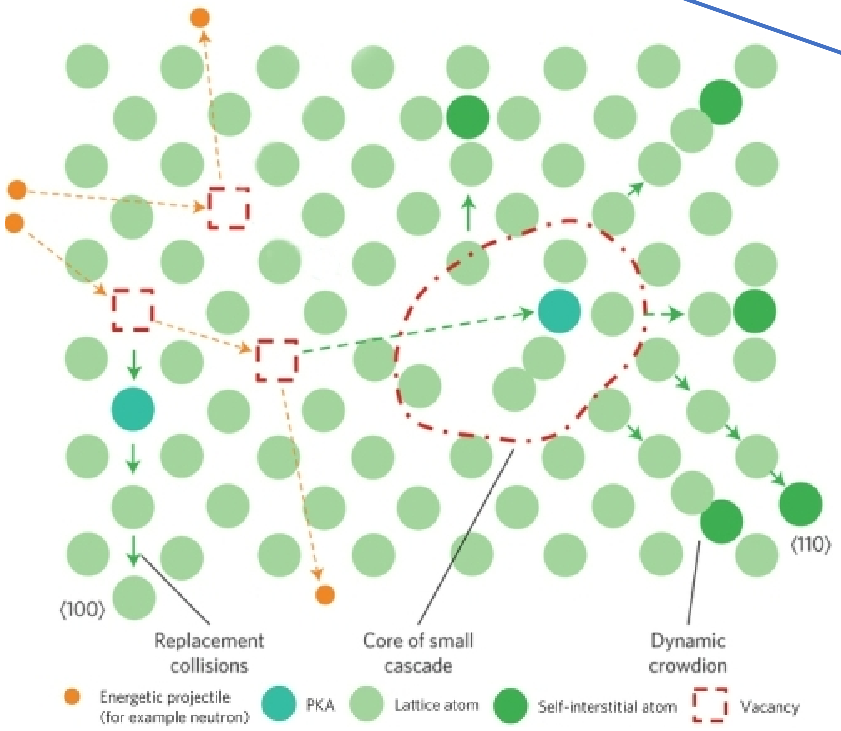
Rajasekhara, et al. (2012) Scripta Mat.
Huang, et al. (2011) Nature Comm.
Dubrovinsky, et al. (2007) PRL

Grain Boundaries, Irradiation Damage, & Defect Absorption

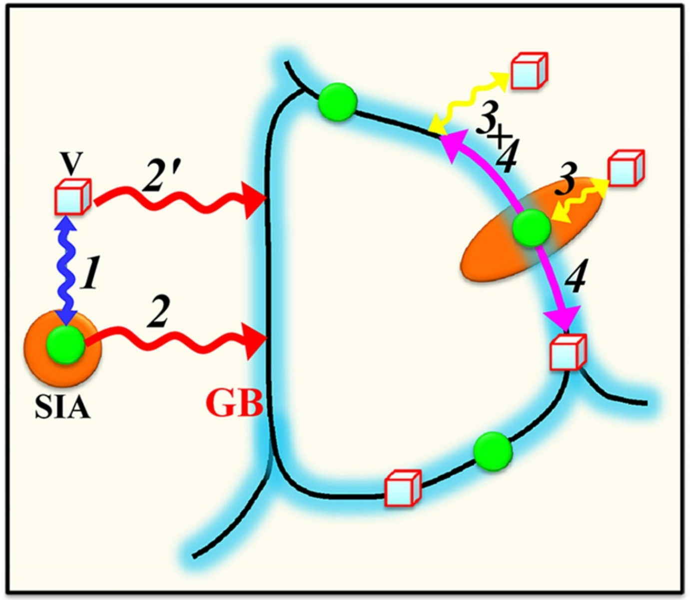
1



2



3



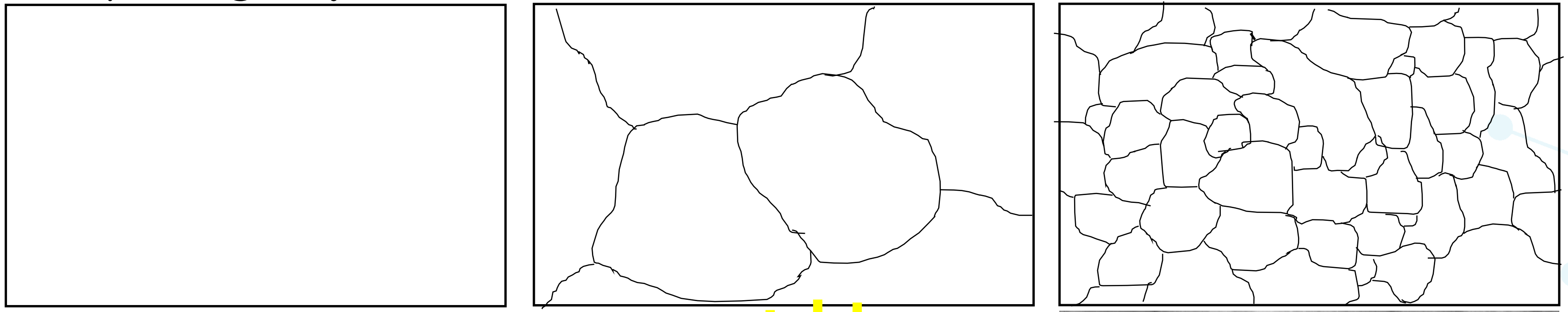
Ashkenazi, Investigating Material Failures (2007)

X. Li, et al. Acta Materialia (2016)

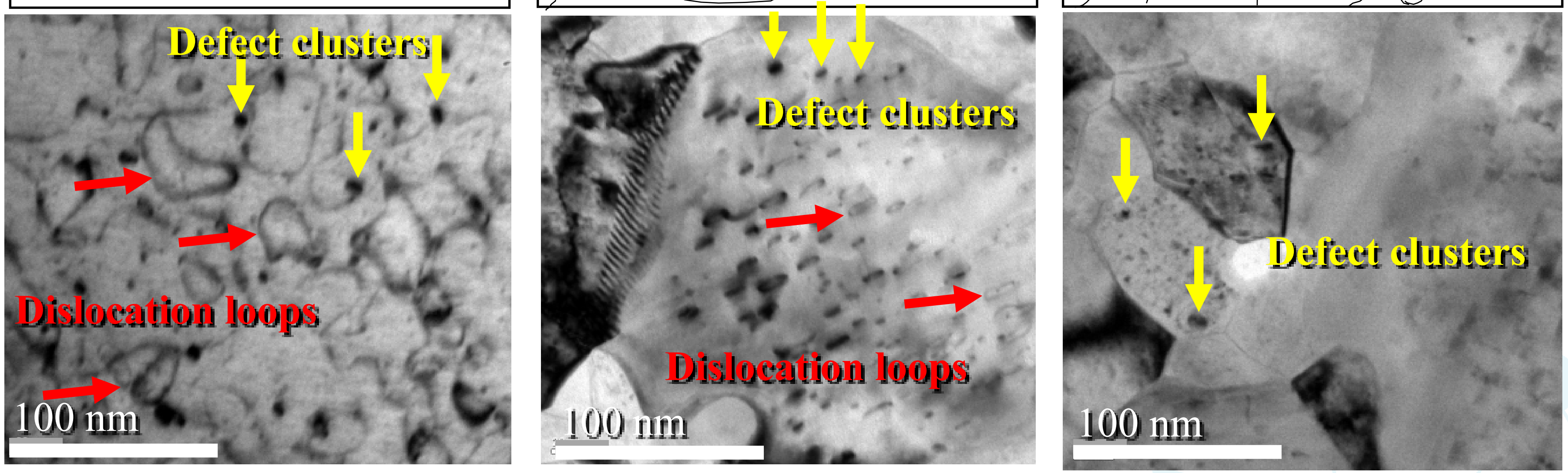
Decreased grain size leads to increased interface/GB density ► Defect sinks ► Increase defect cluster loss

	µm-single crystal	100-500nm	20-50nm

Grain size



Ex. of heavy ion irradiated Fe



No GBs	Some GBs	A Lot of GBs

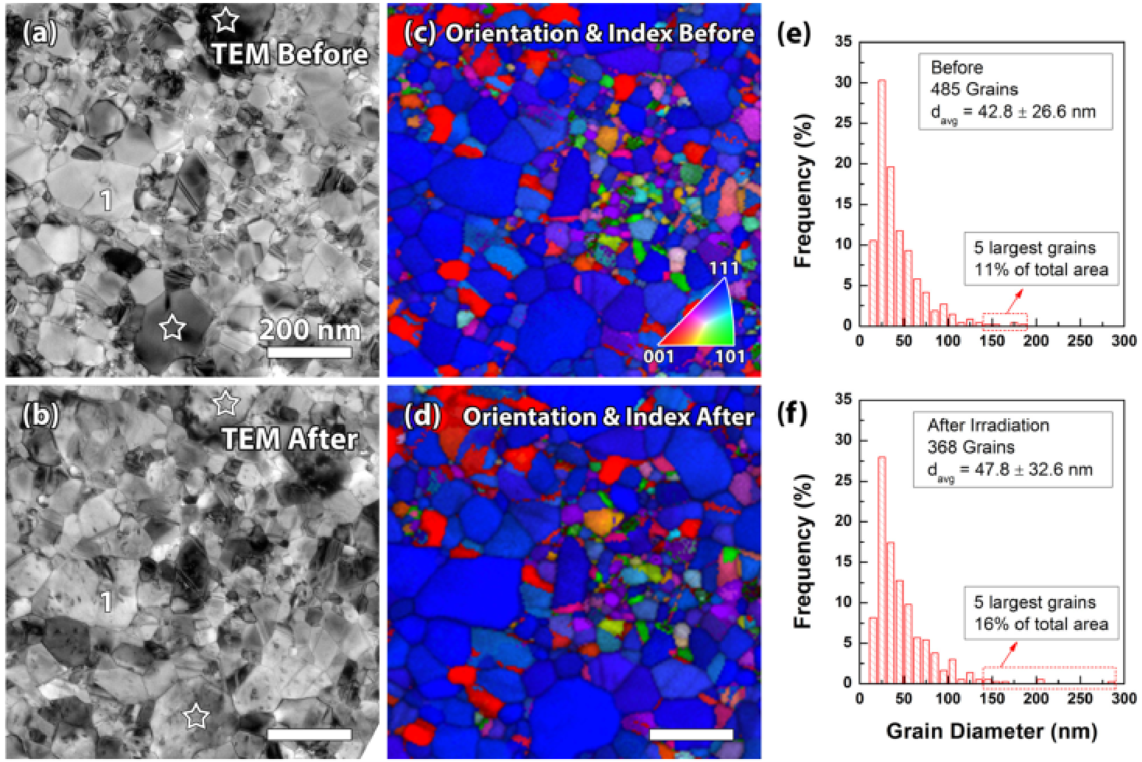
Vetterick, unpublished (2014)

Irradiation Induced Microstructural Changes in NC Metal

Irradiation induced grain growth in NC Au

Before irradiation

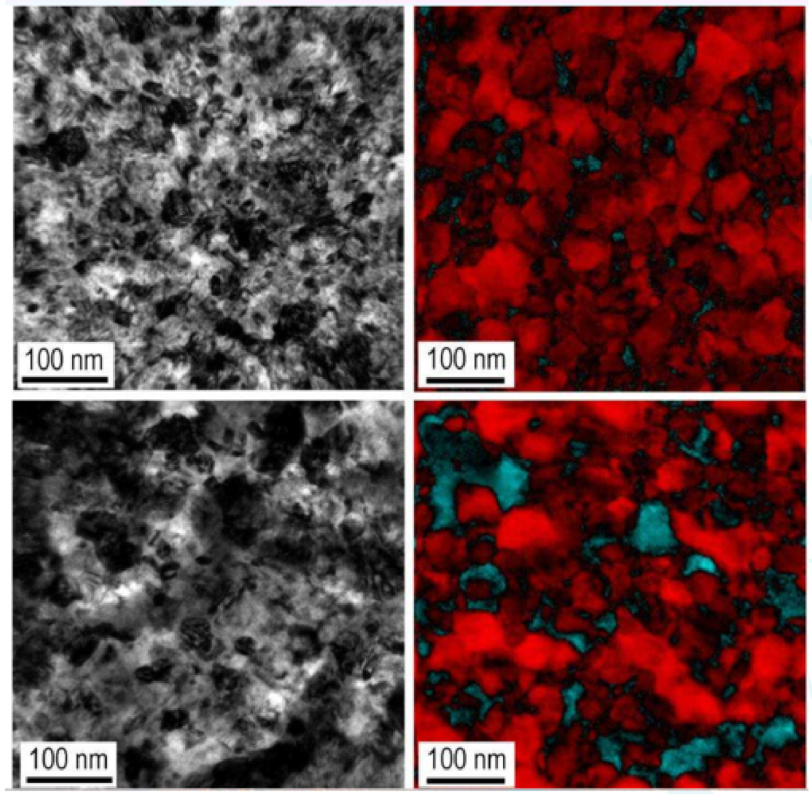
10 MeV Si (0.35 dpa)



Irradiation induced grain growth NC Ni

As deposited

35 MeV Ni (10 dpa)



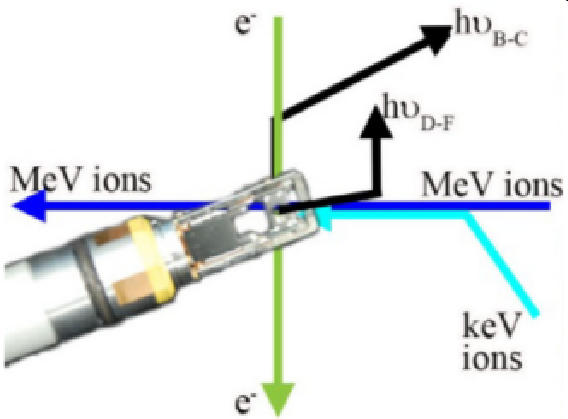
- Irradiation of NC Au lead to grain growth
- Irradiation of NC NiW lead to grain growth and texture evolution <111>

- From 6% HCP to 20%, increase in #
- HCP ave grain size from 8 nm to 14 nm

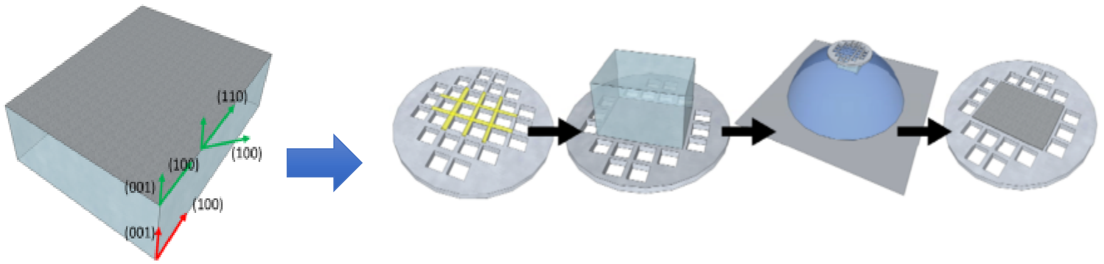
Bufford, et al. (2015), APL
Rajasekhara, et al. (2013) AIP Conference Proceedings
Singh (2017) IEEE Int. Con. On Nanotech. Proceedings

Approach: In-Situ Ion Irradiation in TEM

NC Au thin film

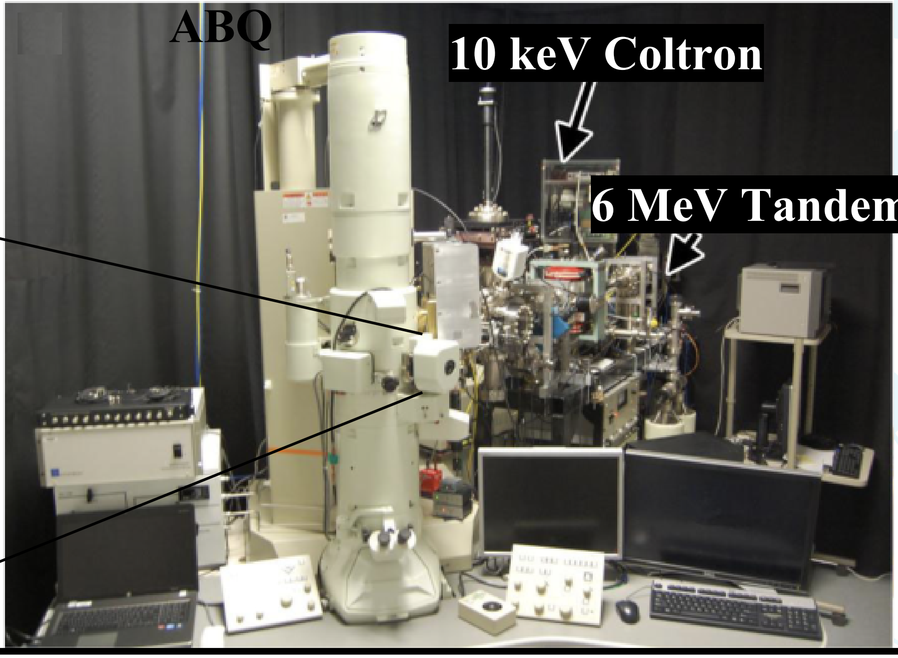


Hattar *et al*., Nuc. Int. Meth B, 2020



Films are floated off onto Mo TEM grids

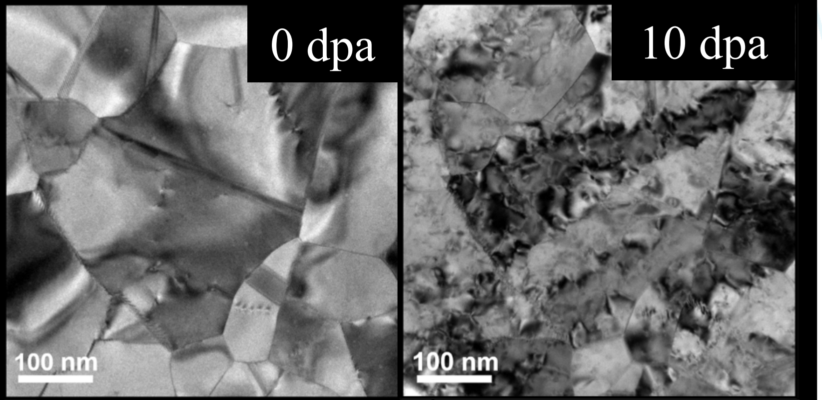
Ion Beam Lab at Sandia ABQ



JEOL 2100 LaB6 equipped with <u>NanoMegas ASTAR PED</u> system & <u>beamlines to accelerators</u>

<u>Additional HR imaging</u>

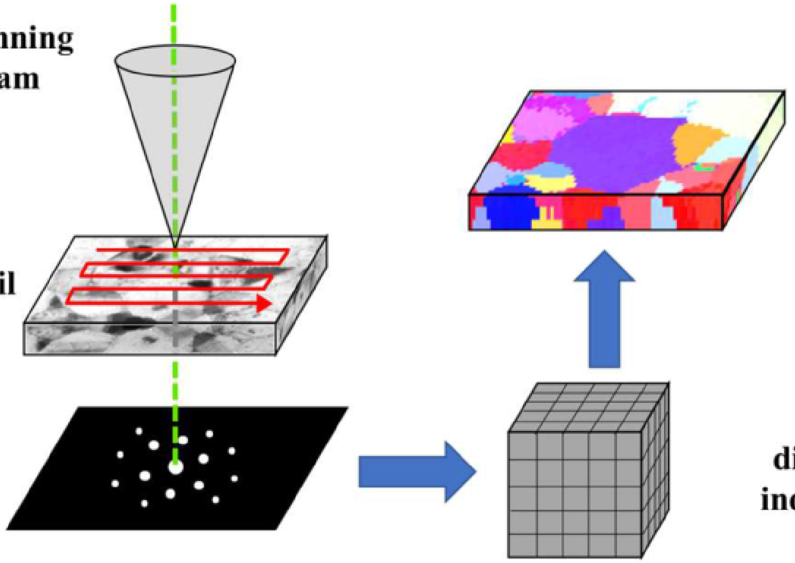
C_s-Corrected FEI Titan G2 (S)TEM



Suri *et al*., Sci Reports, 2020

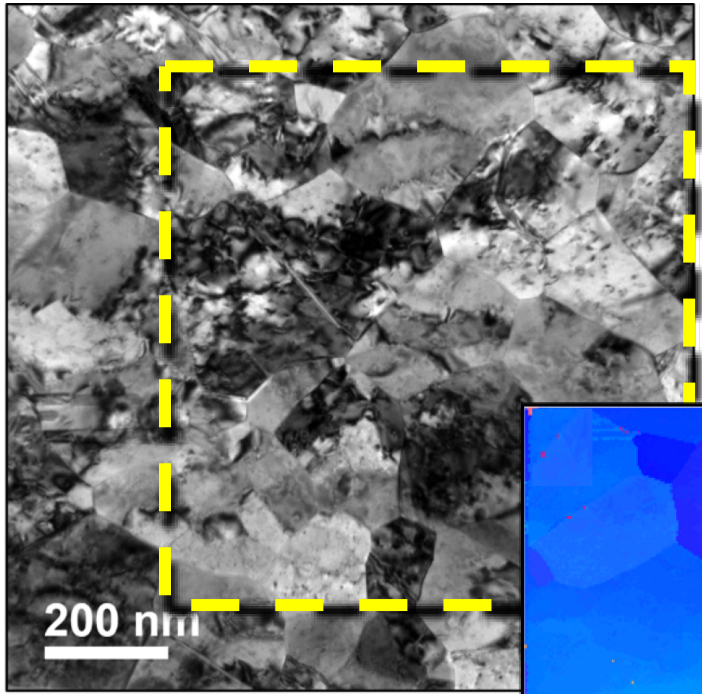
TEM Microstructural Characterization

Automated Crystal Orientation Microscopy (ACOM)

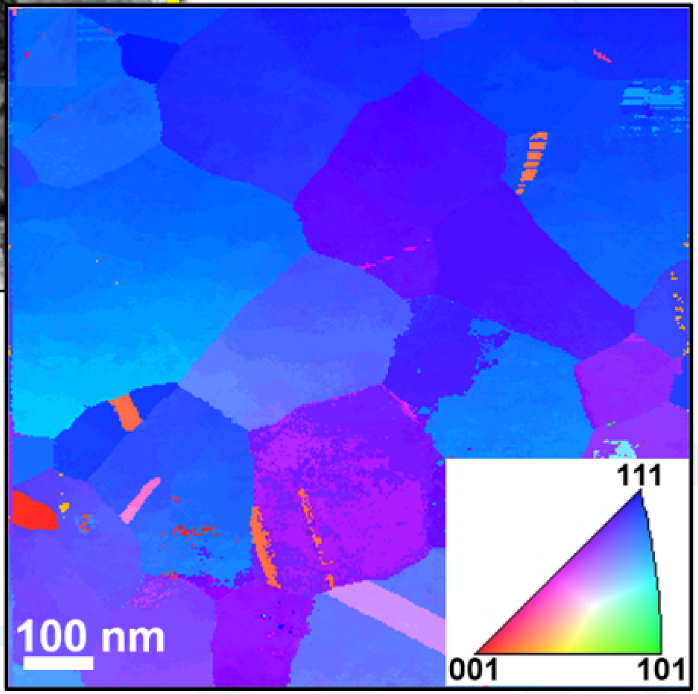


Nathaniel et al (2019) Ultramicroscopy

BF TEM



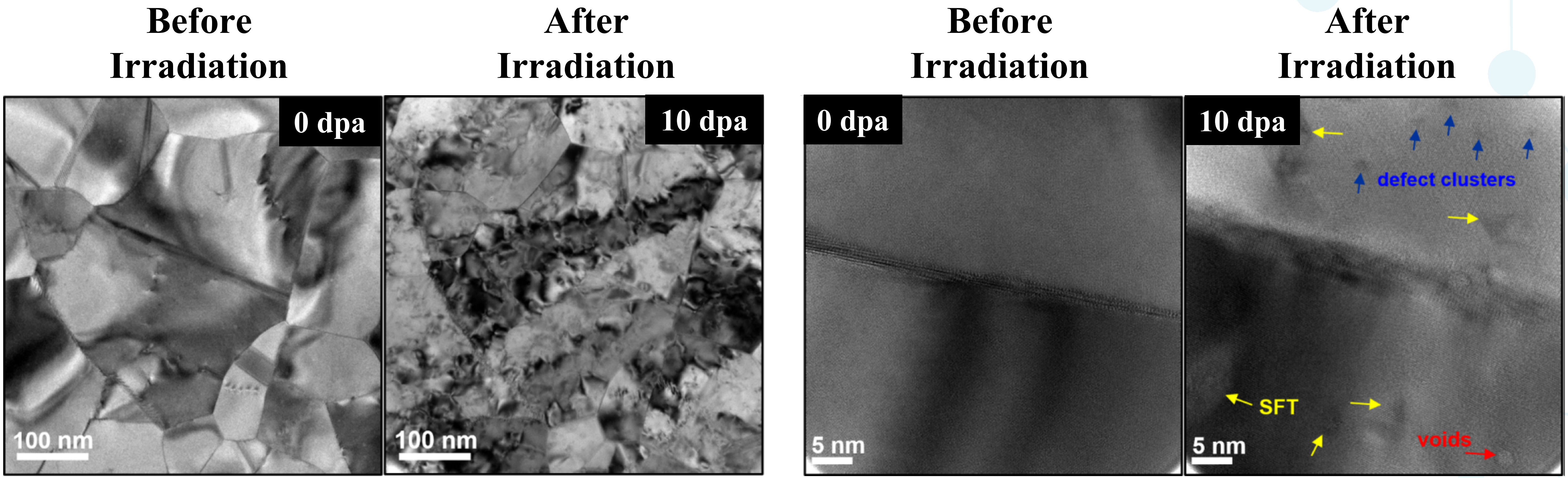
Orientation map



Suri et al (2020) Sci. Reports

- NanoMegas ASTAR precession electron diffraction w/ TopSpin acquisition system was used for crystal orientation and relative strain mapping
- Nano-electron beam is rastered over a selected area and the diffraction patterns are recorded; spot patterns undergo automated indexing/template matching
- Digitized microstructures are constructed from the diffraction data arrays

Microstructural & Defect Morphology Evolution Imaging

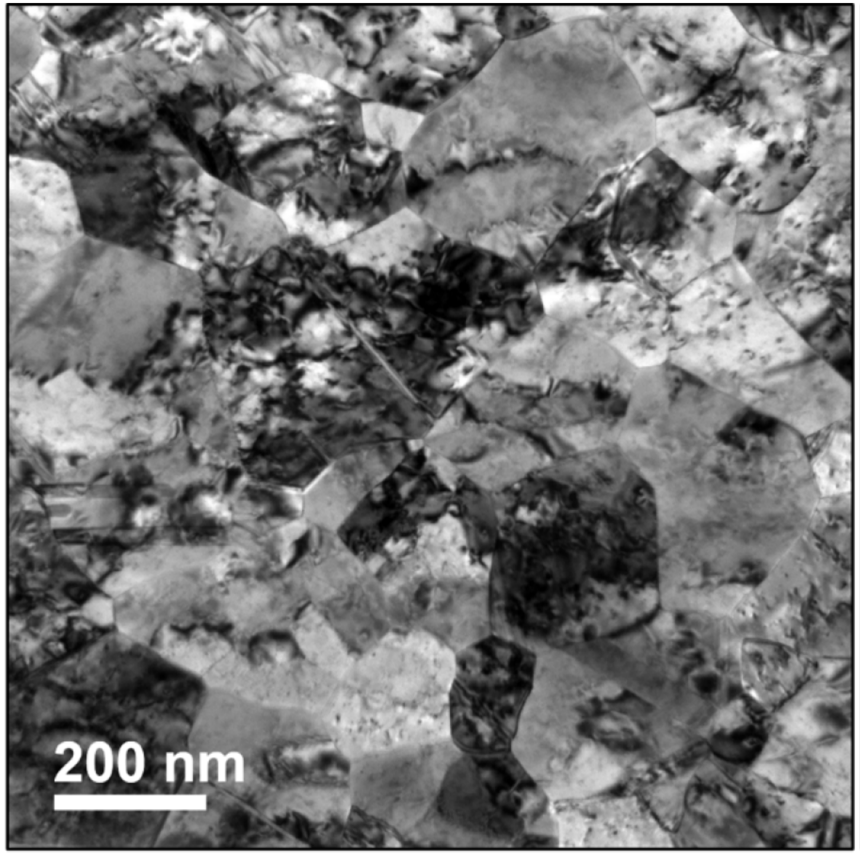


- Sample was irradiated w/ 2.8 MeV Au ions @ 200C to 4.1×10^{14} ions/cm^2 (10 dpa)
- Pre & post-irradiation HR imaged w/ C_s-Corrected FEI Titan G2 (S)TEM
- Post-irradiation microstructure did not exhibit substantial grain growth; however, significant deformation was imparted by radiation induced defects
- Stacking-fault tetrahedron and voids decorate the grain matrices and adjacent to GBs

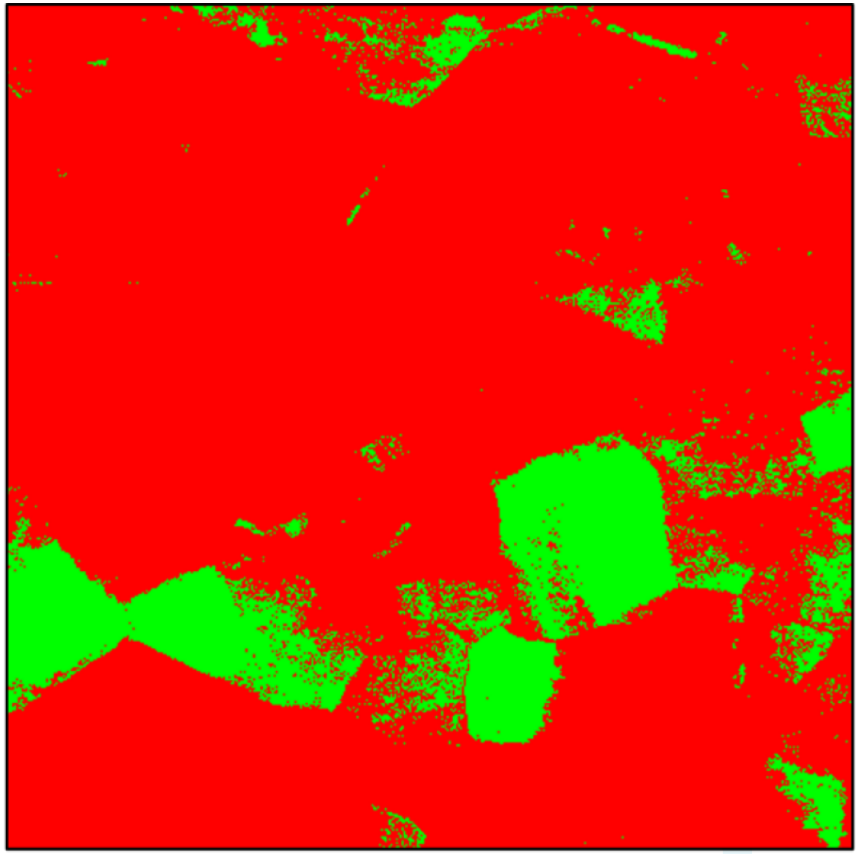
Suri *et al.*, Sci Reports, 2020

Microstructural Characterization

Post irradiation TEM image of ROI



PED phase map



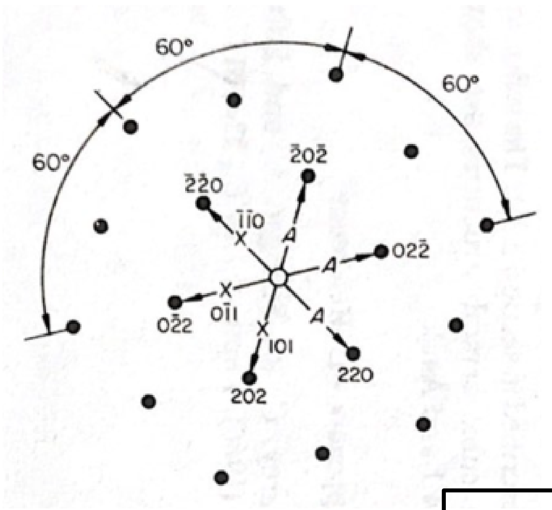
Red – HCP **Green - FCC**

Suri *et al.*, Sci Reports, 2020

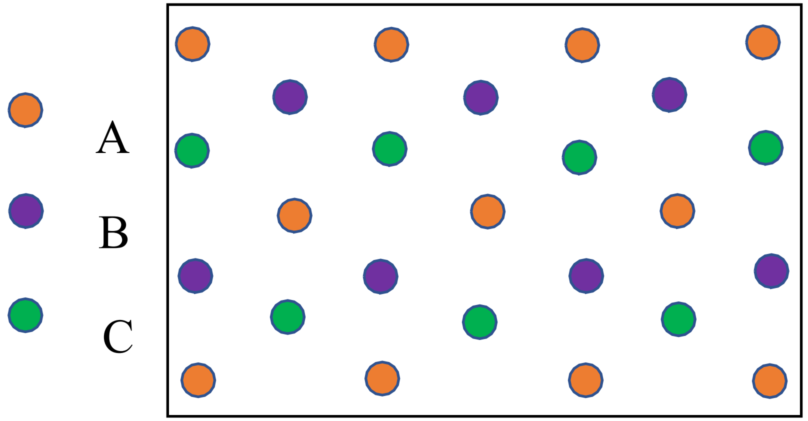
STEM Microstructural Characterization

FCC, [111]

Single crystal diffraction pattern

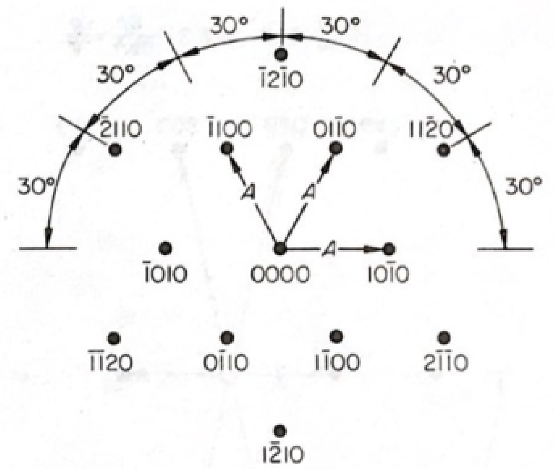


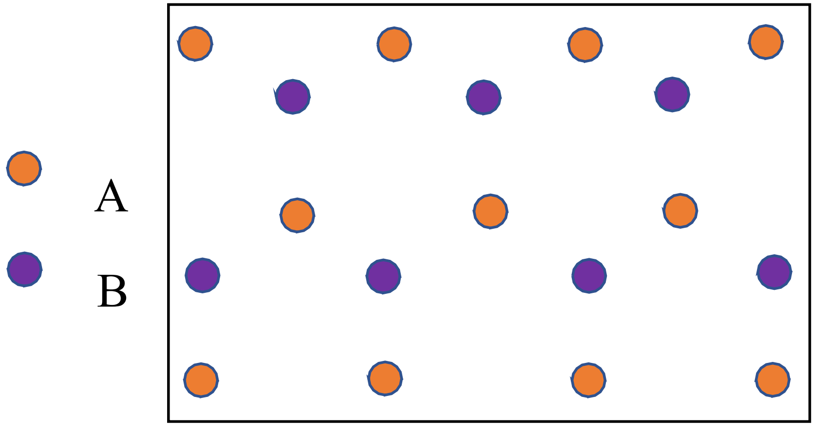
Schematic of down axis projection



A, B & C sites

HCP, [0001]

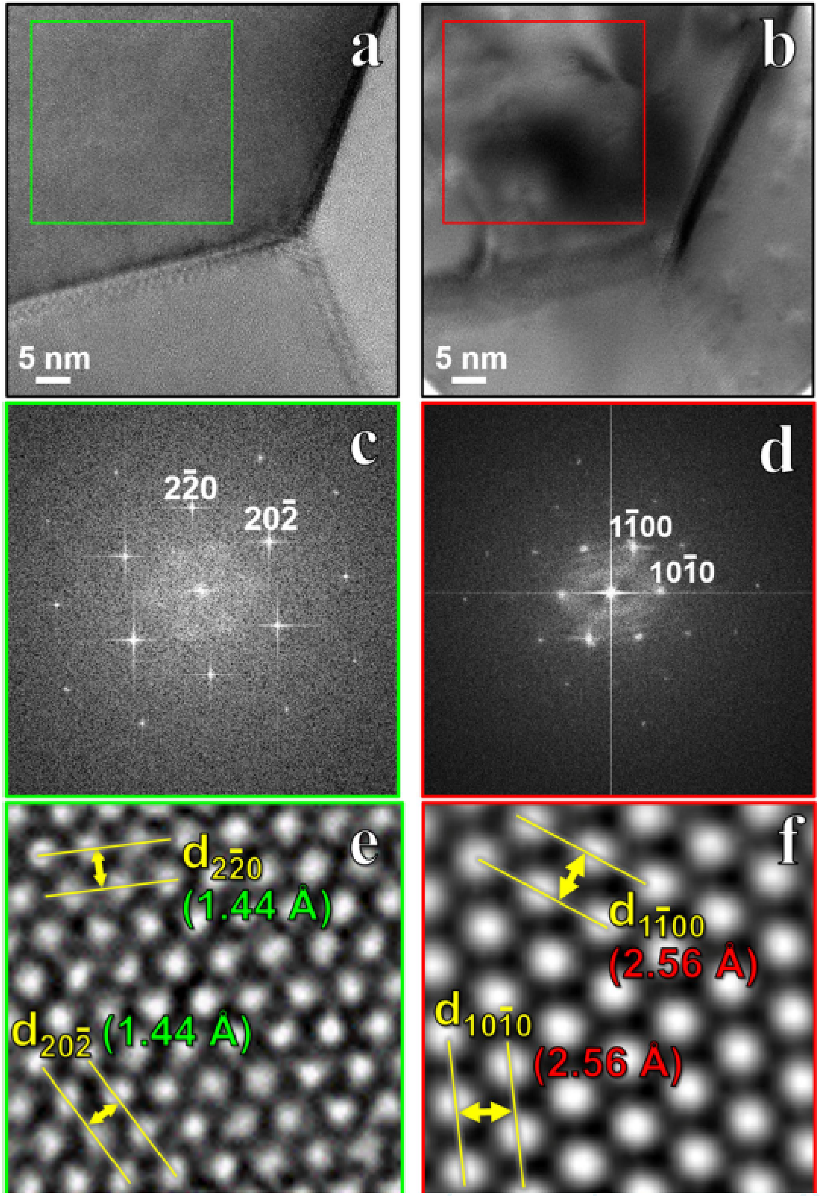




A & B sites only

HR STEM

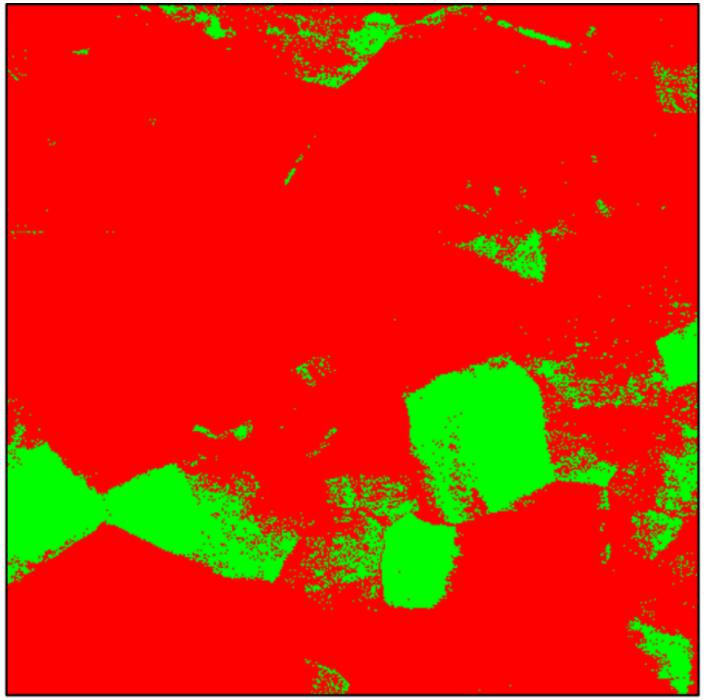
0 dpa *10 dpa*



Suri *et al.*, Sci Reports, 2020

Microstructural Characterization

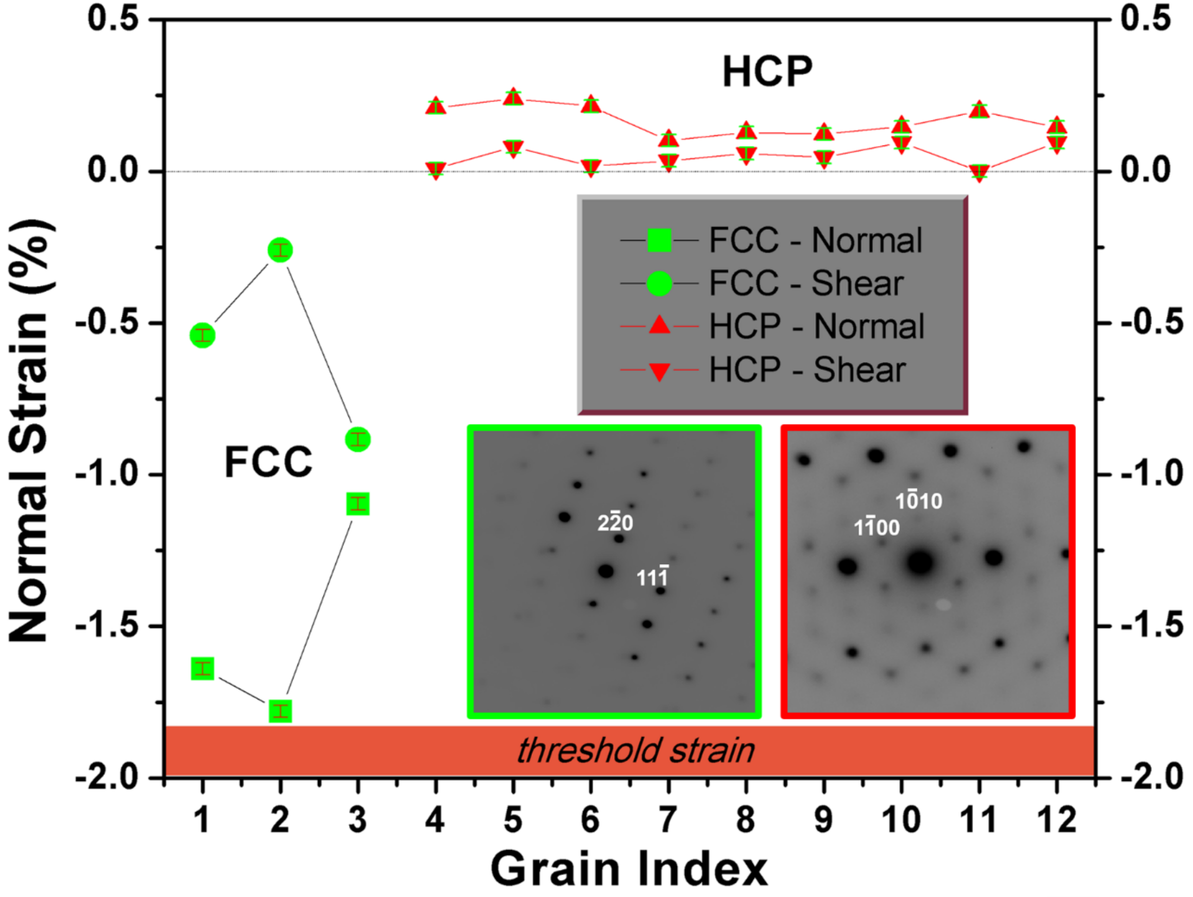
PED phase map



Red – HCP **Green - FCC**

Measured average normal and shear strain in FCC & HCP grains using TopSpin

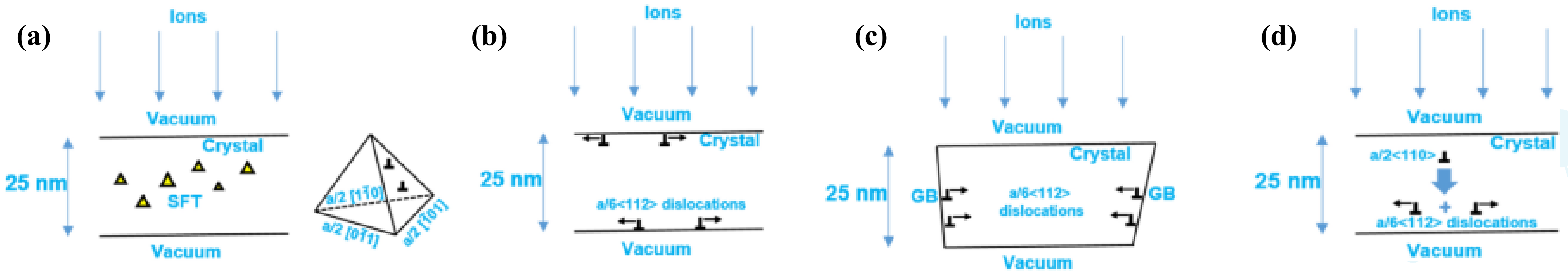
Strain Analysis



Average normal (Max [ε_{xx}, ε_{yy}]) and shear (ε_{xy}) strain plotted for the HCP and FCC phases

Suri *et al.*, Sci Reports, 2020

What is the mechanism that induces the FCC to HCP phase transformation in irradiated nanocrystalline metals?



The propagation of **a/6** <112> Shockley partials is predicted as what's changing the atomic layer packing thus transforming FCC to HCP. We proposed a few mechanisms for the origin of these dislocations:

(a) Formation of SFT, nucleation of Shockley partials on SFT faces

(b) Nucleation of **a/2** <110> dislocations at the crystal-vacuum interface → dissociate to Shockley partials

(c) Nucleation of **a/2** <110> dislocations at the GBs → dissociate to Shockley partials

(d) Nucleation of **a/2** <110> dislocations in the crystal matrix → dissociate to Shockley partials

Suri *et al.*, Sci Reports, 2020

Summary/Conclusions

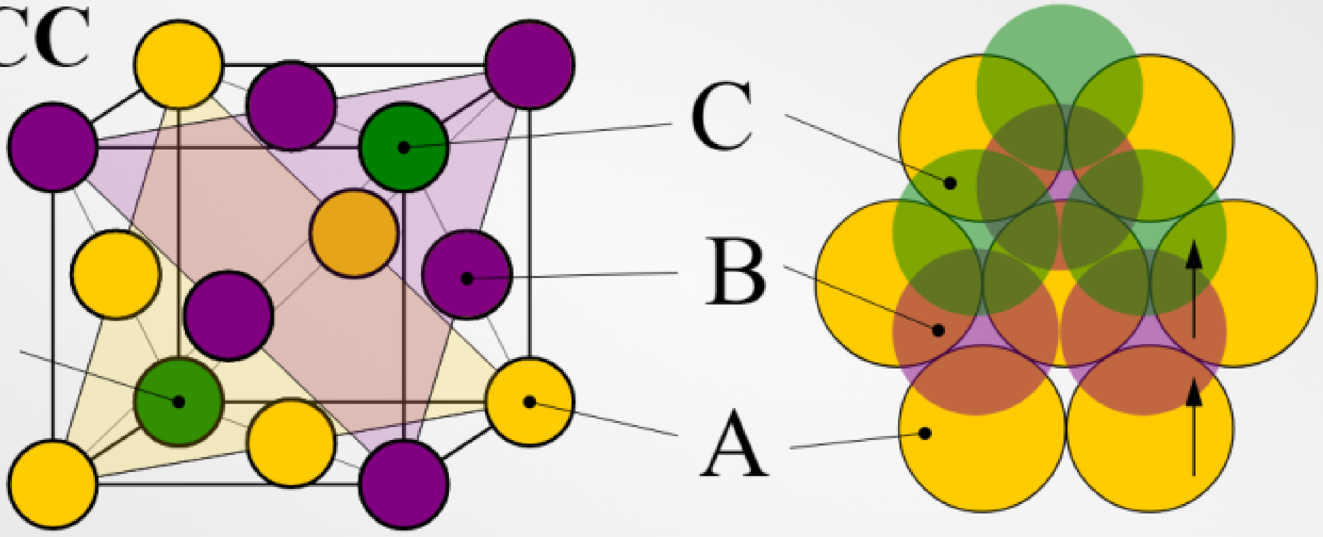
- First documented instance of irradiation induced HCP in Au / HR-TEM
- **Suri, P.K., Nathaniel, J.E., Li, N. *et al.* Ion irradiation induced phase transformation in gold nanocrystalline films. *Sci Rep* 10, 17864 (2020).**
- Unique structures in nanograined material can be induced/stabilized by irradiation
- Precession electron diffraction microscopy provides a unique tool to study the grain orientations, microstructure, and properties at the smallest scale
- Thermal, mechanical, and radiation stability of nanograined metals are probably intertwined

Thank You

- Collaborators: Pranav K. Suri, Nan Li, Yongqiang Wang, Khalid Hattar, and Mitra Taheri
- Continuing research team: Doug Medlin, Ryan Schoell, Zachary Milne, Eric Lang
- Sandia, CINT, BES, NNSA, & DOE

HCP vs FCC Stacking: ABABAB vs ABCABC

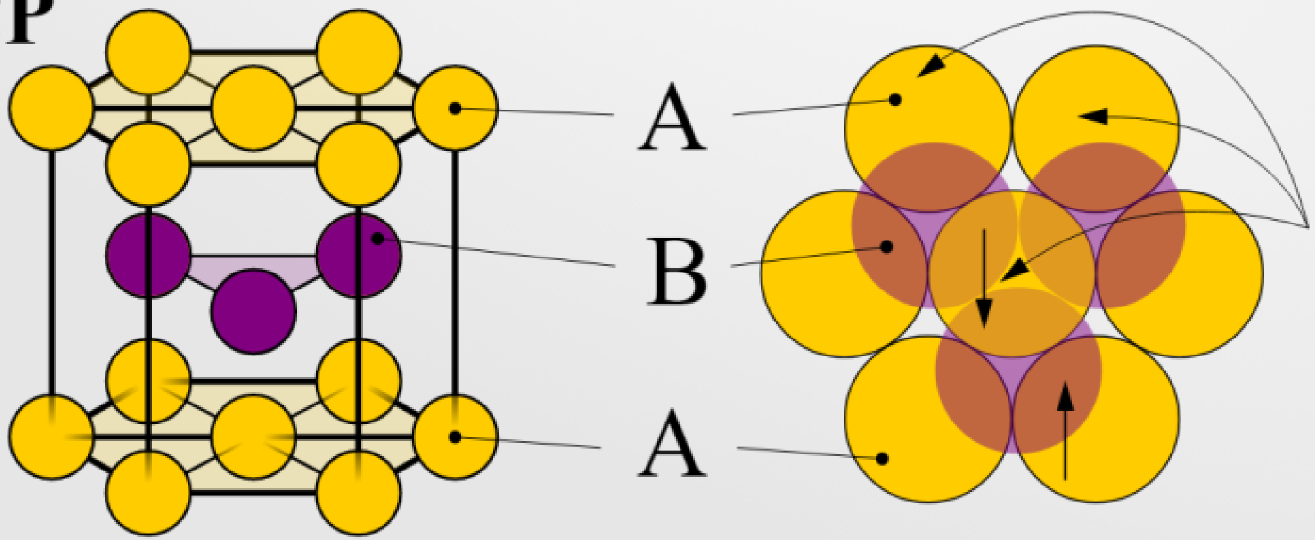
FCC



Sequence: ABCABC

There are **3 possible positions** for a close-packed layer. If **all 3** positions are used in ABC pattern, this makes an **FCC** crystal. This is hard to see because the close-packed planes are along the body diagonal.

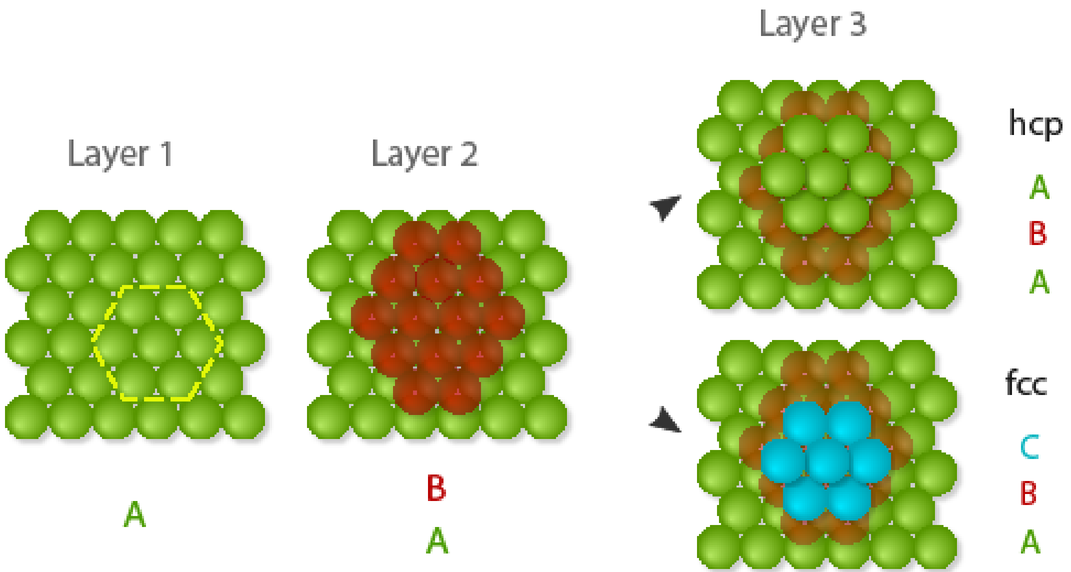
HCP



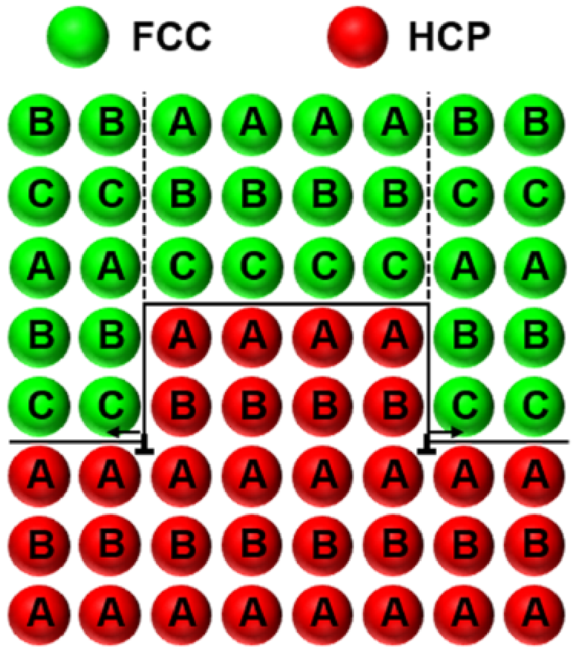
Sequence: ABABAB

The stacking is easier to see in HCP because the layers stack within the unit cell. For an HCP crystal, **only 2** of the 3 possible positions are used, so the third layer is a repeat of the first layer.

Irradiation-induced microstructural changes in nanocrystalline metals



Schematic of atomic layer packing change from ABCABC… (FCC) to ABABAB… (HCP)



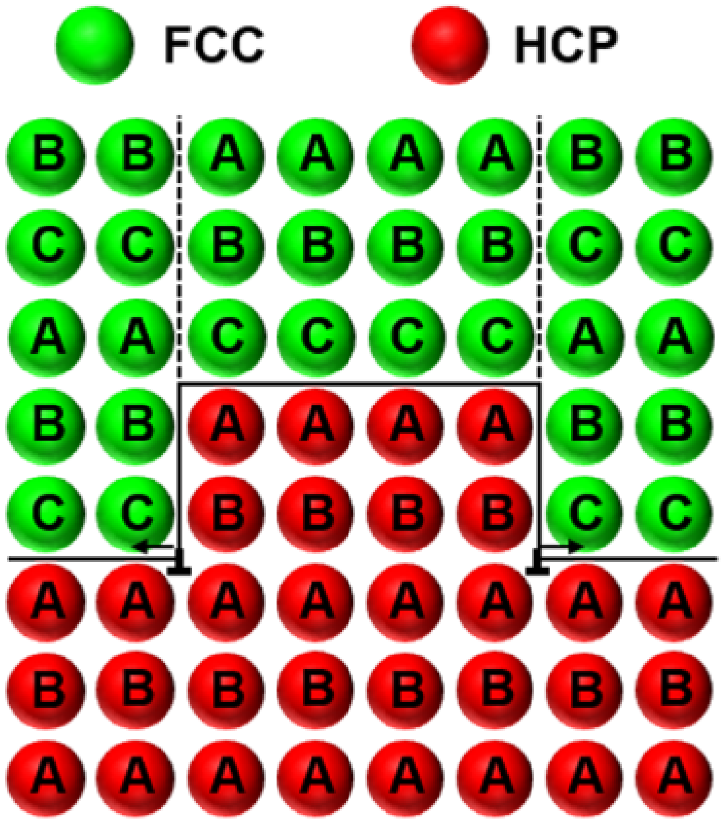
Propagation of **a/6**<112> Shockley partials transforming two layers of FCC to HCP

<u>Though this phenomena has been documented, the mechanism(s) of transform have yet to be identified</u>

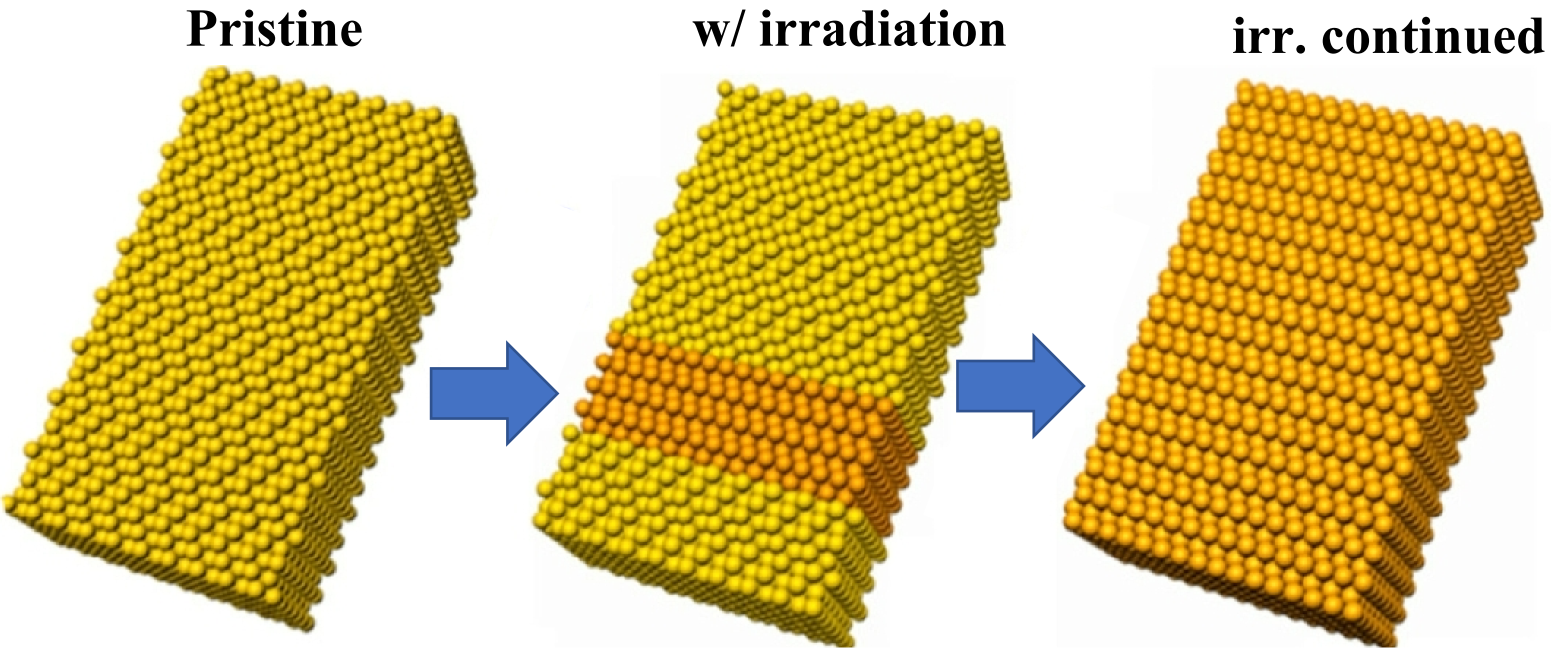
physicsinanutshell.com, solid state physics – common crystal structures

Suri *et al.*, Sci Reports, 2020

Schematic of atomic layer packing change from ABCABC… (FCC) to ABABAB… (HCP)



Propagation of **a/6**<112> Shockley partials transforming two layers of FCC to HCP



Yellow: FCC Orange: HCP