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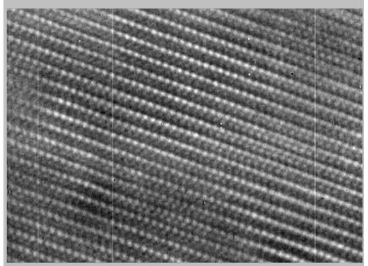

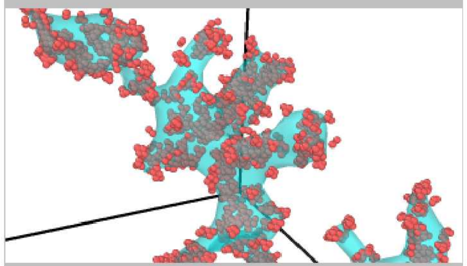

Characterization of Single Ion Strikes using In-Situ Dynamic Transmission Electron Microscopy

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Rio Grande Symposium
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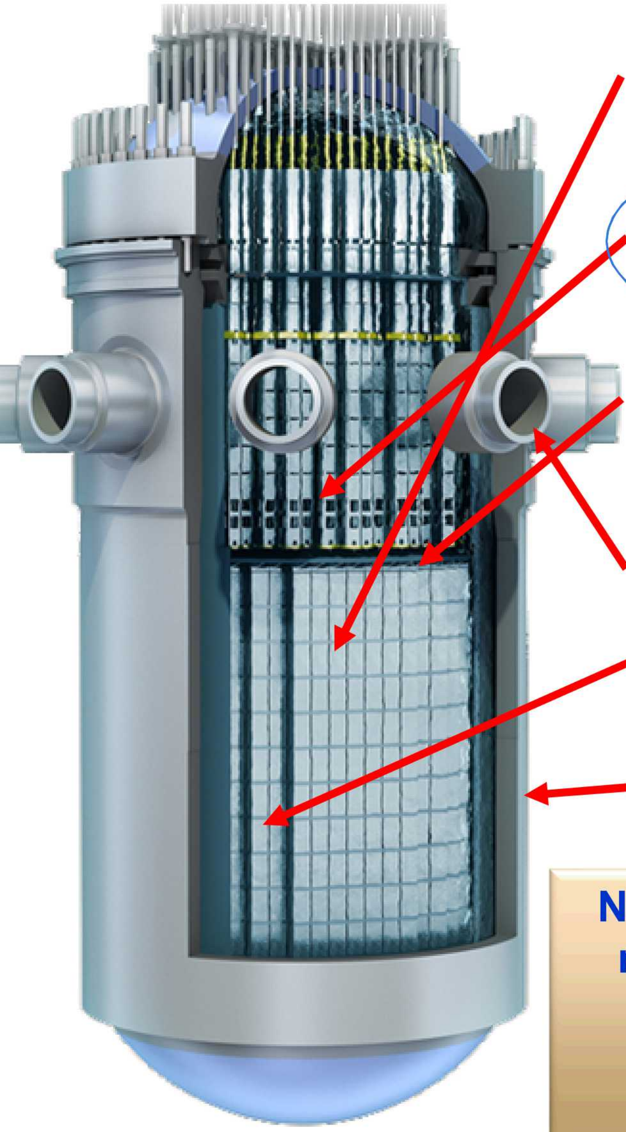
Reactor Materials Challenges



Nuclear Fuels

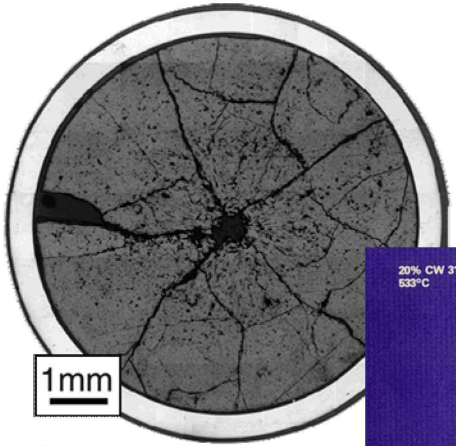
Displacement Damage Effects

Transmutation Effects

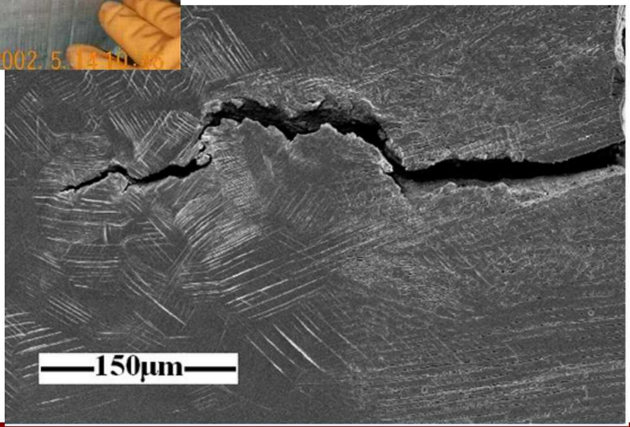
Corrosion

Creep

Fatigue

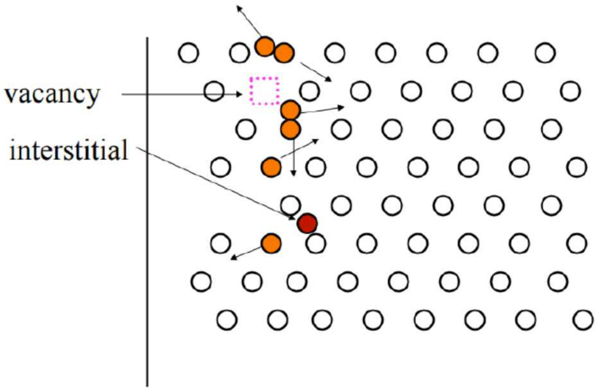


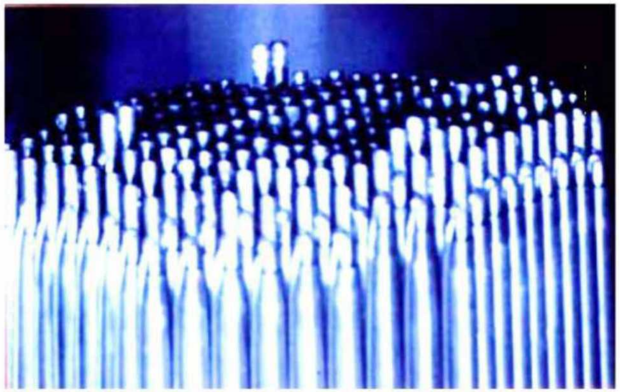
Nuclear reactors pose unique materials challenges due to degradation effects from combined environmental stressors



Radiation Displacement Damage

- Energetic particles (neutrons or ions) cause cascades of atomic displacements.
- High concentrations of interstitials and vacancies form under irradiation and agglomerate into voids, dislocation loops, etc.





Swelling in Fuel Pin Bundles

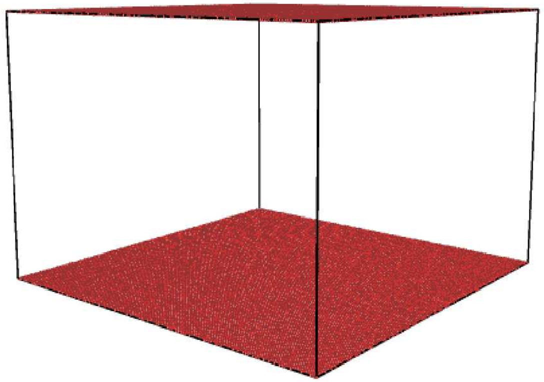
Radiation Effects on Microstructure:

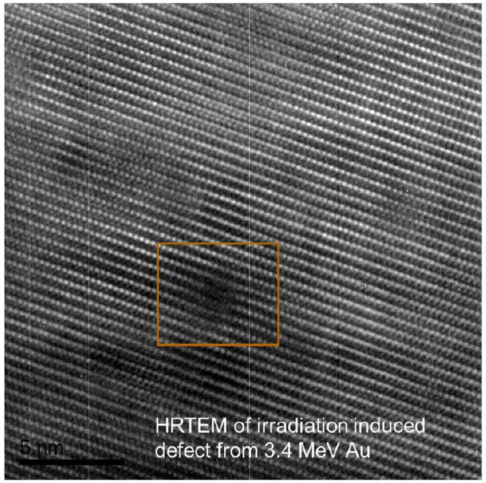
- Dislocations
- Voids and Bubbles
- Segregation
- Precipitation

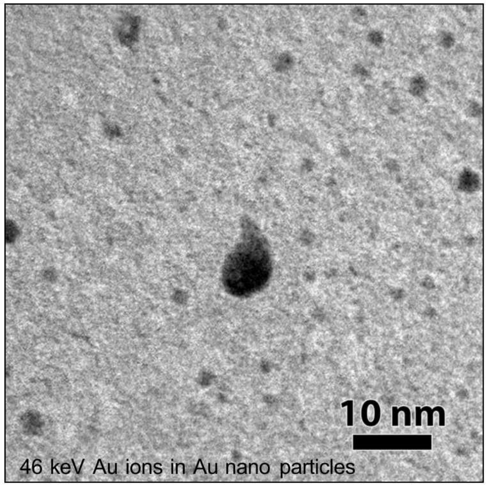
Mechanical Effects

- Irradiation Hardening/Deformation
- Fracture and Embrittlement
- Irradiation Creep and Growth
- Irradiation-Assisted Stress Corrosion Cracking (IASCC

Temporal Gap Between Modeling and Experiments







Molecular Dynamics Modeling of Cascades:
Temporal Resolution: < picoseconds
Spatial Resolution: single atoms

Experimental Observations of Radiation Damage:
Temporal Resolution: 1/30th of a second (standard camera)
Spatial Resolution: single atoms

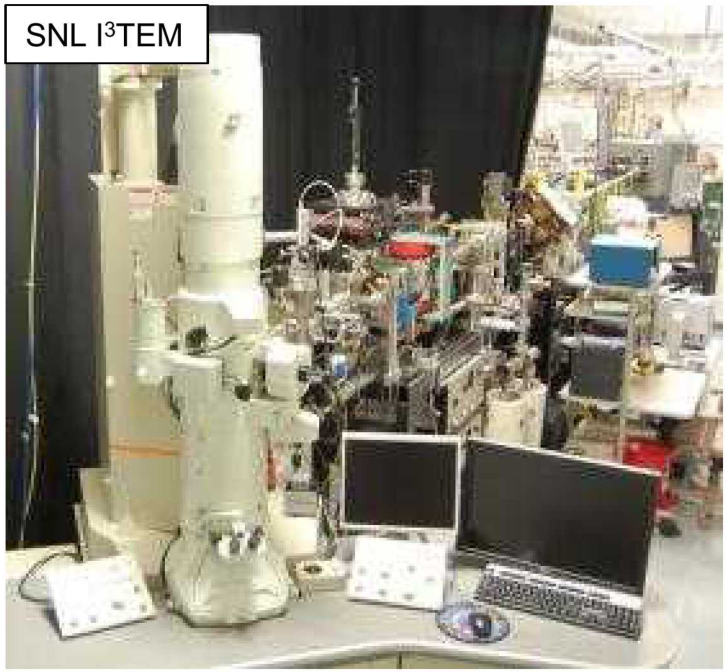
Computational models can be scaled up but is expensive and requires multidimensional physics.

Advances in experimental capabilities can be used to approach the scale of computational models.

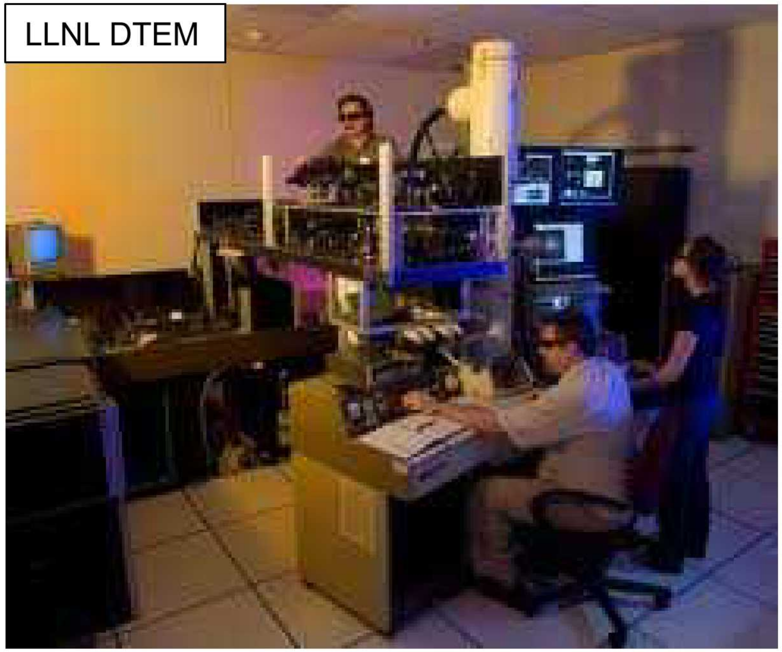
Objective

Discuss the development of **high spatial and temporal resolution microscopy** techniques that allow for characterization of **single ion strikes**.

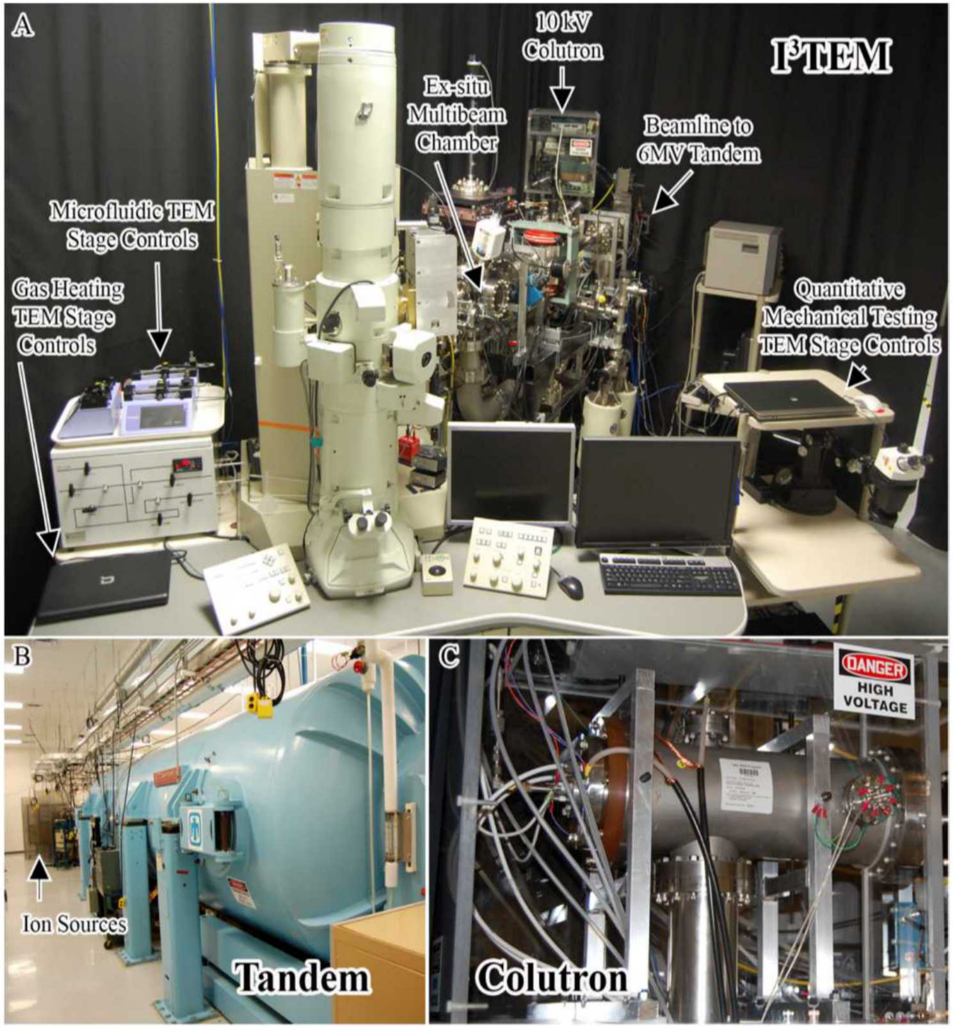
SNL I^3TEM



LLNL DTEM



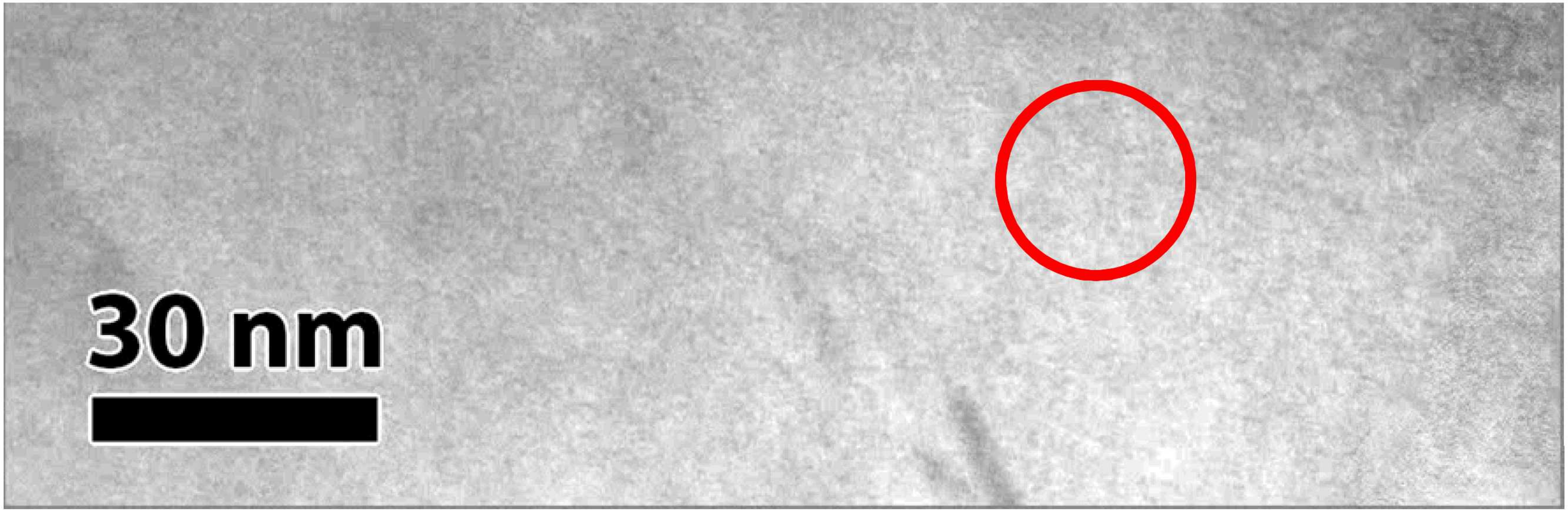
Sandia's In-Situ Ion Irradiation Transmission Electron Microscope (I^{3}TEM)

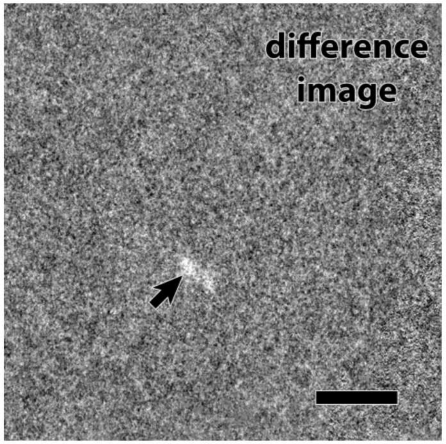


Hattar, K., D. C. Bufford, & D. L. Buller. "Concurrent in situ ion irradiation transmission electron microscope." *NIM:B* **338**, 56-65 (2014).

1.7 MeV Au ions into Au nanofilm

Single Ion Strikes Explored Experimentally





- 1.7 MeV Au into single crystal Si
- Single ion strikes can be observed in semiconductors
- Non-symmetric structure in contrast to the spherical approximation
- Effects were observed down to 46 keV Au & up to 48 MeV Si

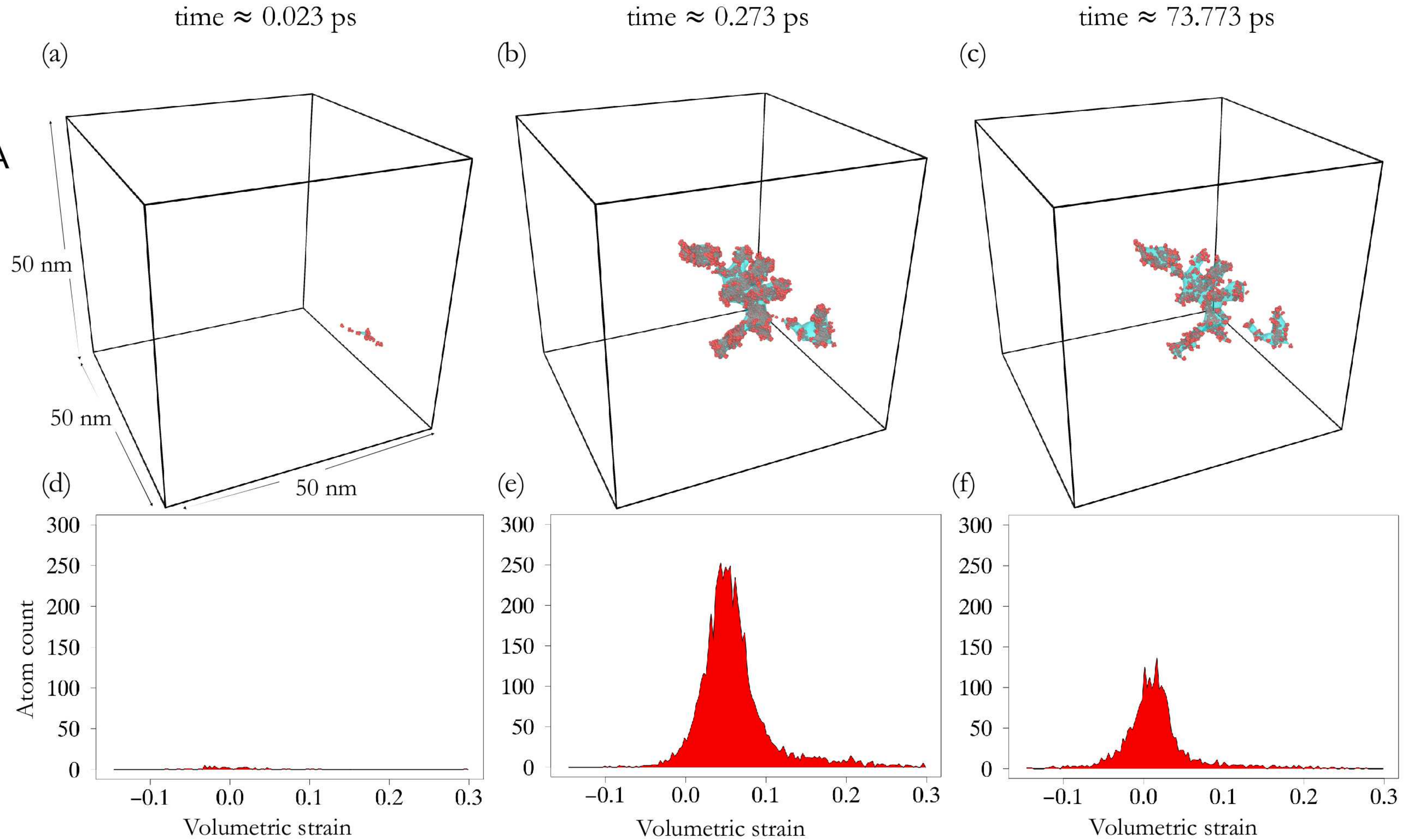
Can we go beyond this to observe:

- the important aspects of structural evolution as a function of time (ns to hrs.)?
- the evolution in more complex systems (GaAs)?
- Directly correlate it to key model parameters?

Single Ion Strikes Explored with Molecular Dynamics

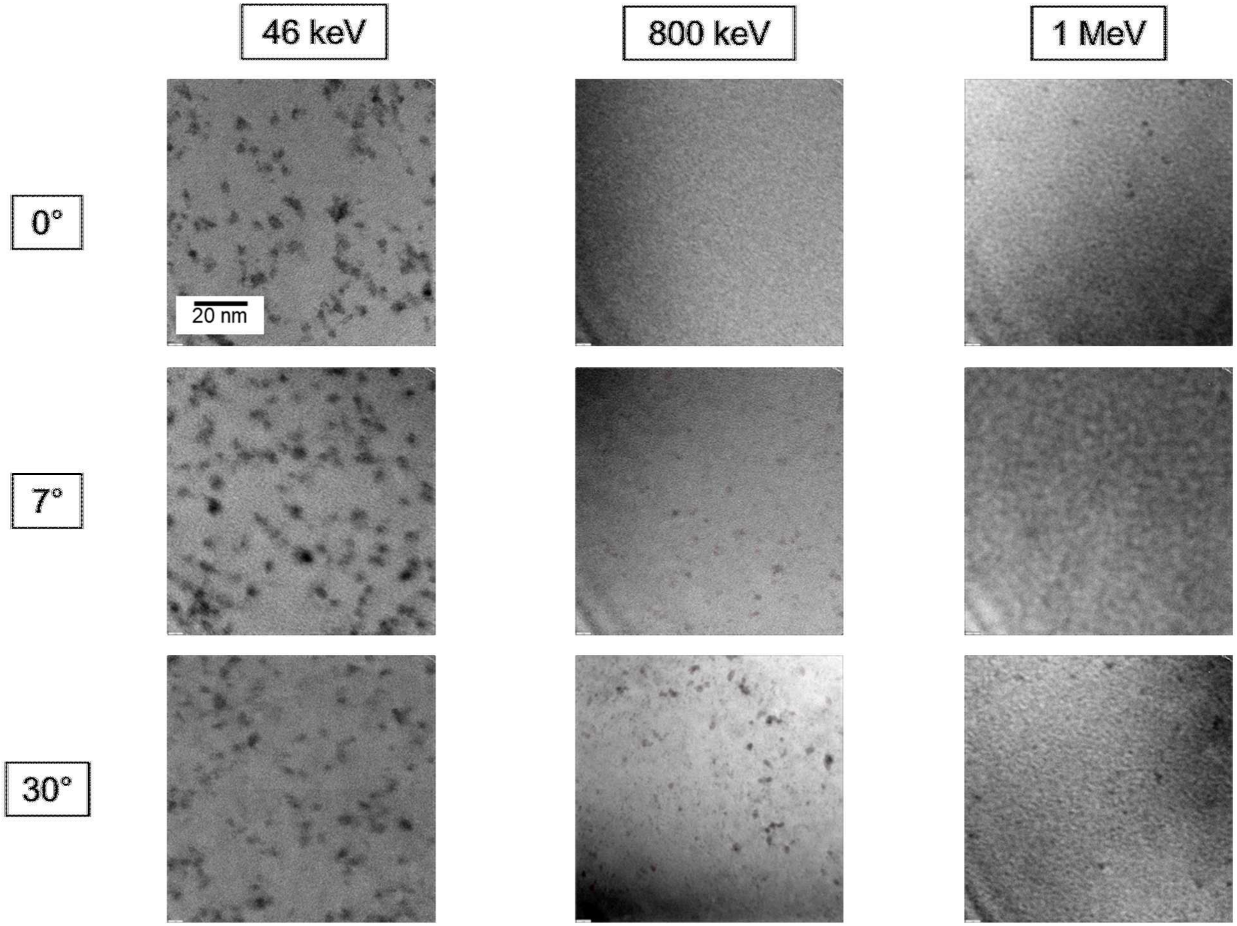
INITIATION PEAK DAMAGE DEFECT RECOMBINATION

20 keV PKA
in bulk Si



[Stewart et al. (2016) J. *Appl. Phys.*]

Black Dot Damage Distributions



Single crystal <100> Si irradiated ex-situ with Au ions at a variety of energies and angles of incidence.

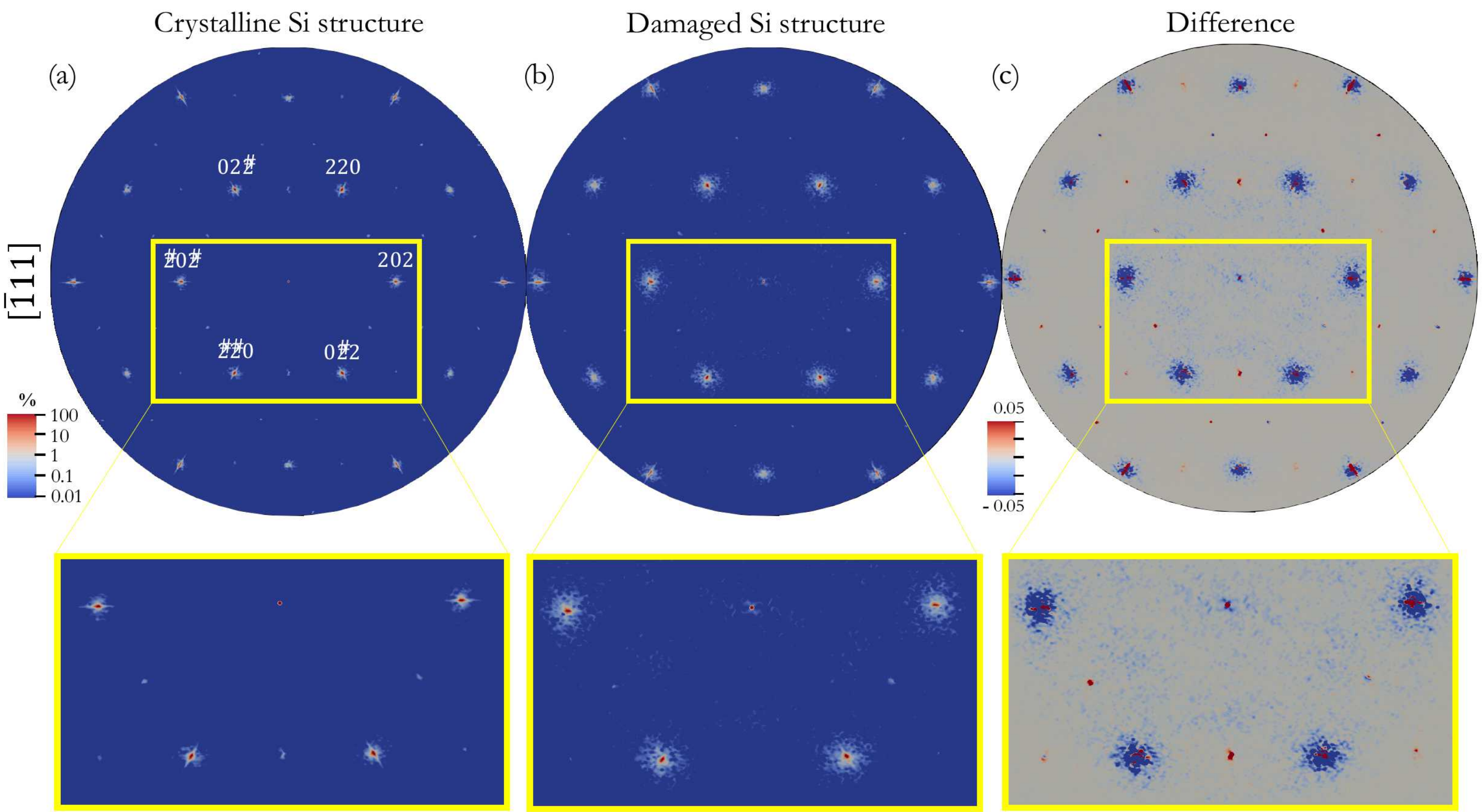
Probing the damage cascade experimentally is limited to contrast from black-dot damage.

Can a coupling of modeling and experiment be used to:

- Obtain defect distributions represented in real-space images?
- Directly correlate quantitative experimental results with modeling results?
- Account for contrast variation seen in TEM?

Using Diffraction to Compare Modeling and Experiments

It is difficult to correlate real-space images with molecular dynamics modeling. Diffraction patterns can be used by both as a more reliable method of comparison.

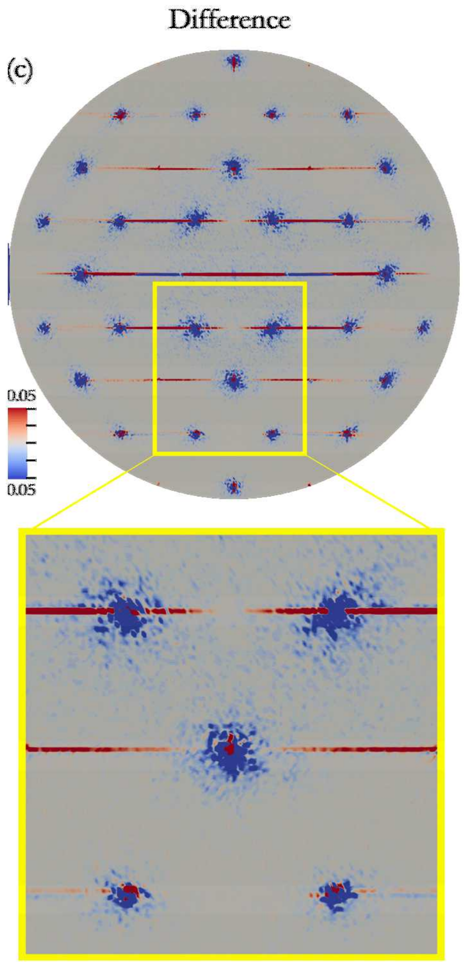


Calculated SAED patterns for [111] Si before and after a single ion cascade event

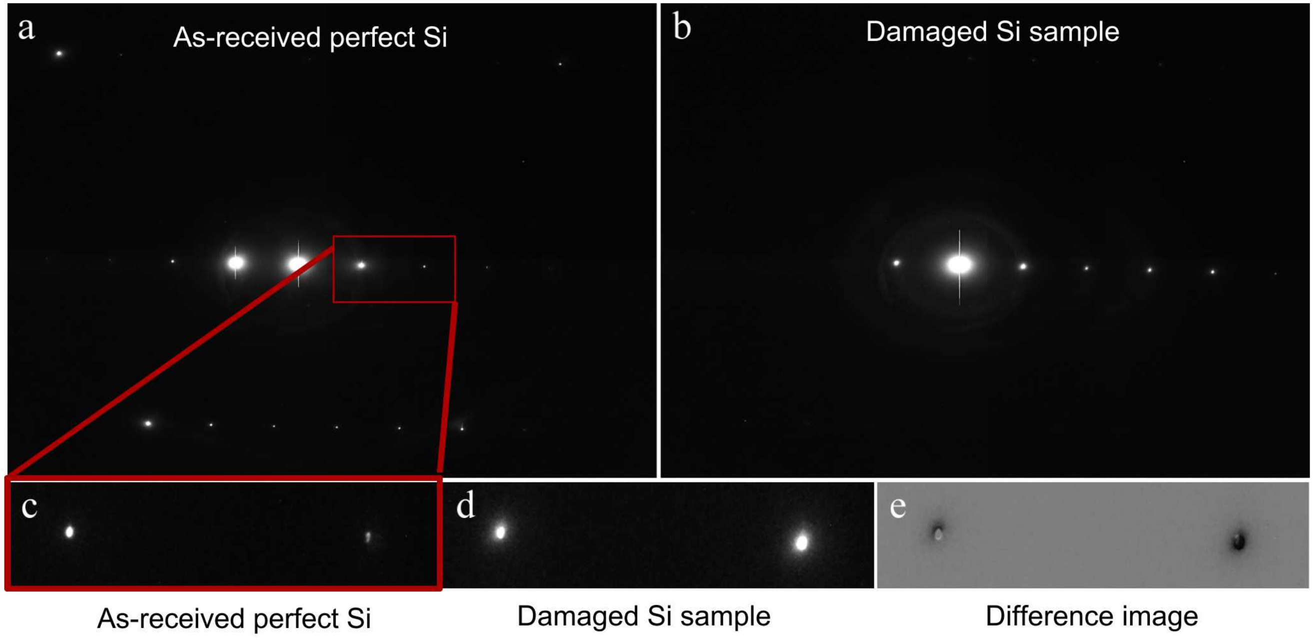
[Stewart et al. (2018) J. Appl. Phys.]

Using Diffraction to Compare Modeling and Experiments

Simulated Image



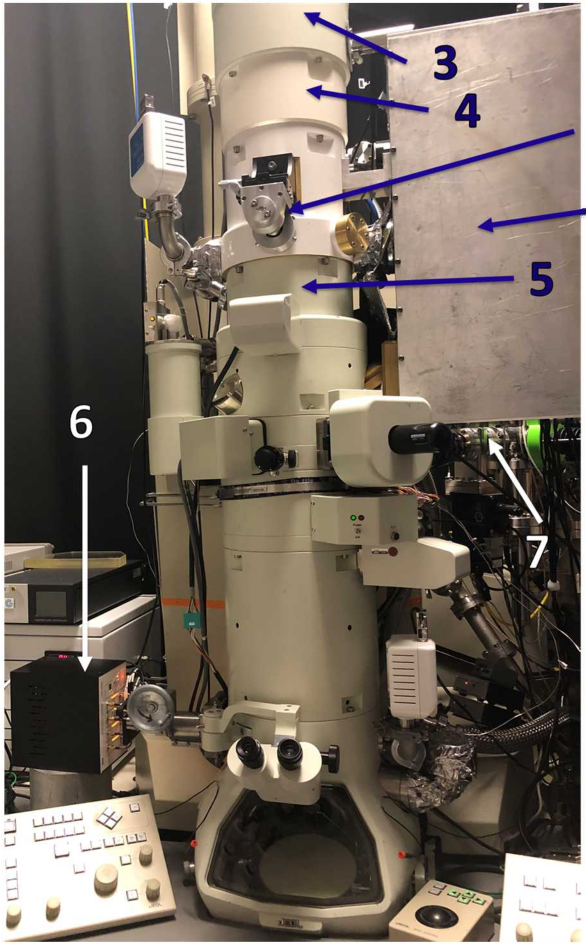
Experimental Image

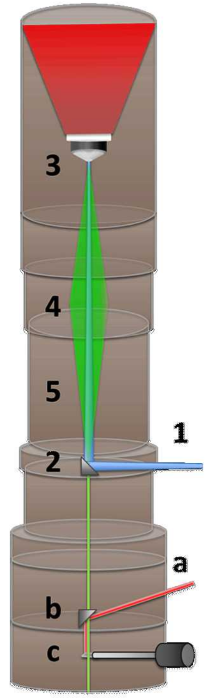


Diffraction techniques can be used as a viable comparison. But **high temporal resolution is required** to probe the damage cascade itself.

[Stewart et al. *J Appl. Phys.* (2018)]

Closing the Temporal Gap with Dynamic TEM (DTEM)

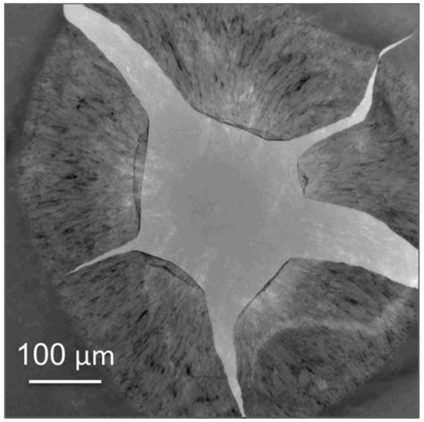




1. UV and IR laser optics system
2. Molybdenum mirror
3. Tantalum cathode
4. C_0 lens
5. Drift section
6. Ultrafast deflector
7. Ion irradiation beamline

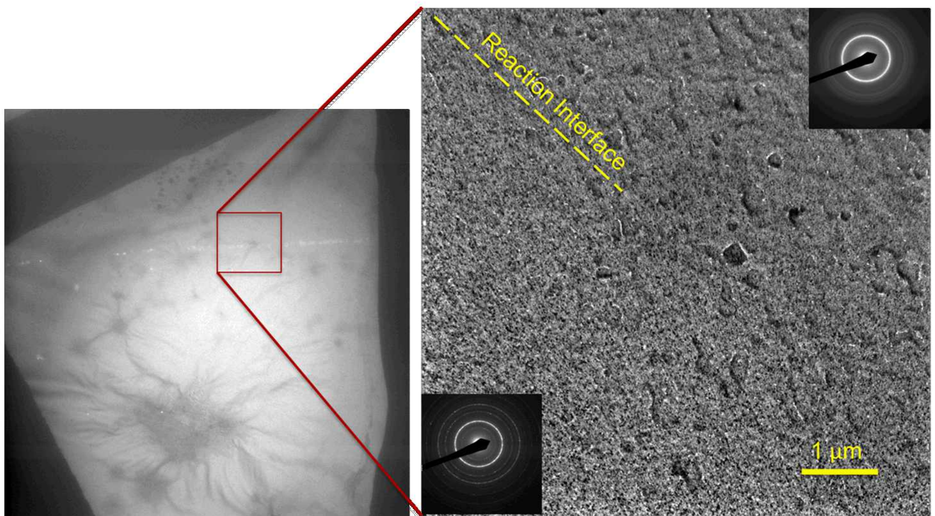
- Further modifications were added to the I^{3}TEM converted to a Dynamic Transmission Electron Microscope (DTEM)
- UV laser is directed at a Ta cathode to photoexcite a nanosecond pulse of electrons
- IR laser is directed to the sample to incite a reaction
- Photoexcited electrons produce an image of the reaction occurring
- Conversion marks the **world's first in-situ ion irradiation dynamic transmission electron microscope (I^{3}DTEM)!**

Alignment and Timing Challenges of UV and IR System

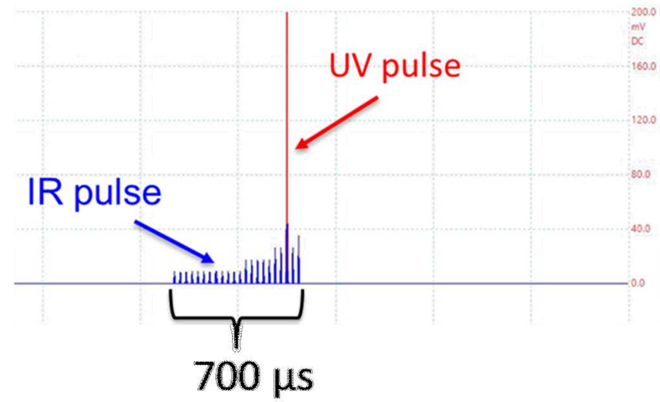


Final alignment of IR laser spot coincident with electron beam on Si_3N_4

- Mirrors in the laser box system are carefully aligned to ensure UV and IR lasers are directed to cathode and sample, respectively.
- UV and IR pulses can be triggered and timed with extreme precision to capture the events at the desired timing.



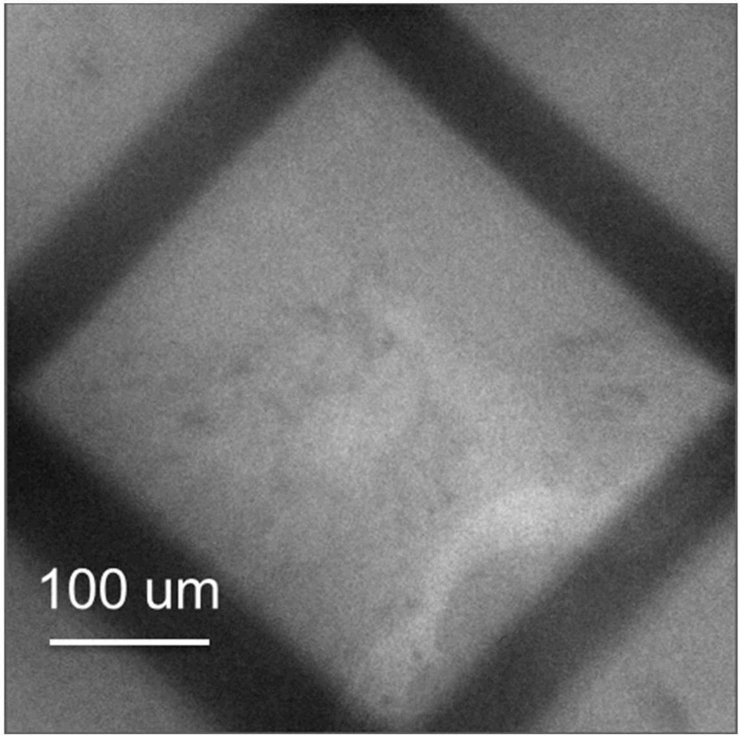
Al/Co multilayer sample **after IR laser shot** and closeup of reaction interface



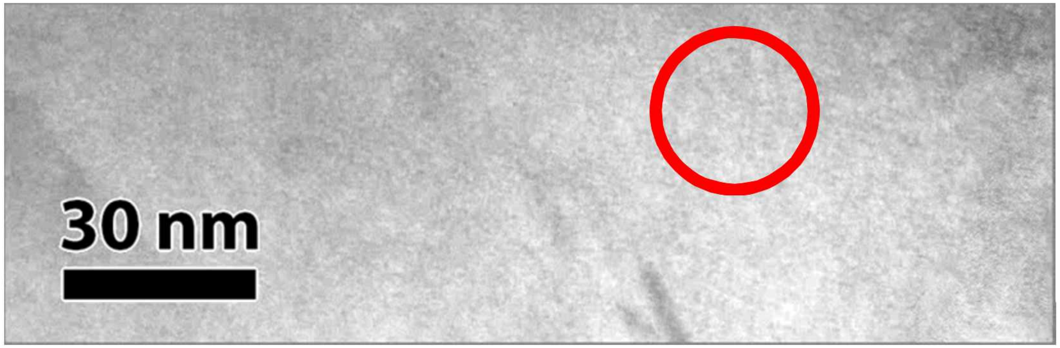
Timing of UV and IR laser pulses when triggered simultaneously

Current State and Challenges for DTEM

- Successful photoemission and images obtained with DTEM
- Timing of IR and UV lasers determined
- UV and IR pulses can be triggered and timed with extreme precision to capture the events at the desired timing.
- Intensity needs to be increased to explore high magnification
- Timing between ion beam and UV laser for capture of single ion cascade needs to be determined

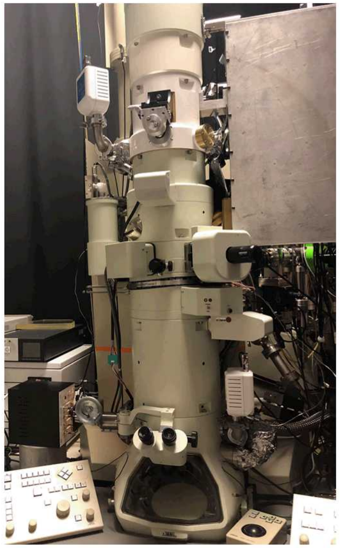


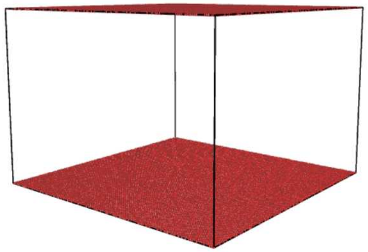
Single shot DTEM of unreacted Al/Co multilayer sample

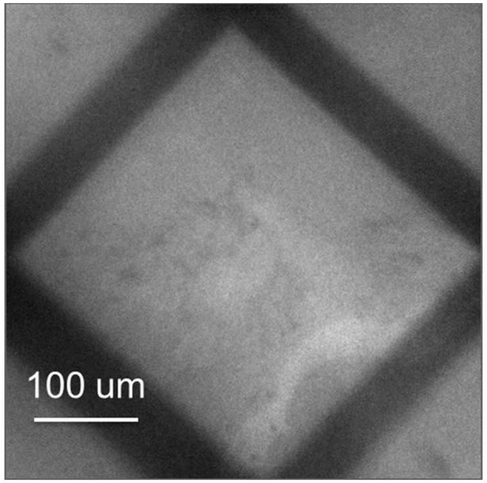


Conclusions

- The **world's first in-situ ion irradiation dynamic transmission electron microscope (I^3DTEM)** has been created at Sandia National Laboratories.
- This microscope allows for the study of processes/reactions occurring on the **nanosecond and nanometer scale**, including the study of single ion strikes.
- DTEM is an essential tool to guide modeling of processes which are limited to this small spatial and temporal scale.
- Future upgrades and improvements:
  - Increase in electron beam intensity for high magnification imaging
  - Synchronizing timing of UV laser with the capture of a single ion strike







QUESTIONS?

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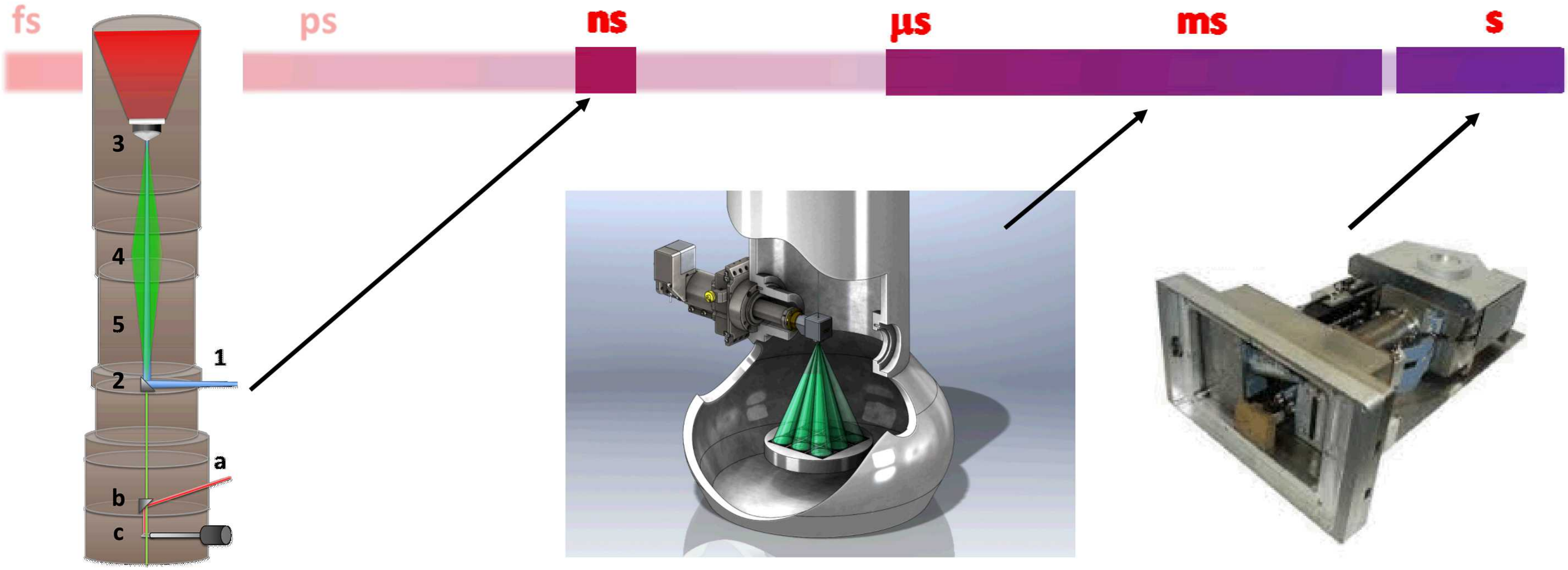
Extra Slides

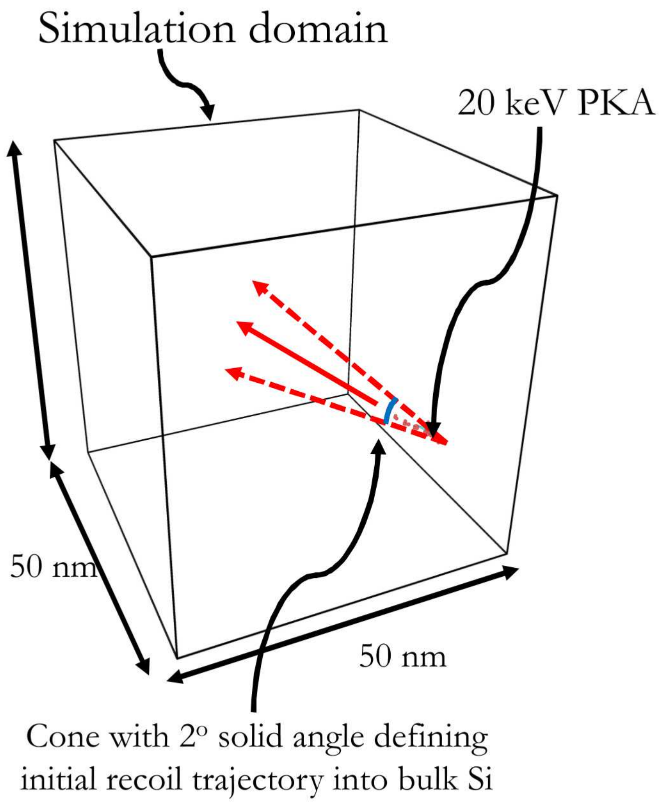
Increasing Temporal Resolution



- **DTEM**
  - Laser induced photoemission of electrons is needed to achieve sufficient current density to produce an image
  - Provides nanosecond imaging of irreversible process
- **Deflector System**
  - Multiple images acquired on single frame
  - Microsecond imaging possible
  - Virtually no missing data (nanosecond gaps)
- Standard 1K TVIPS camera
  - Due to camera read out rate few images can be acquired
  - 10-20fps maximum
  - Missing data during camera readout

CASCADE DAMAGE WITH MOLECULAR DYNAMICS SETUP

- LAMMPS code improved to perform simulation of PKA displacement cascade:
  - Improvement of electronic stopping effects (beyond SRIM)

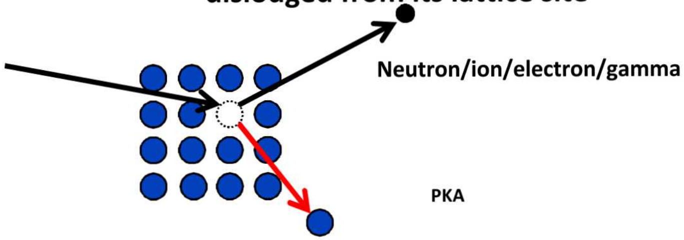


Computational Tools for Studying Radiation Damage

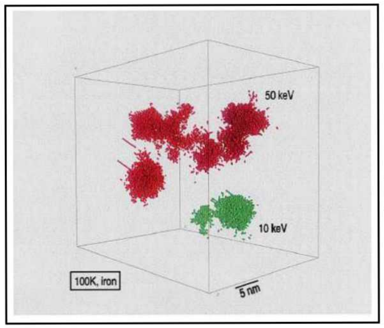
Tool Name	Uses
Density Functional Theory (DFT)	Defect binding and migration energies Developing atomic potentials
Atomistics: Molecular Dynamics (MD) and Molecular Statics (MS)	Cascade damage Defect binding and migration energies Defect interactions with other objects (dislocations, impurities)
Kinetic Monte Carlo methods: Object kinetic Monte Carlo (OKMC) Event kinetic Monte Carlo (EKMC) others	Cascade annealing Defect interactions with objects (dislocations, impurities) Sink strengths Defect accumulation Annealing
Rate theory methods: Mean field rate theory (MFRT) Cluster dynamics (CD)	Defect accumulation Annealing Large doses and times
Phase field models	Large-scale defect accumulation Bubble growth Interfacial behaviors
Discrete dislocation dynamics (DDD)	Defect-dislocation interactions Hardening

How does Radiation Damage Occur?

Damage is caused when a primary knock-on atom (PKA) is dislodged from its lattice site

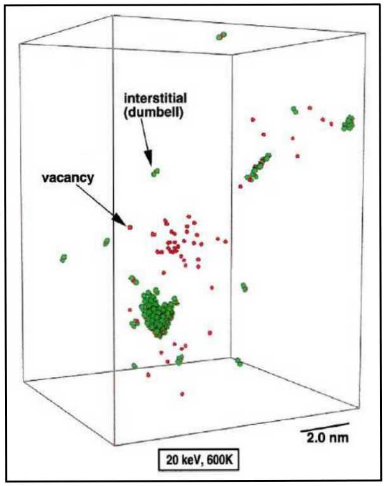


Displacement damage is in the form of cascades or Frenkel pairs (single vacancy + single interstitial)



Stoller and Calder (2000)

30 ps



Source	Damage Type	Dose Rate (DPA/s)
Electron (1-3 MeV)	Frenkel Pair	10^{-9} - 10^{-3}
Neutron (1-14 MeV)	Cascade	10^{-7} - 10^{-6} (fast fission, fusion) 10^{-12} - 10^{-11} (thermal fission)
Light ion (~ 10 keV)	Frenkel Pair	10^{-4} - 10^{-2}
Heavy ion (keV – MeV)	Cascade	10^{-4} - 10^{-2}
Gamma (>2 MeV)	Frenkel Pair	1-5 x neutron DPA rate (in HFIR)