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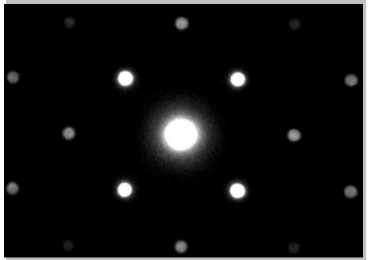

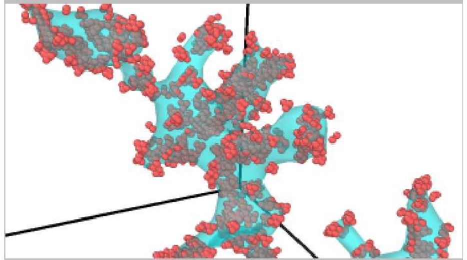

In-Situ Characterization of Single Ion Strikes in Single Crystal Silicon

Anthony M. Monterrosa, James Stewart, Patrick Price, Remi Dingreville, Khalid Hattar

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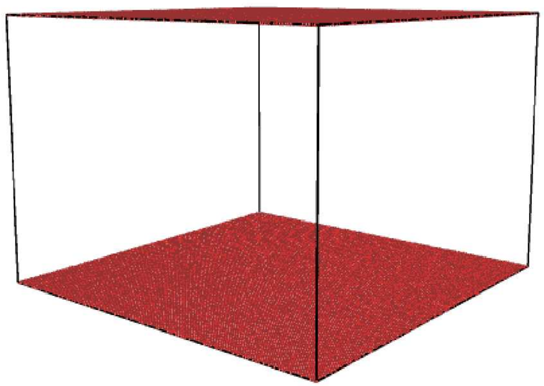
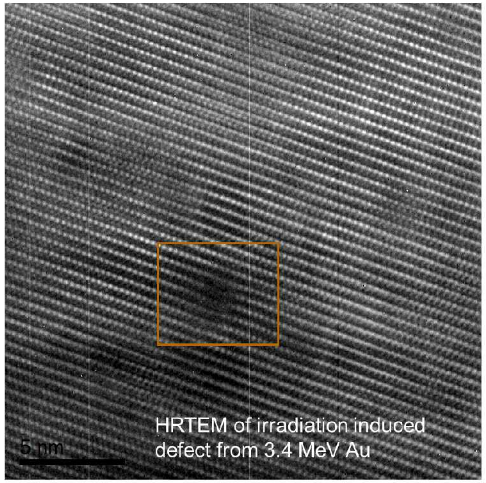
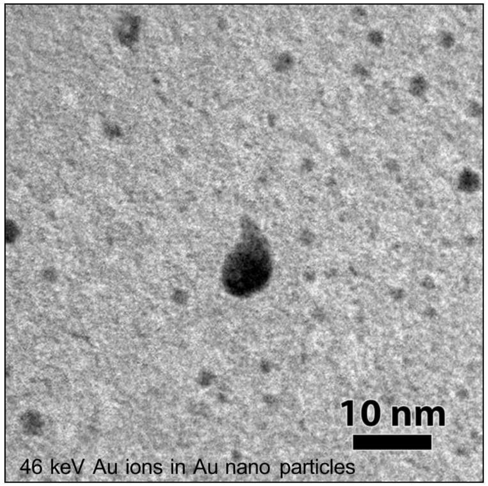
Temporal Gap Between Modeling and Experiments







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

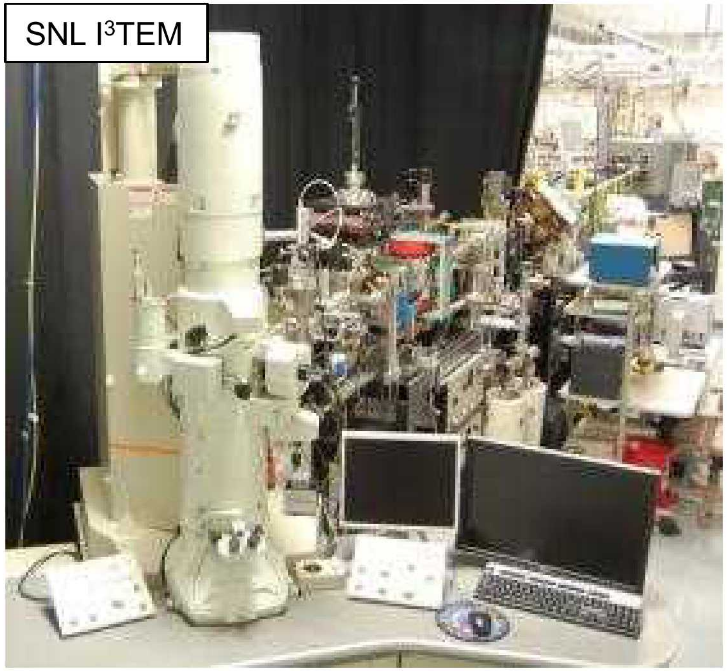
Computational models can be scaled up but is expensive and requires multidimensional physics.

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

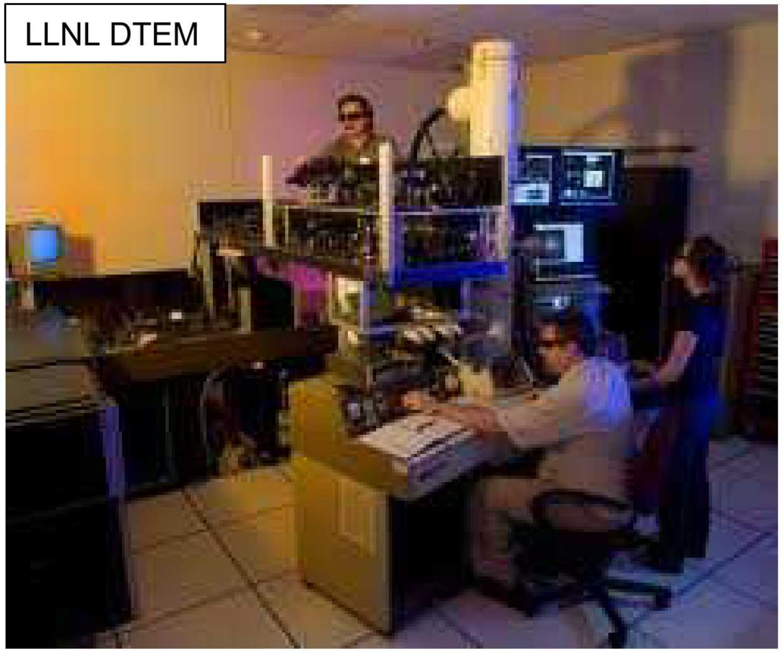
Objective

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

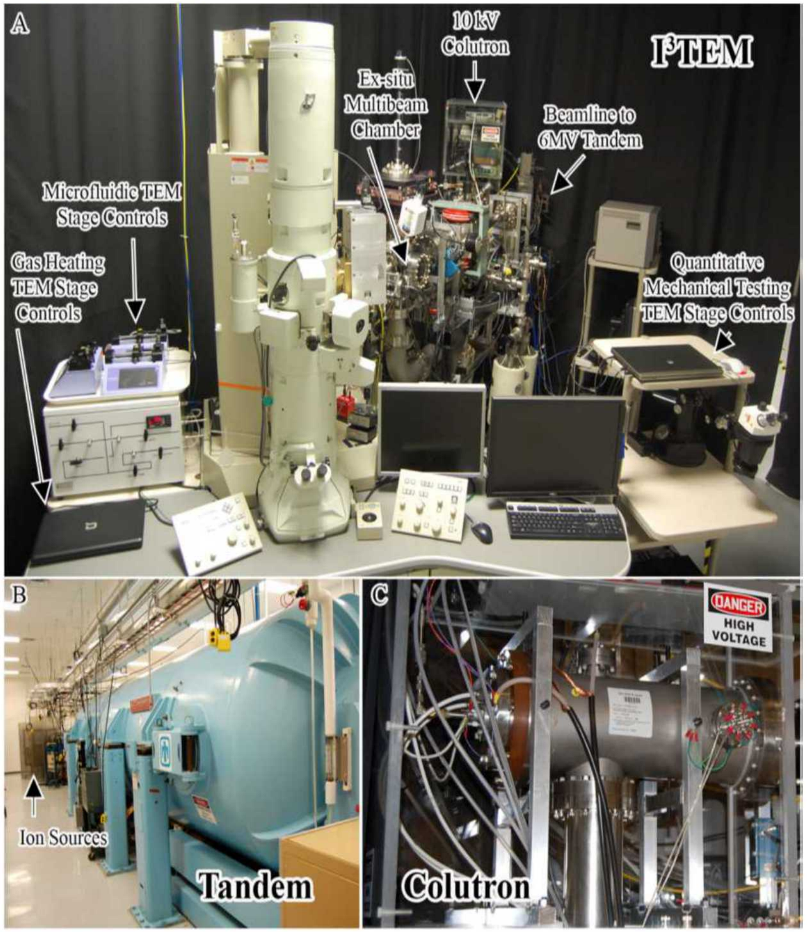
SNL I^3TEM



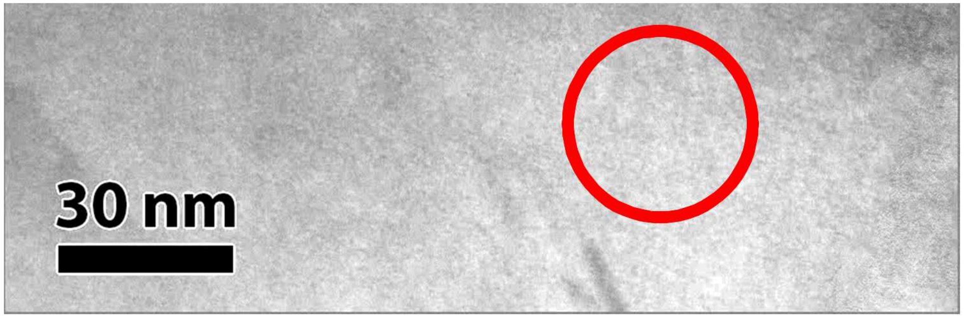
LLNL DTEM



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).



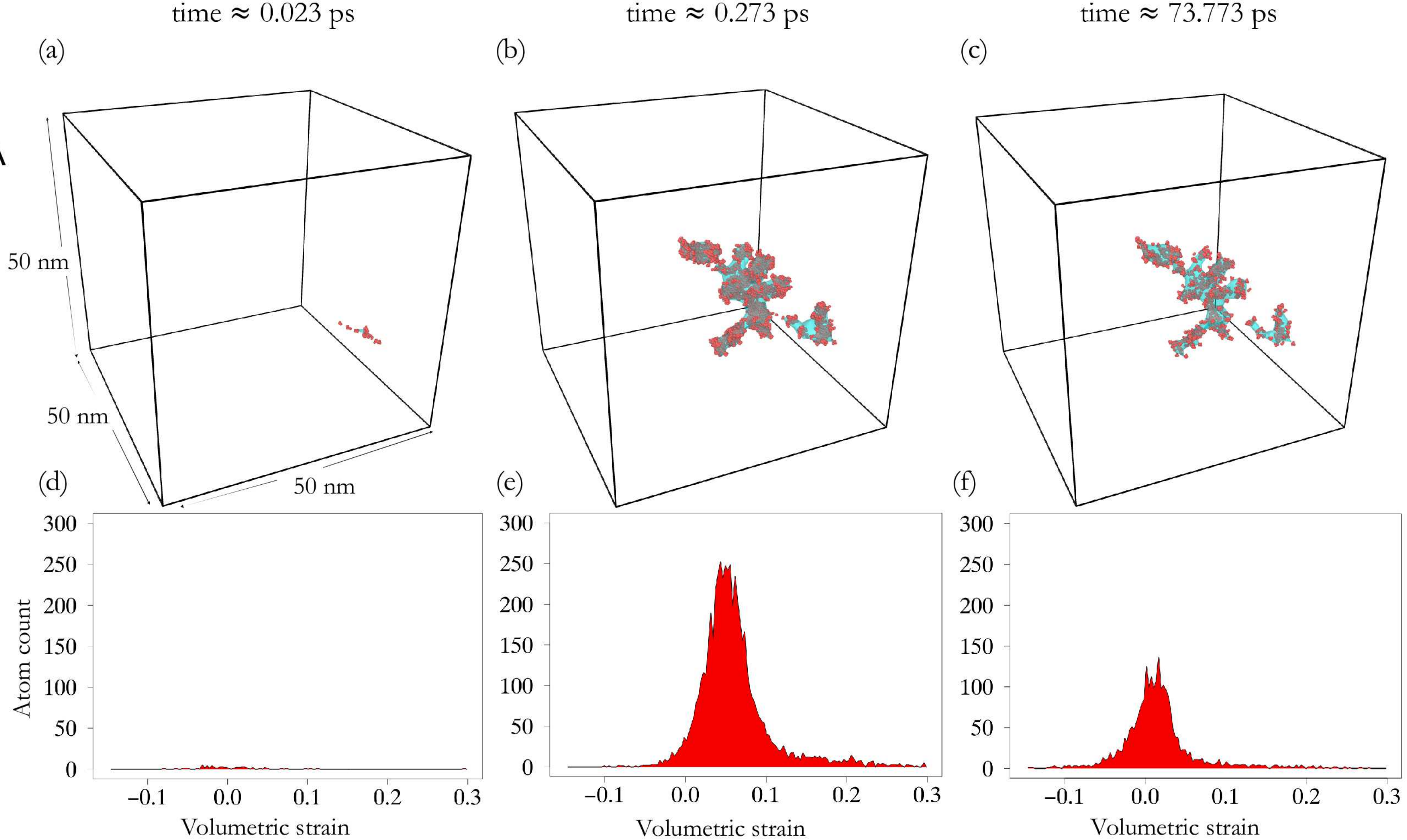
- 1.7 MeV Au into single crystal Si
- Single ion strikes can be observed in semiconductors
- Non-symmetric structure in contrast to the spherical approximation

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

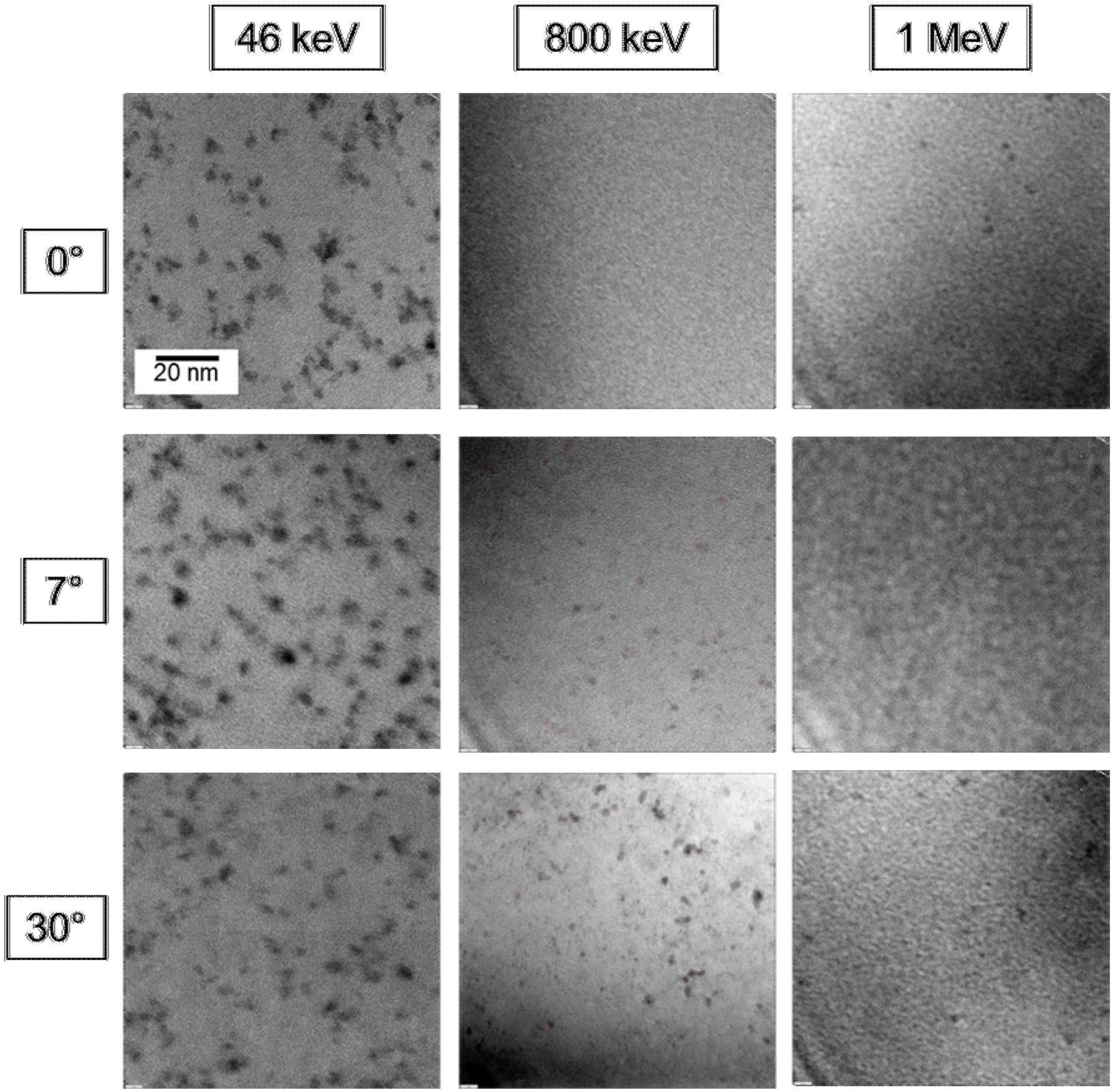
INITIATION PEAK DAMAGE DEFECT RECOMBINATION

20 keV PKA in bulk Si



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

Black Dot Damage Distributions



Single crystal <001> Si irradiated ex-situ with Au ions at a variety of energies and angles of incidence.

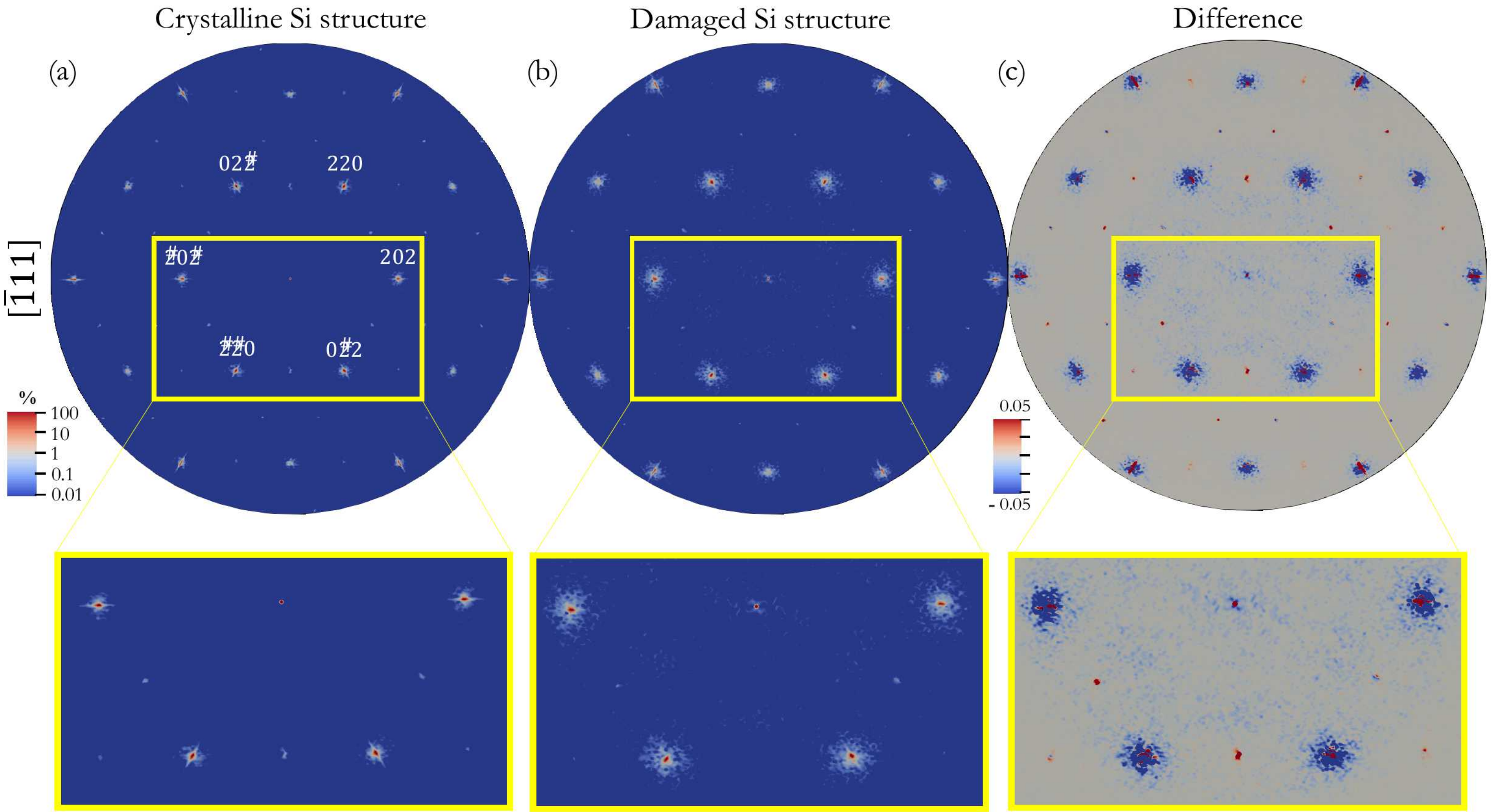
Probing the damage cascade experimentally is limited to contrast from black-dot damage.

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.

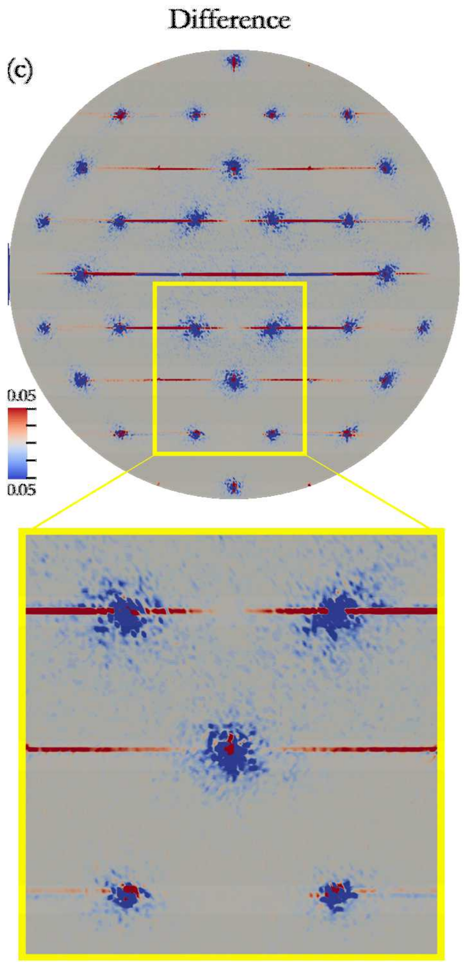


Calculated SAED patterns for [111] Si before and after a single ion cascade event

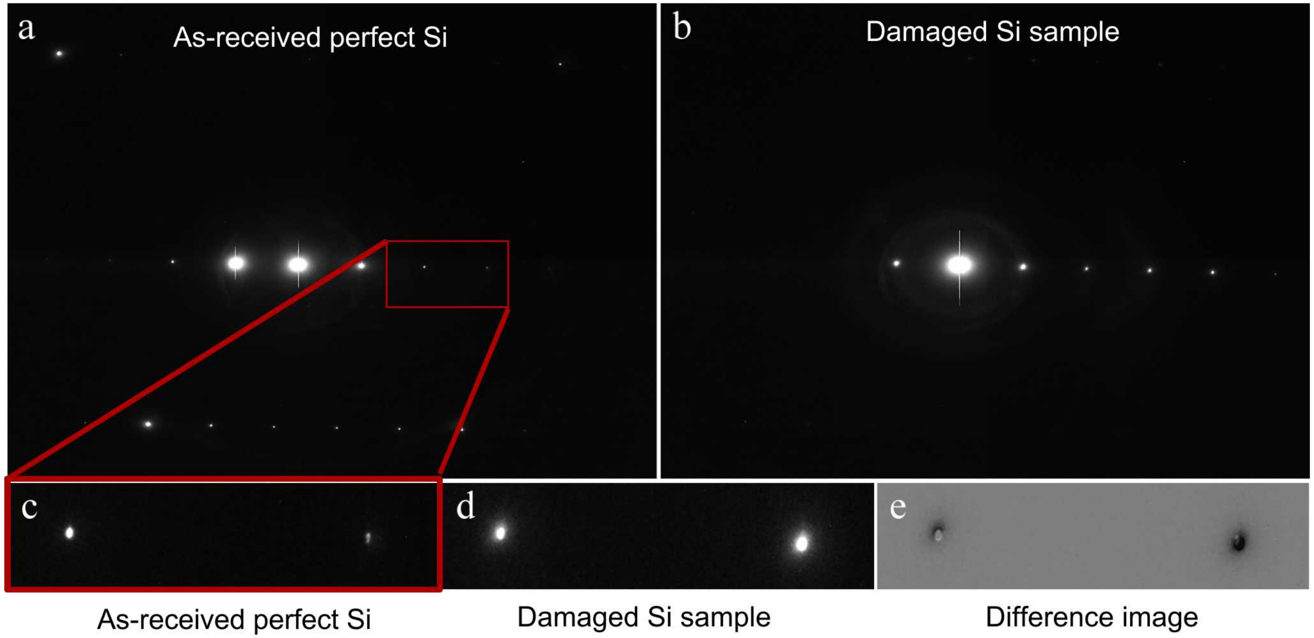
[Stewart et al. (2018) J. Appl. Phys.]

Using Diffraction to Compare Modeling and Experiments

Simulated Image



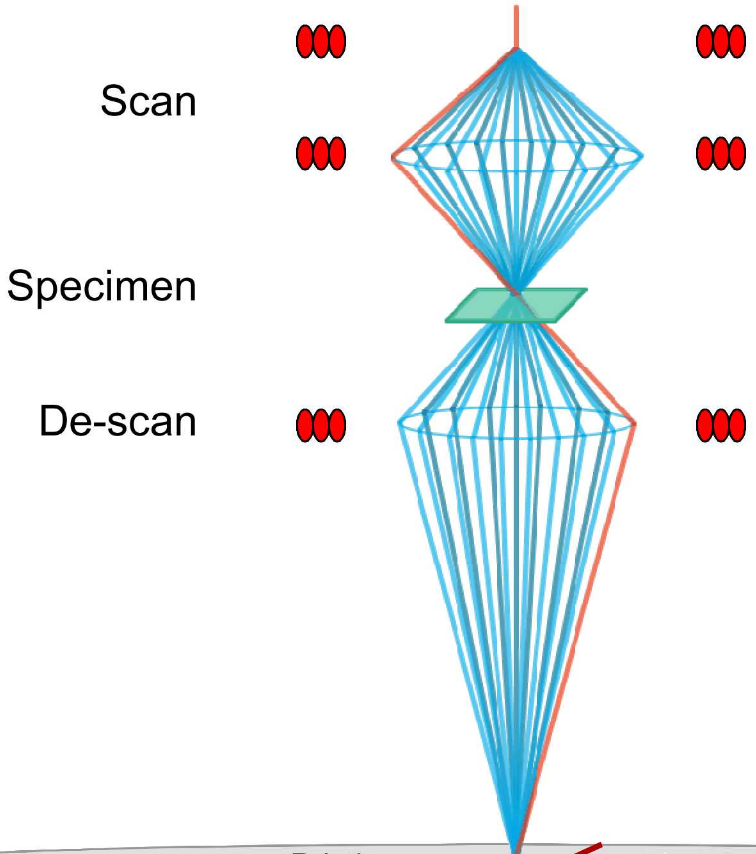
Before and After In-Situ Irradiation

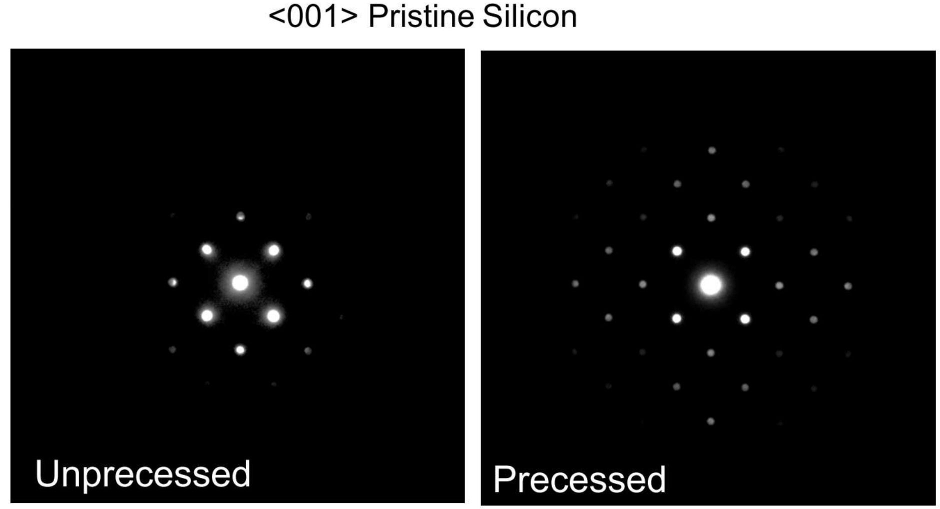


SAED may suggest a similar trend. However, many factors can influence diffraction spot size. **Precession electron diffraction (PED), may provide a more consistent pattern.**

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

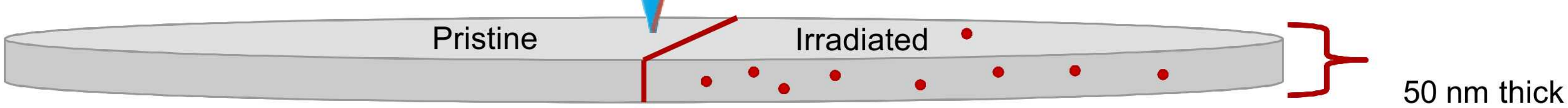
Precession Electron Diffraction (PED) Microscopy





Advantages for interpretation of radiation damage

- ~ 10 nm spatial resolution
- Near kinematical electron diffraction, allowing for interpretation of intensities
- Symmetry ambiguities are resolved

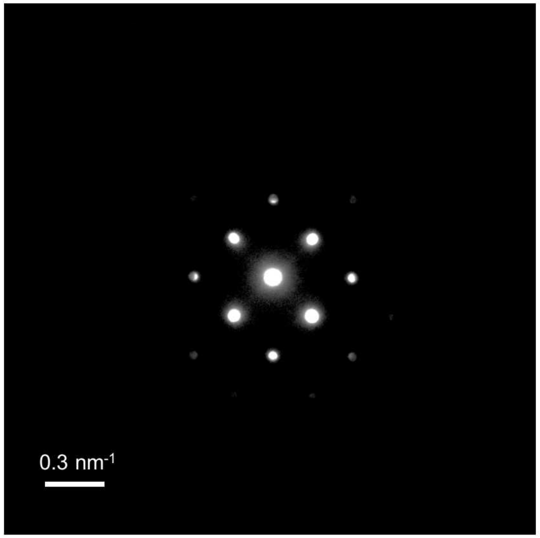


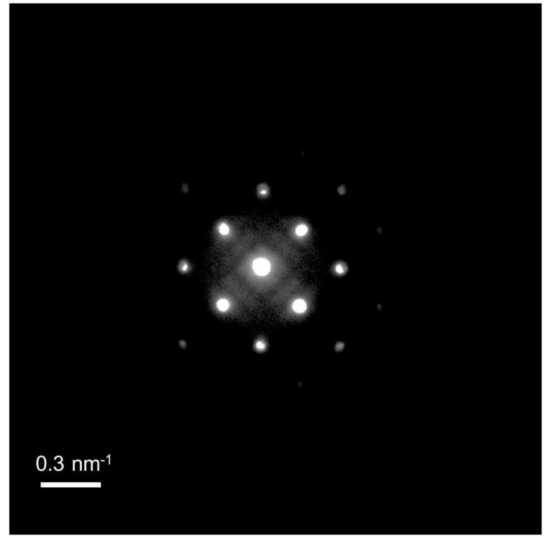
50 nm thick <001> Si irradiated with 46 keV Au^+ to a fluence ~10^{13} ions/cm^2. Samples were half irradiated so that each sample had a pristine and an irradiated region. The same exact imaging, precession, and camera conditions were used to capture patterns from each area.

Precession Electron Diffraction of Irradiated Silicon

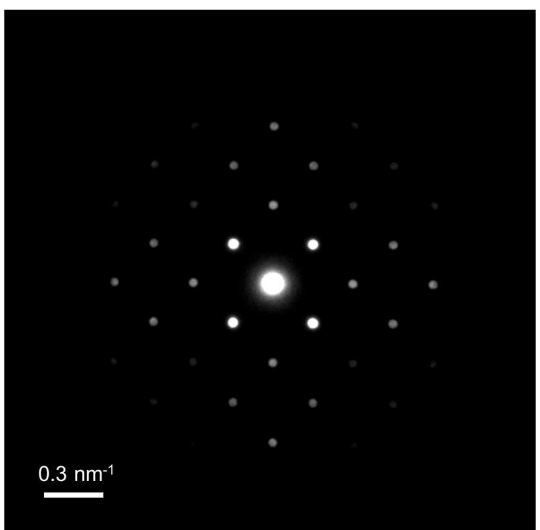
Pristine | Irradiated

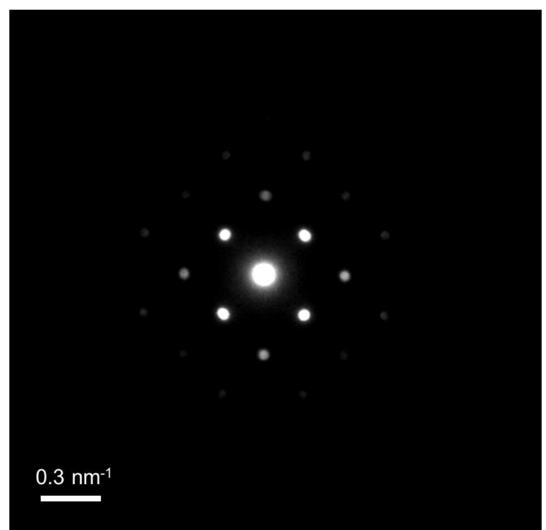
0 ° Precession Angle





1.2 ° Precession Angle





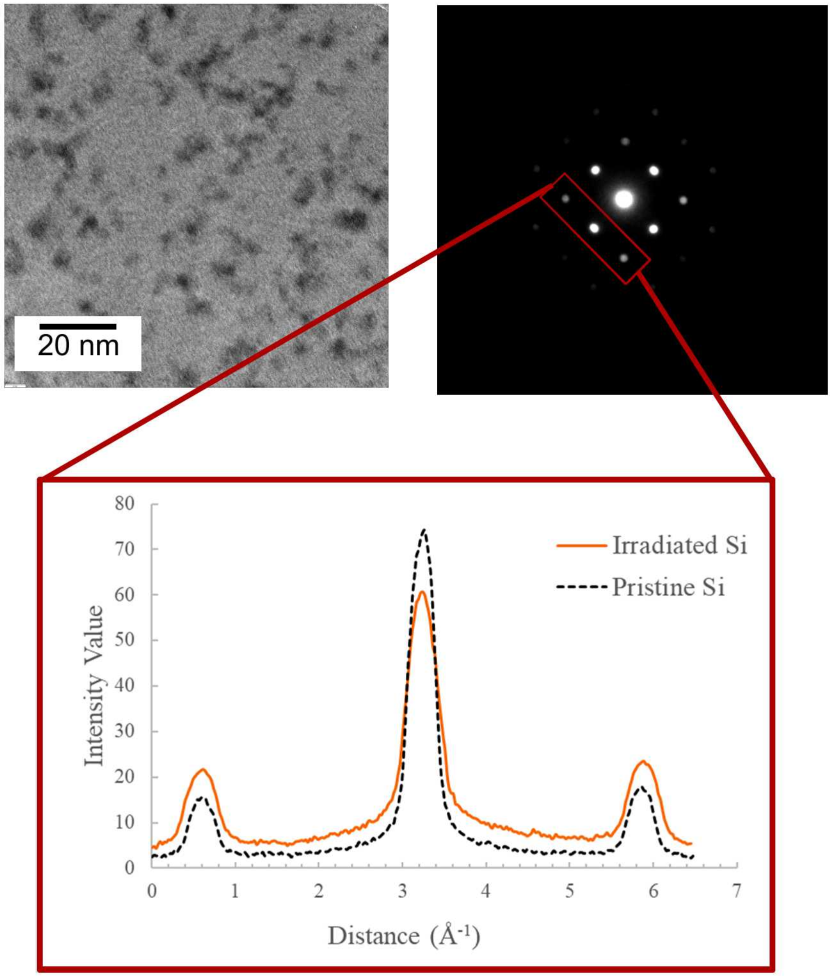
<001> Single Crystal Silicon irradiated to 10^{13} ions/cm^2 with 46 keV Au^+.

Precession patterns taken on pristine and irradiated sections of samples.

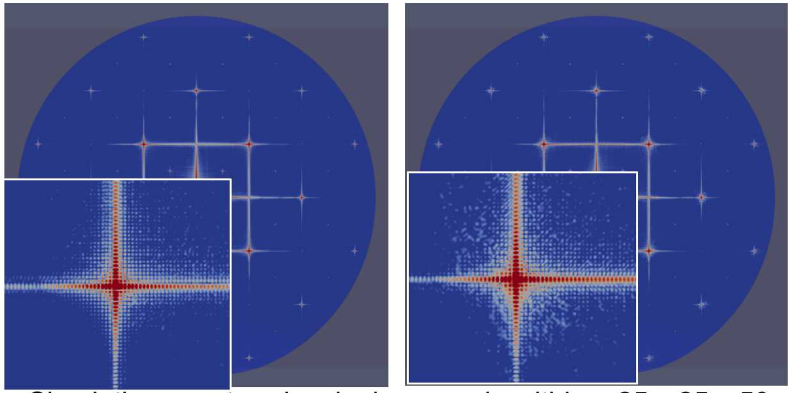
Precession spot size ~10 nm captured 2-3 black dot damage sites.

PED Comparison with Simulation

Experimental bright field and PED pattern of Irradiated Si



Simulated <001> Pristine Si   Simulated <001> Irradiated Si

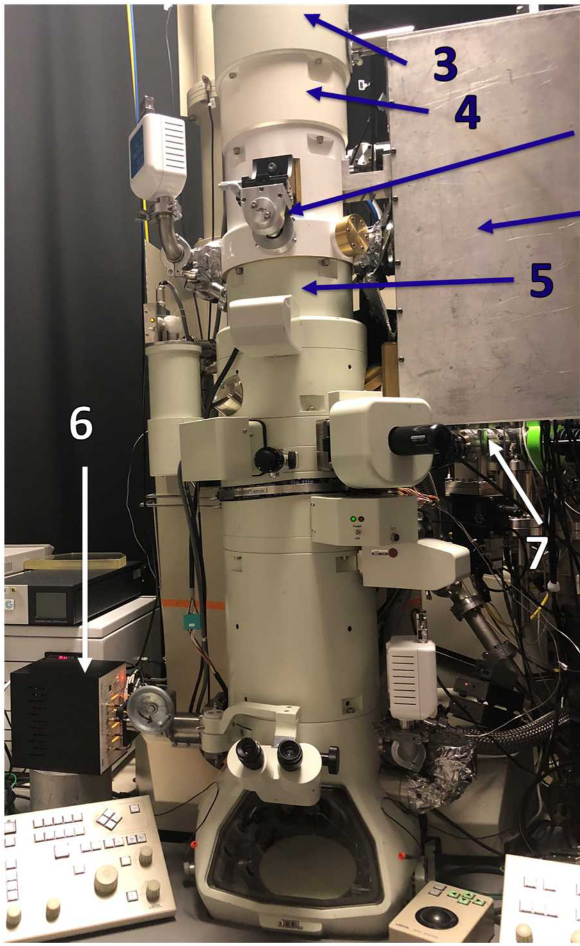


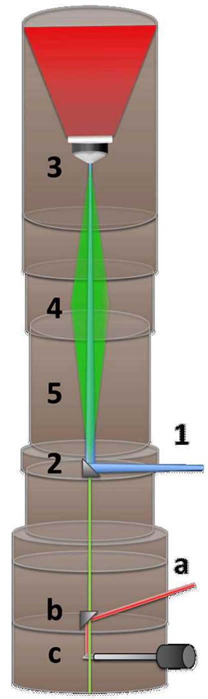
Simulations captured a single cascade within a 25 x 25 x 50 nm volume. Simulations were **not** precessed.

Radiation damage resulted in **broader and more diffuse diffraction spots** in both the experimental and simulated PED patterns.

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

Closing the Temporal Gap with Dynamic TEM (DTEM)

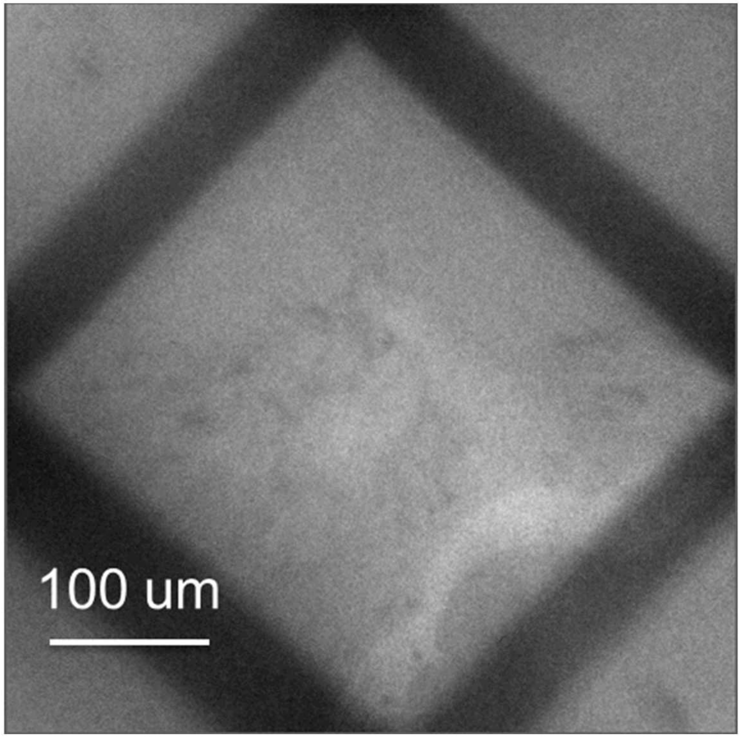




1. UV and IR laser optics system
2. Molybdenum mirror
3. Tantalum cathode
4. C_0 lens
5. Drift section
6. Ultrafast deflector
7. Ion irradiation beamline

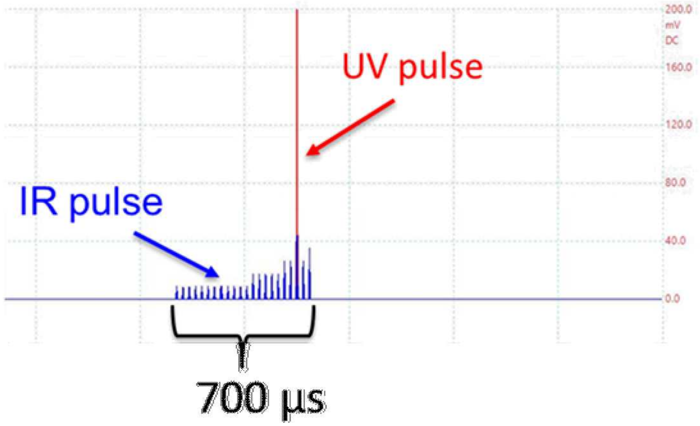
- Further modifications were added to the I^{3}TEM converted to a Dynamic Transmission Electron Microscope (DTEM)
- UV laser is directed at a Ta cathode to photoexcite a nanosecond pulse of electrons
- IR laser is directed to the sample to incite a reaction
- Photoexcited electrons produce an image of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

Current State and Challenges for DTEM



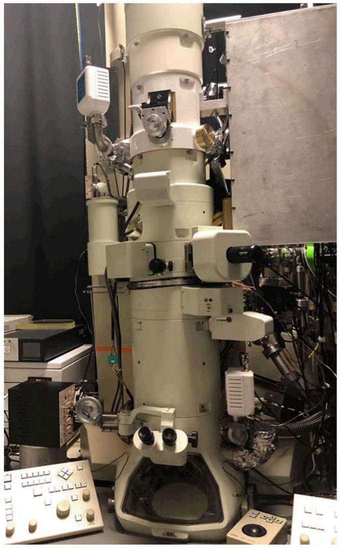
Single Shot DTEM image of unreacted Al/Co multilayer sample

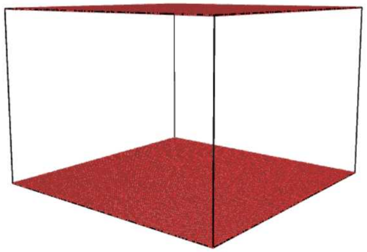
- Successful photoemission and images obtained with DTEM
- Timing of IR and UV lasers determined
- UV and IR pulses can be triggered and timed with extreme precision to capture the events at the desired timing.
- Intensity needs to be increased to explore high magnification
- Timing between ion beam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

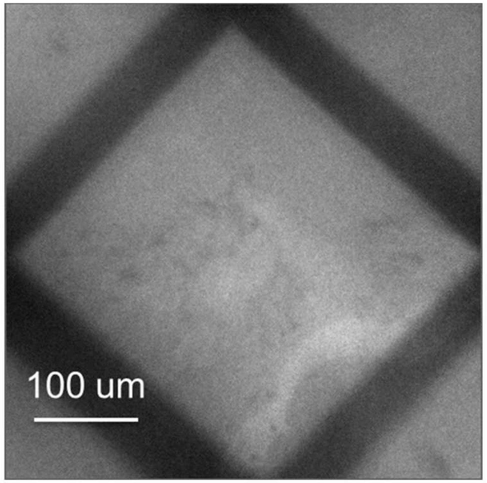


Conclusions

- 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.
- Radiation damage produces diffuse diffraction spots which can be probed by PED
- Future work and improvements:
  - Utilize higher resolution PED (using STEM)
  - Couple experimental PED with simulated PED
  - Increase in electron beam intensity for high magnification imaging
  - Synchronizing timing of UV laser with the capture of a single ion strike



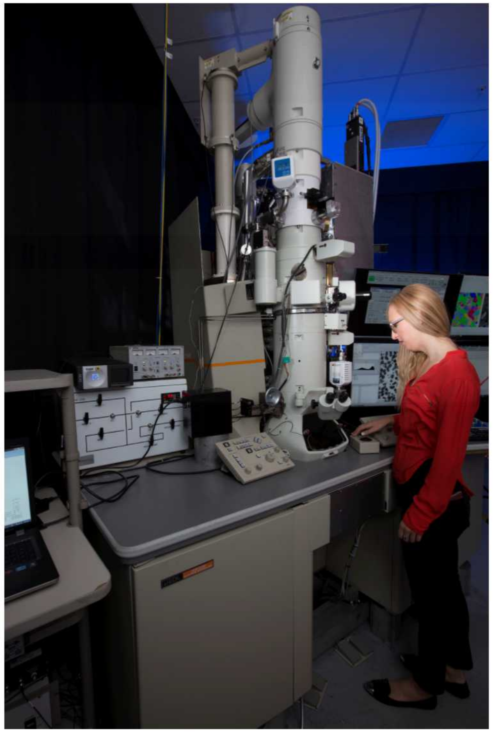




QUESTIONS?

Anthony Monterrosa: amonter@sandia.gov

Khalid Hattar: khattar@sandia.gov



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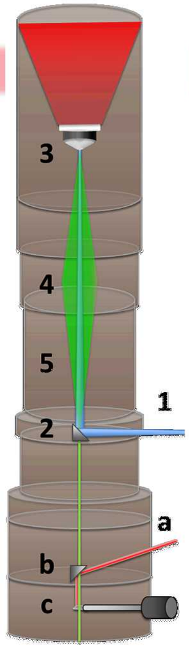
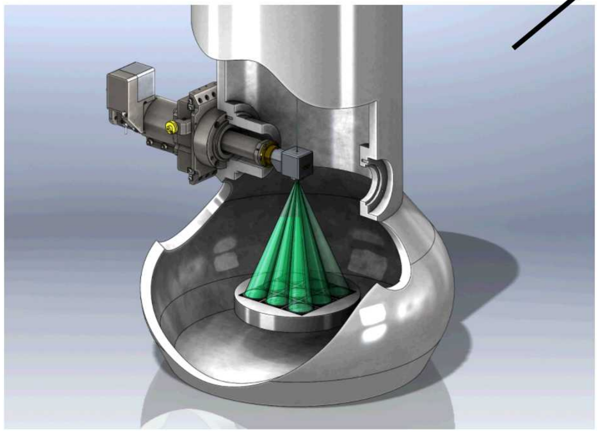
Extra Slides

Increasing Temporal Resolution

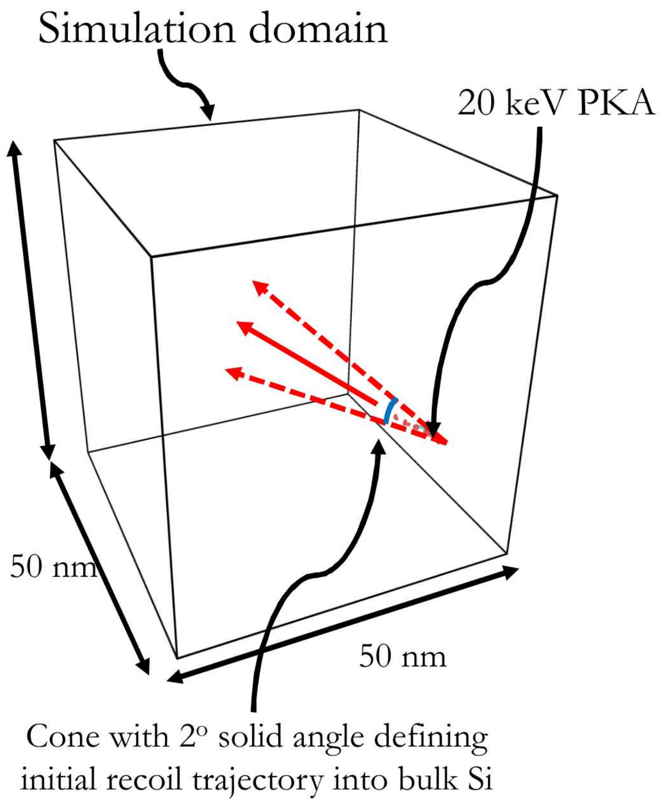




- **DTEM**
  - Laser induced photoemission of electrons is needed to achieve sufficient current density to produce an image
  - Provides nanosecond imaging of irreversible process
- **Deflector System**
  - Multiple images acquired on single frame
  - Microsecond imaging possible
  - Virtually no missing 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)

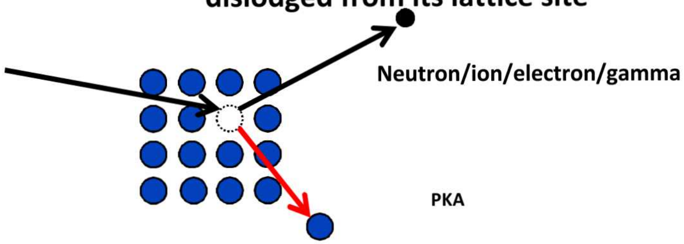


Computational Tools for Studying Radiation Damage

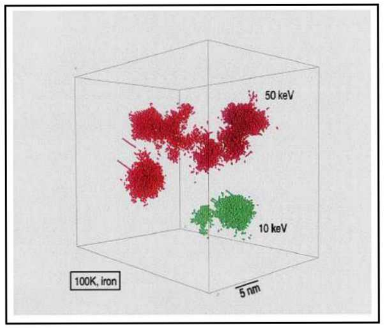
Tool Name	Uses
Density Functional Theory (DFT)	Defect binding and migration energies Developing atomic potentials
Atomistics: Molecular Dynamics (MD) and Molecular Statics (MS)	Cascade damage Defect binding and migration energies Defect interactions with other objects (dislocations, impurities)
Kinetic 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?

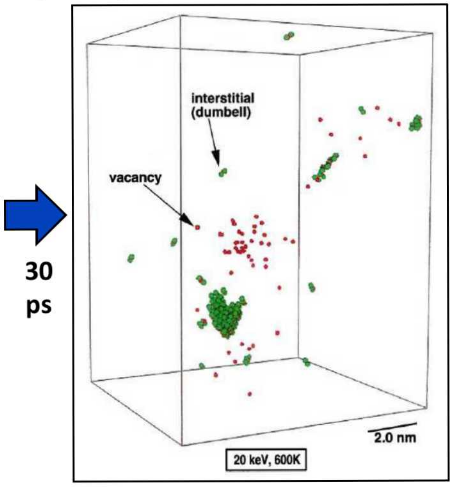
Damage is caused when a primary knock-on atom (PKA) is dislodged from its lattice site



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



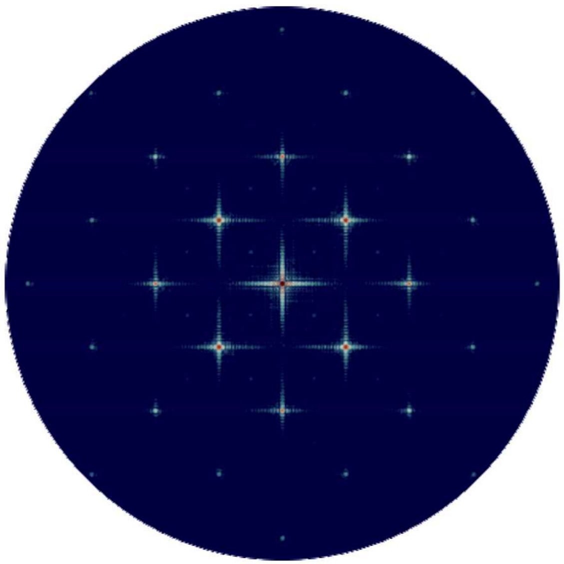
Stoller and Calder (2000)

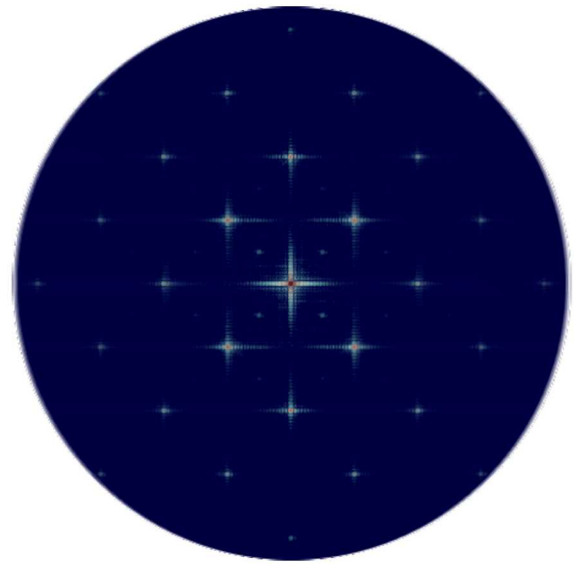


Source	Damage Type	Dose Rate (DPA/s)
Electron (1-3 MeV)	Frenkel Pair	10^{-9} - 10^{-3}
Neutron (1-14 MeV)	Cascade	10^{-7} - 10^{-6} (fast fission, fusion) 10^{-12} - 10^{-11} (thermal fission)
Light ion (~ 10 keV)	Frenkel Pair	10^{-4} - 10^{-2}
Heavy ion (keV – MeV)	Cascade	10^{-4} - 10^{-2}
Gamma (>2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

Preliminary Precession

[001]





Average of:
[001]
$[\overline{0.04}\ 0\ 1]$
$[\overline{0.02}\ 0\ 1]$
$[0.02\ 0\ 1]$
$[0.04\ 0\ 1]$