The response of solids to irradiation by massive particles

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Support:
- Center for Materials at Irradiation and Mechanical Extremes (CMIME), a DOE EFRC
- DOE-NE, NEET Reactor Materials program
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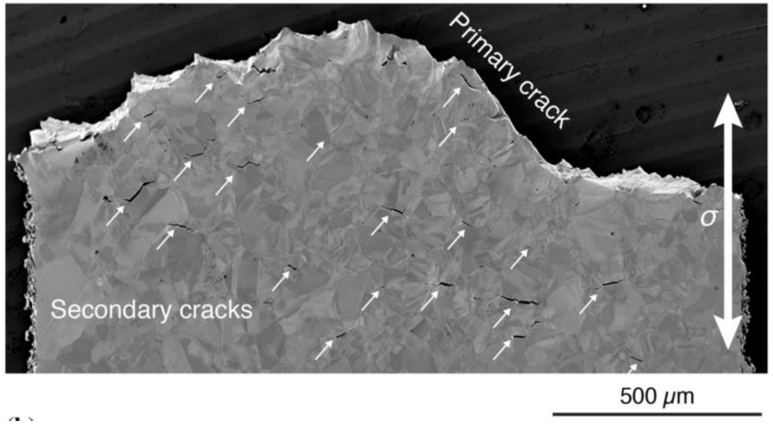
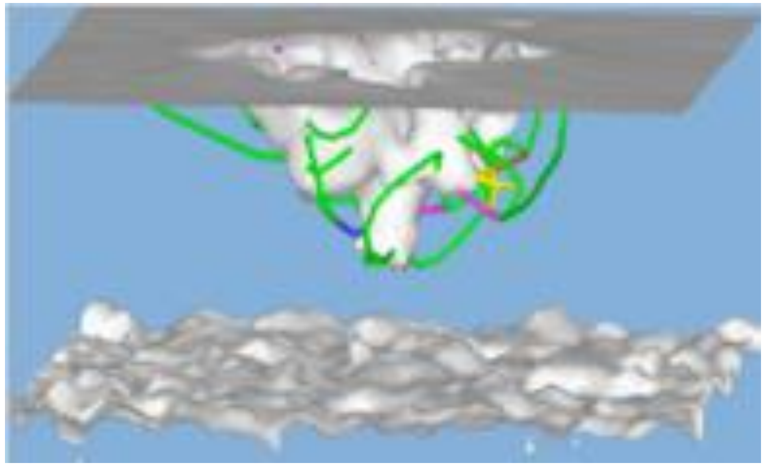
Acknowledgements:
R. E. Baumer, L. Zhang, S. A. Skirlo

Metallic materials in extremes

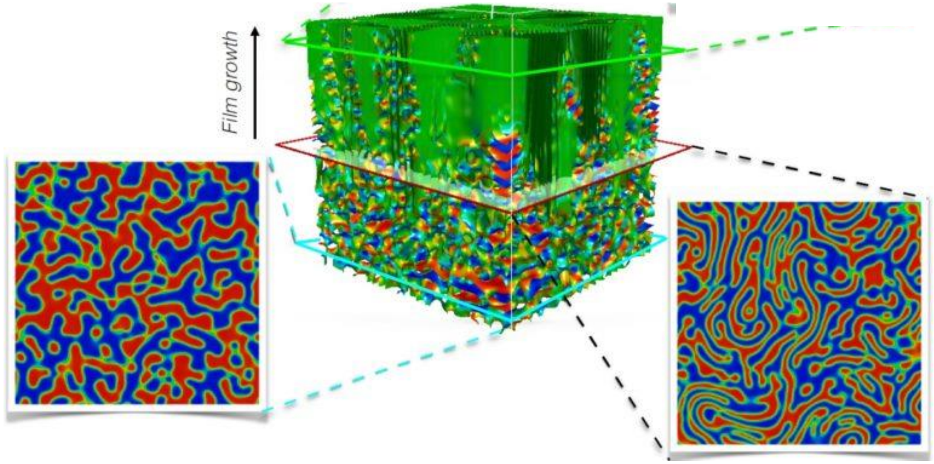
Hydrogen embrittlement, corrosion



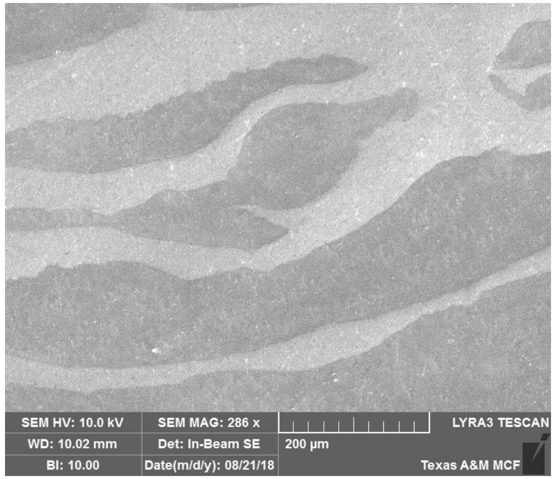
High strain rate deformation



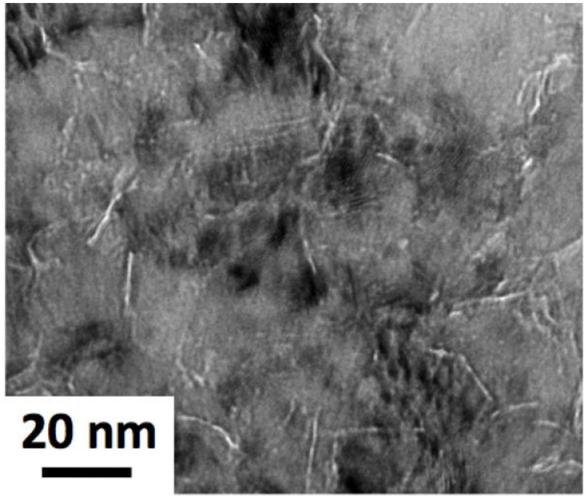
High temperature microstructure evolution



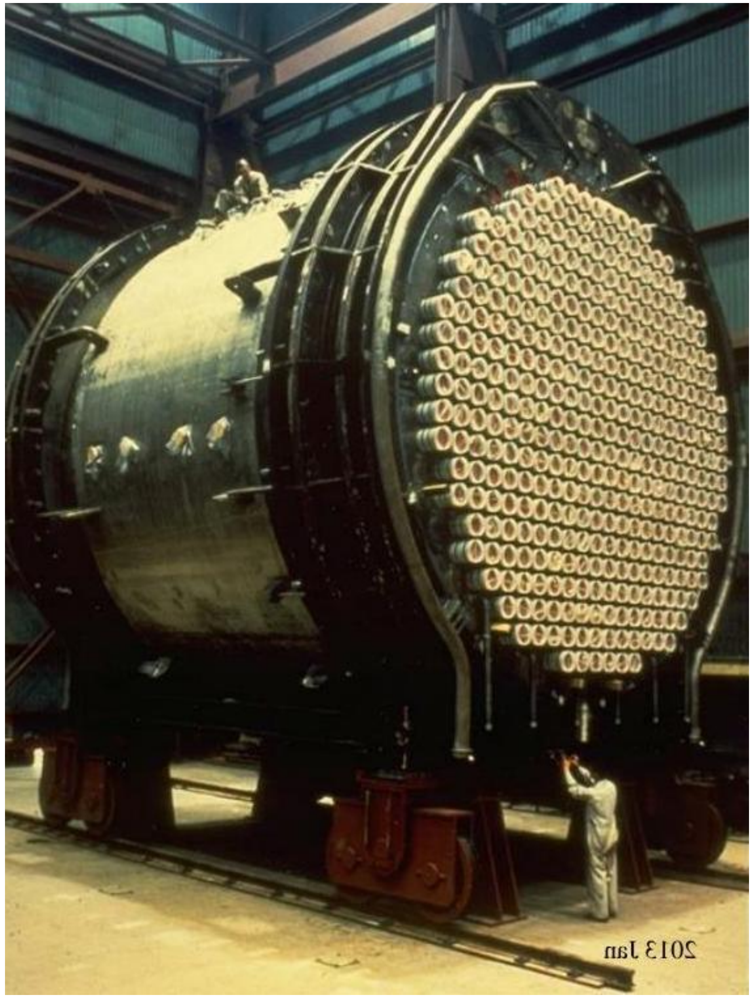
Severe plastic deformation



Radiation response



Nuclear fission: energetic particles generated inside the solid



CANDU reactor fuel assembly

A single fuel bundle

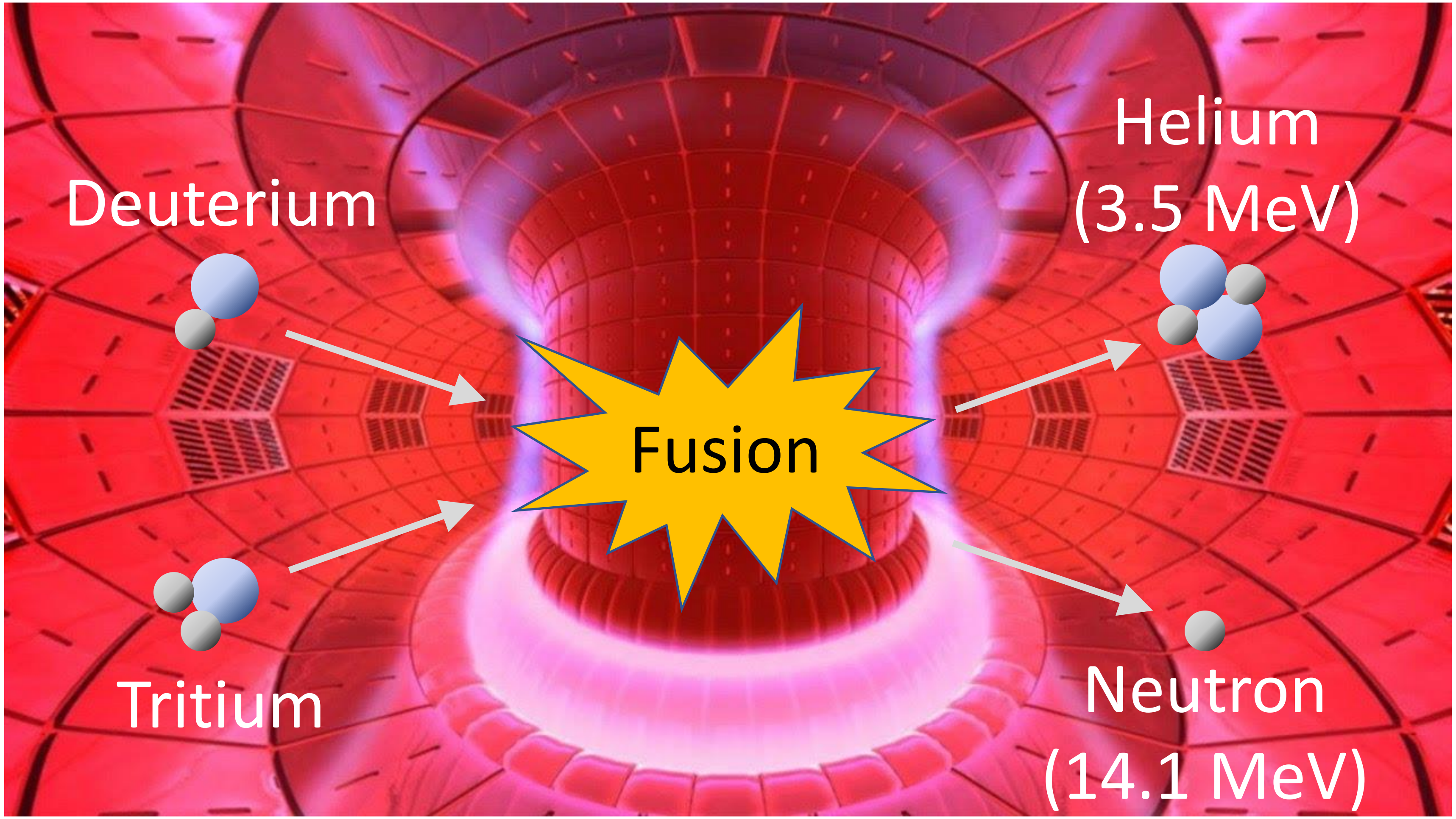
Fuel (UO_2):
^{235}U → neutrons, fission products with ~200MeV of kinetic energy

Garter spring (Ni-base alloys X-750):

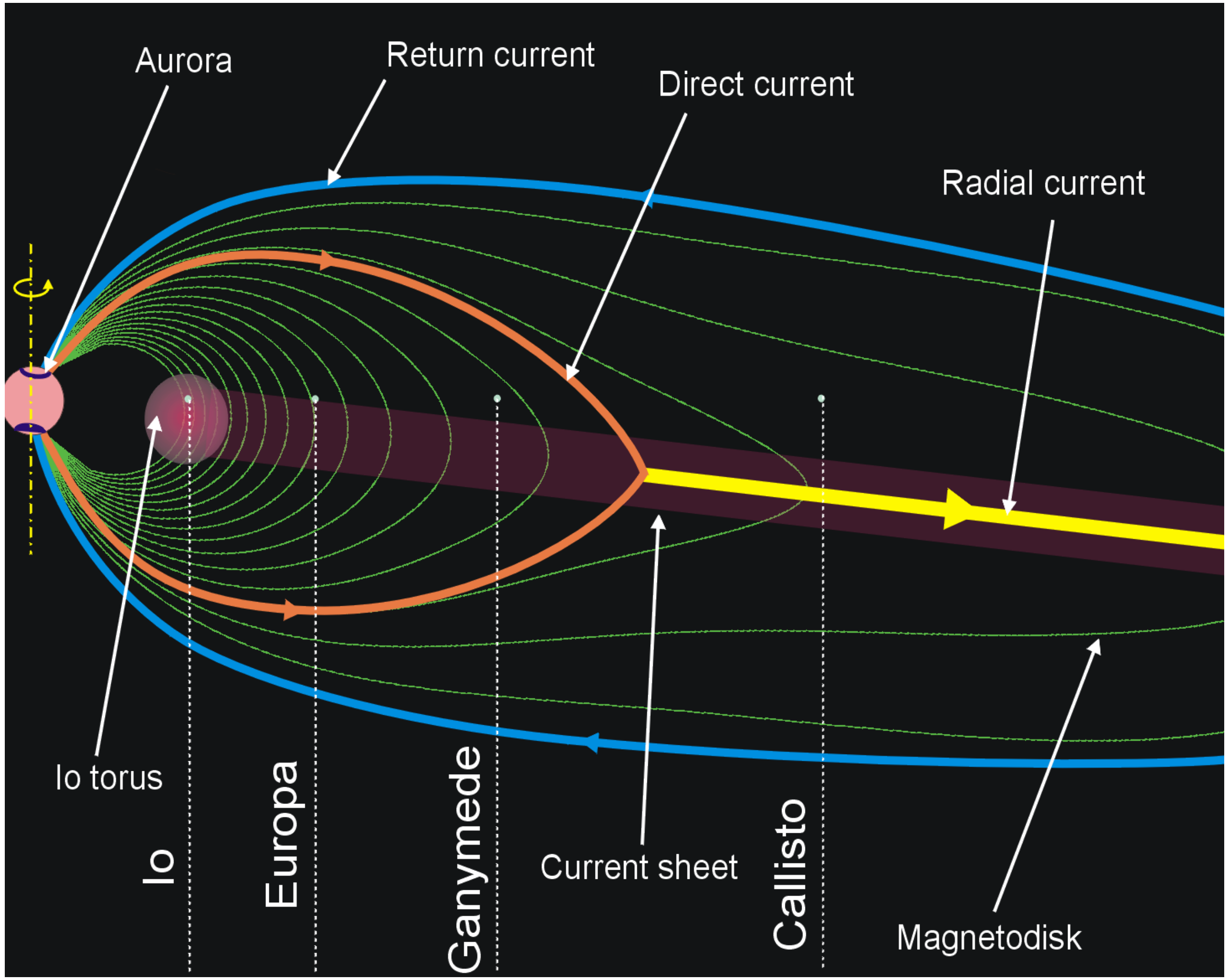
$$^{58}Ni + n \rightarrow {}^{59}Ni + \gamma$$

$$^{59}Ni + n \rightarrow \underbrace{{}^{56}Fe}_{0.3MeV} + \underbrace{\alpha}_{4.8MeV}$$

Magnetic confinement fusion: particles from the plasma

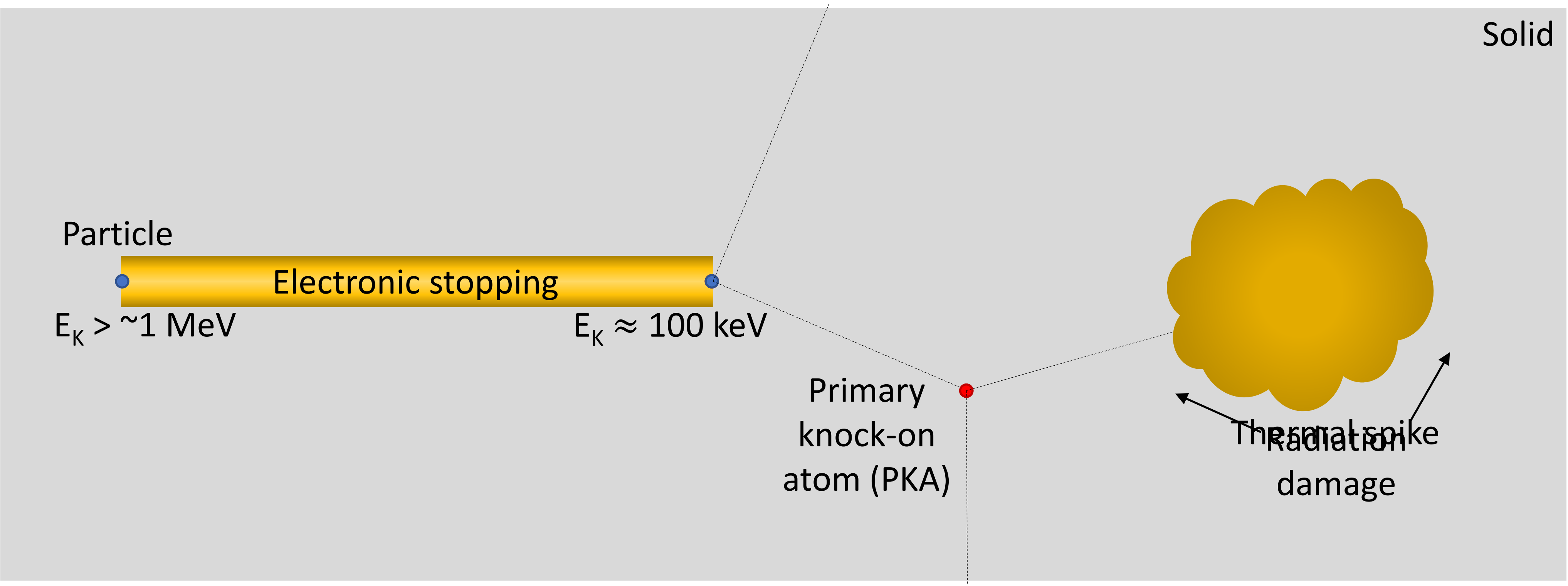


Space: particles accelerated by massive bodies, powerful fields



Jovian magnetosphere: electrons, protons, and ions at 10s of keV

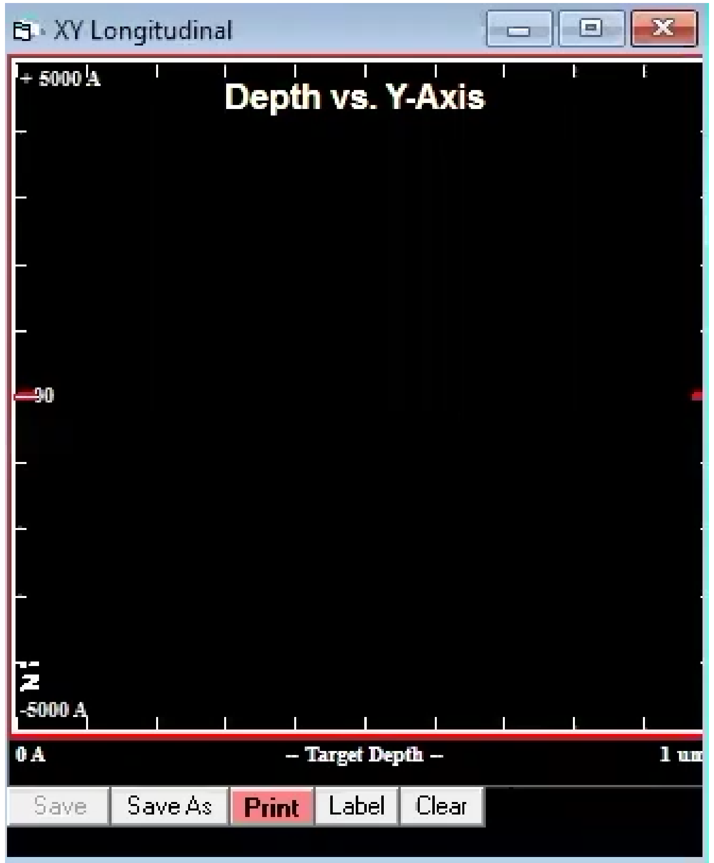
Interaction of massive particles with solids



My talk will focus on the thermal spike phase, where point defects are created

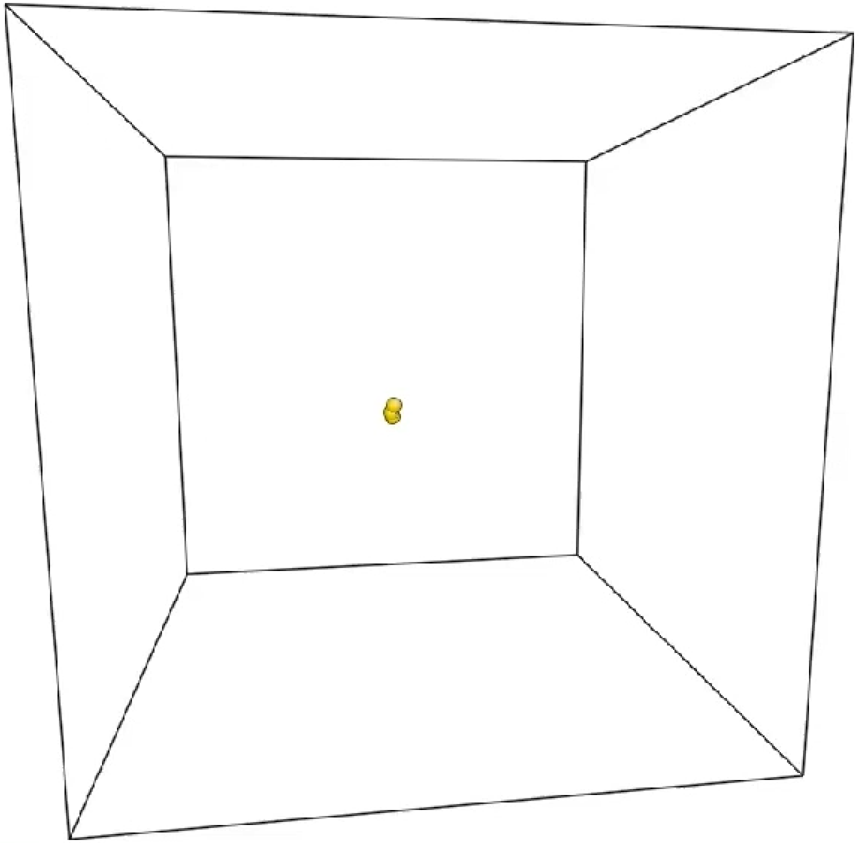
Modeling radiation effects in solids

Collision cascades: SRIM



5MeV Ni self-ion irradiation

Thermal spikes: molecular dynamics



1.5keV self-ion in Cu

My talk will focus on this

Defect diffusion:

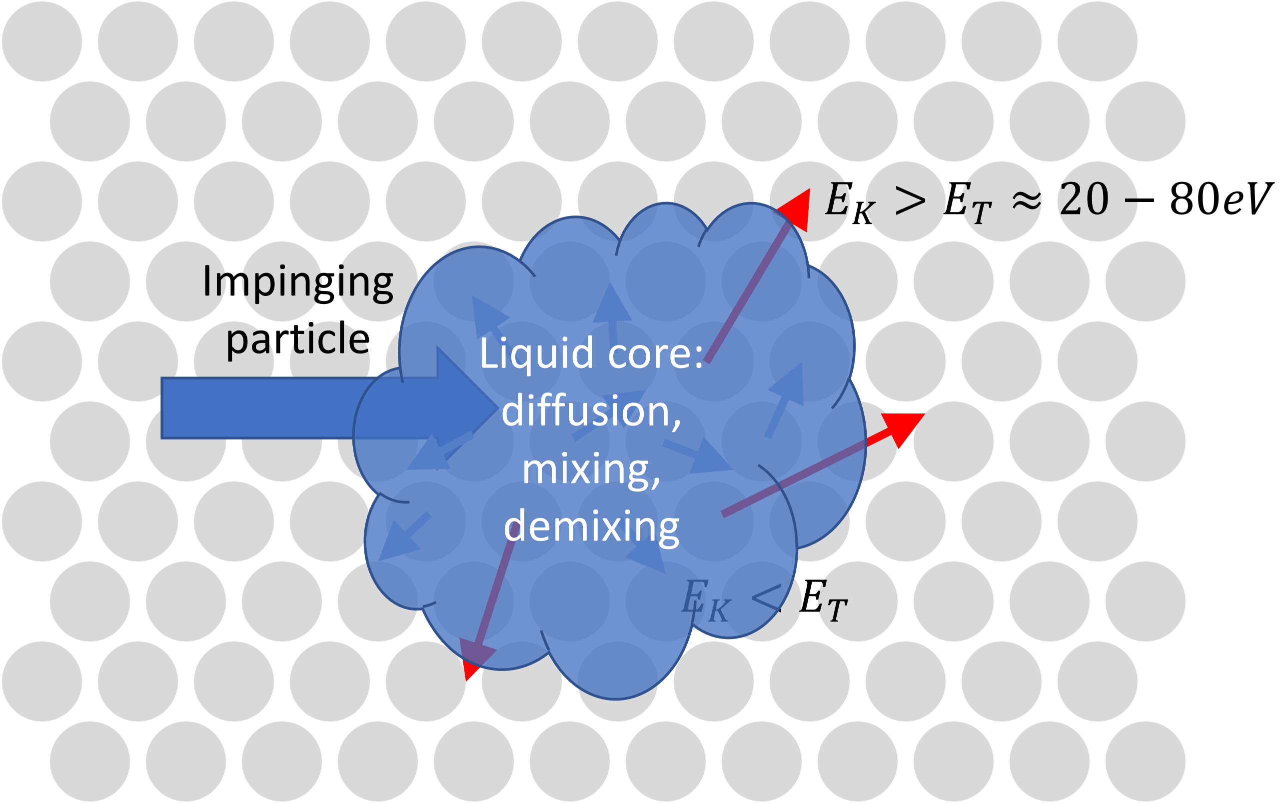
- Rate theory
- Reaction-diffusion
- Phase field
- Kinetic Monte Carlo
- Accelerated MD
- …

Modeling the fate of defects over long times is very challenging.

A closer look at thermal spikes

Thermal phase, 2-3ps

Ballistic phase, ≲1ps

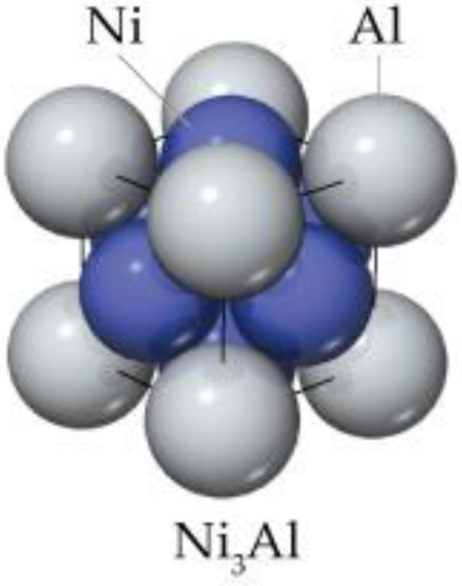


A closer look at thermal spikes

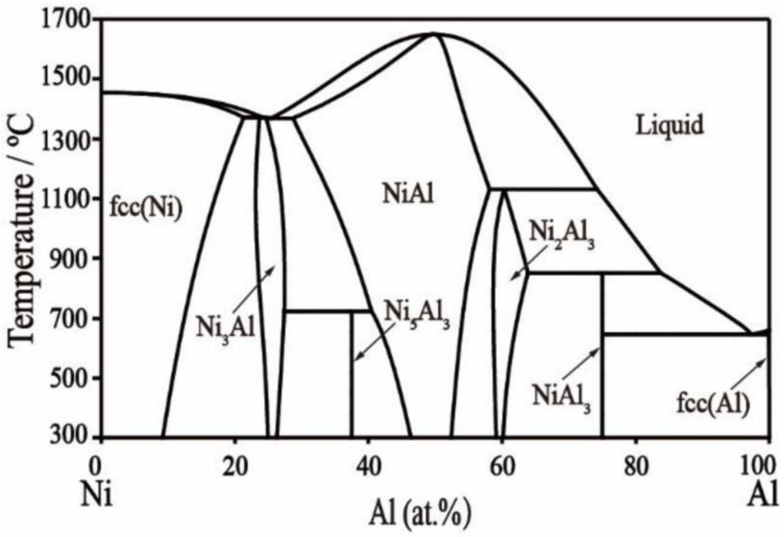
Resolidification, ~5ps

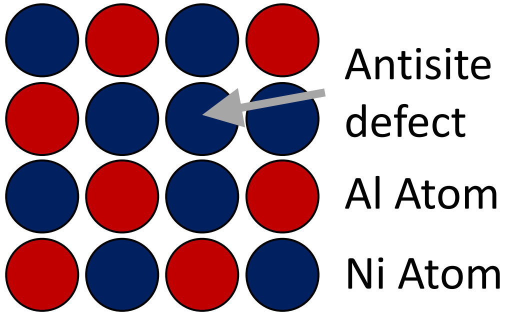
Thermal spikes in Ni_3Al

Lowest energy state is an $L1_2$-ordered compound

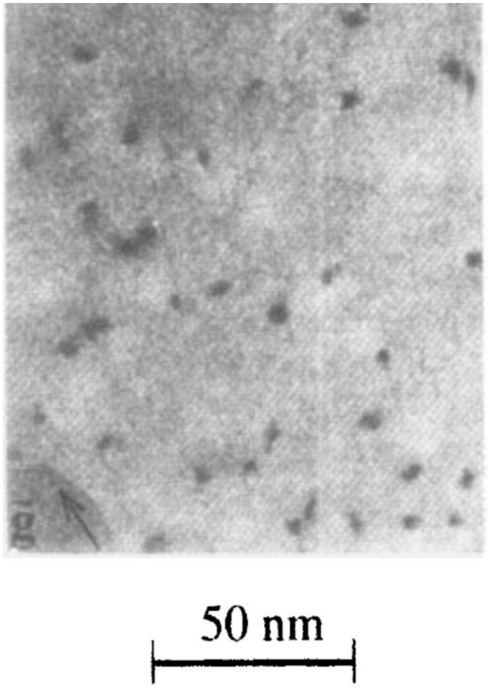


Rapid quenching yields a metastable FCC solid solution



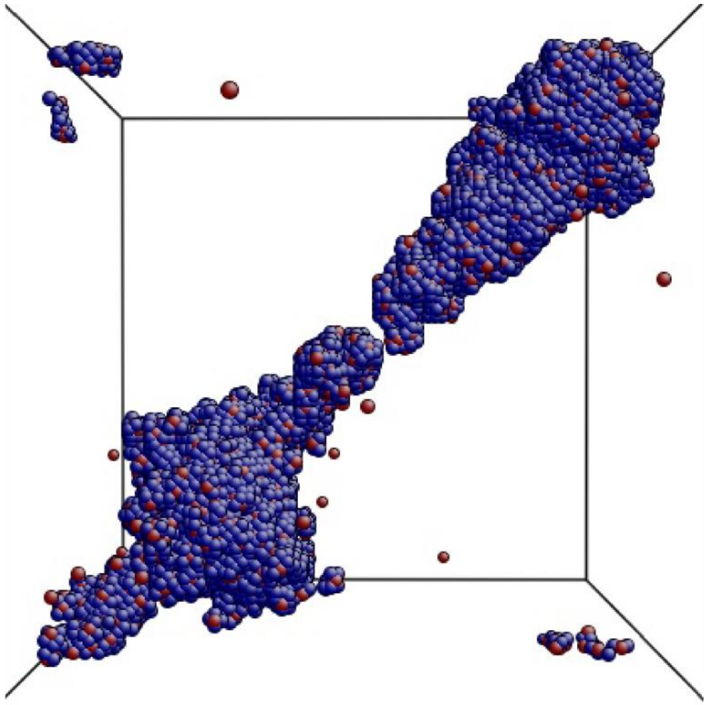


Disordered zones (dark) in TEM of ion irradiated Ni_3Al

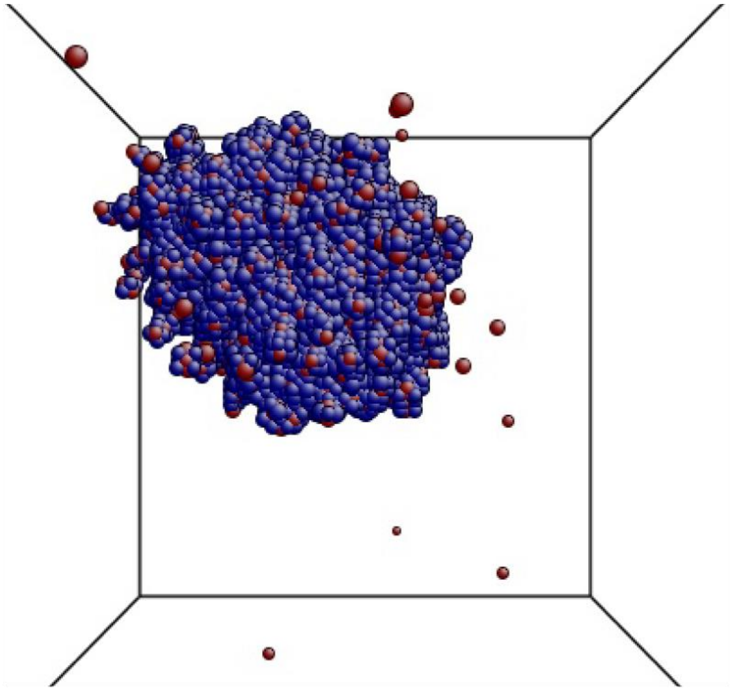


S. Muller *et al.*, Phil. Mag. A **75**, 1625 (1997)

Thermal spikes shapes in Ni_3Al



C=0.39

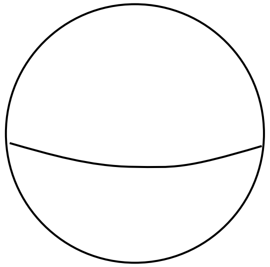


C=0.54

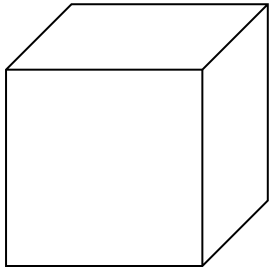
Compactness

$$C = (36\pi)^{1/3} \frac{V^{2/3}}{S}$$

C=1

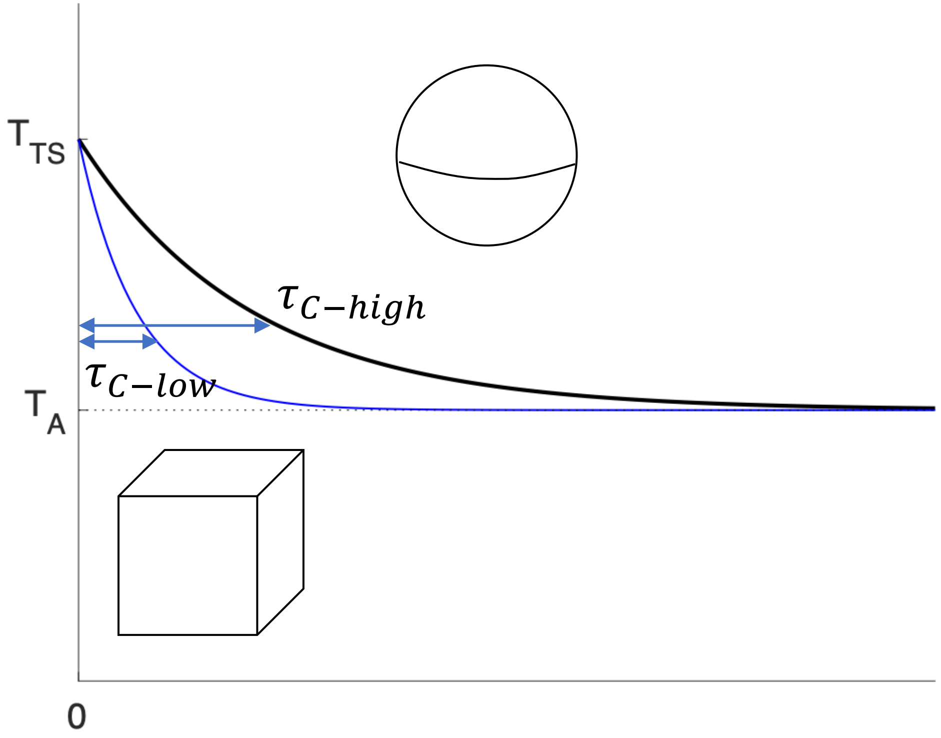


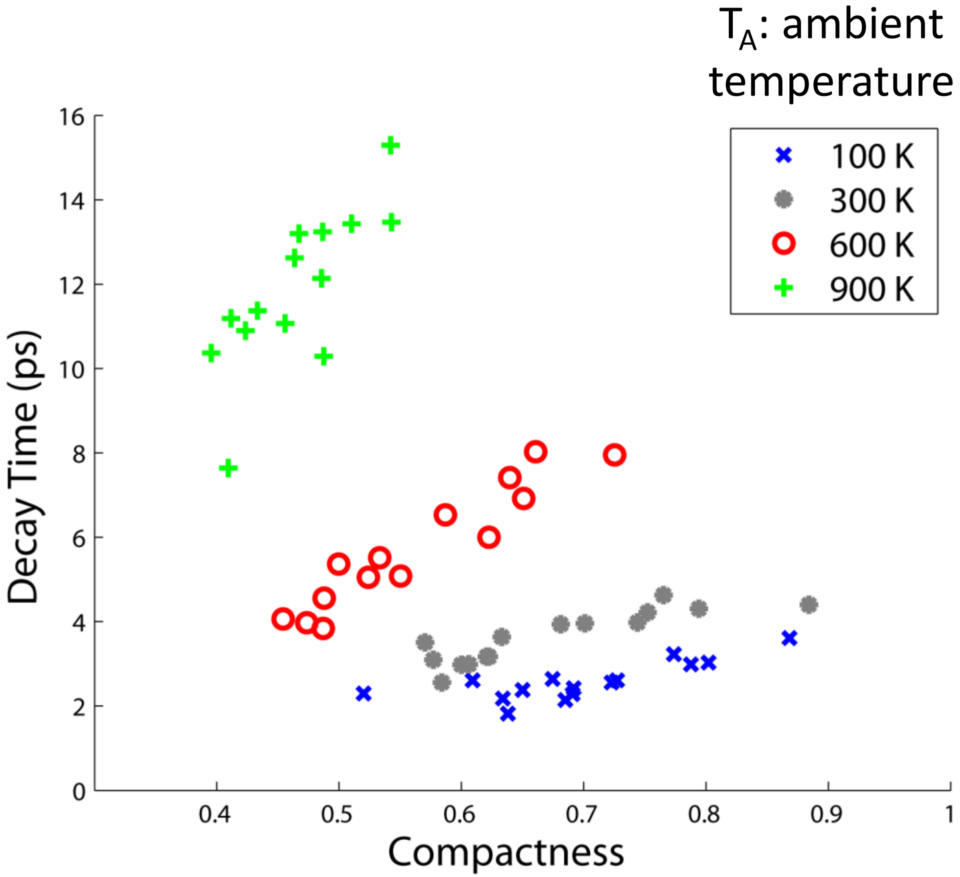
C=0.806



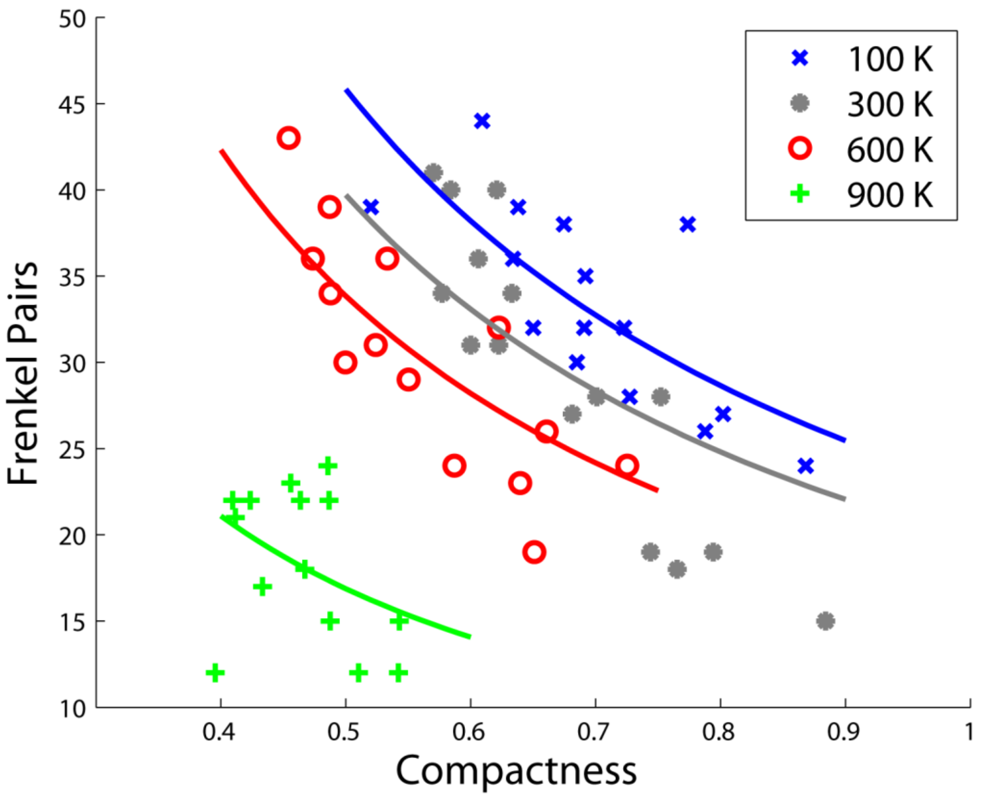
S. A. Skirlo and M. J. Demkowicz, Scripta Mater. **67**, 724 (2012)

Compact thermal spikes cool slower



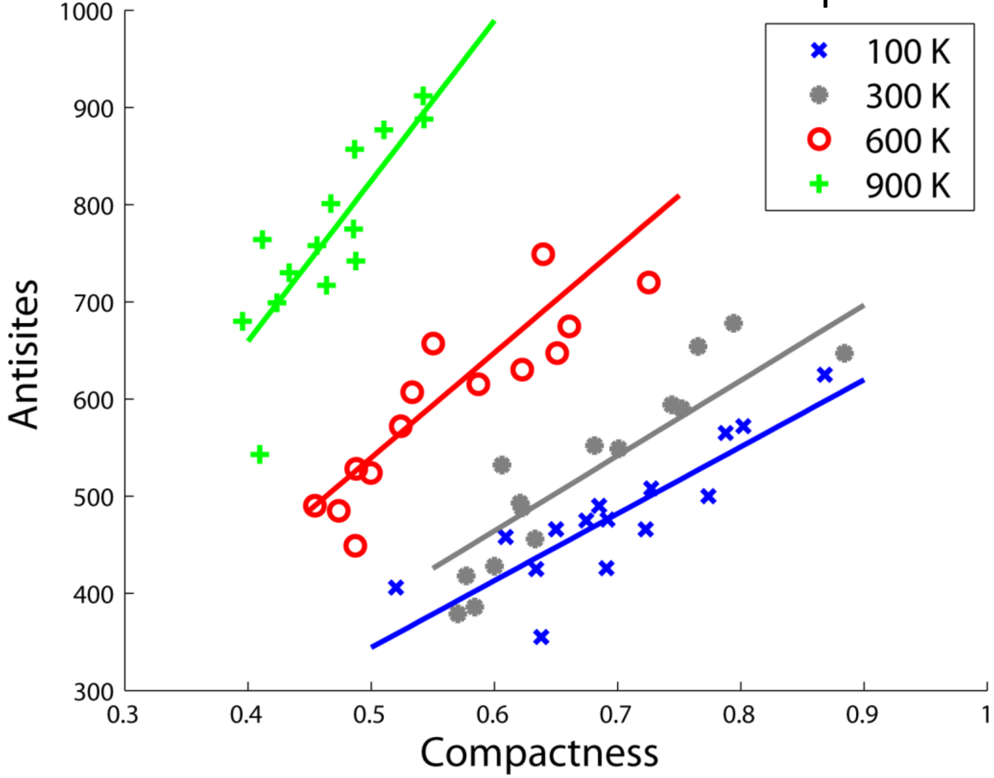


Defect formation depends on compactness



Long decay times favor Frenkel pair recombination

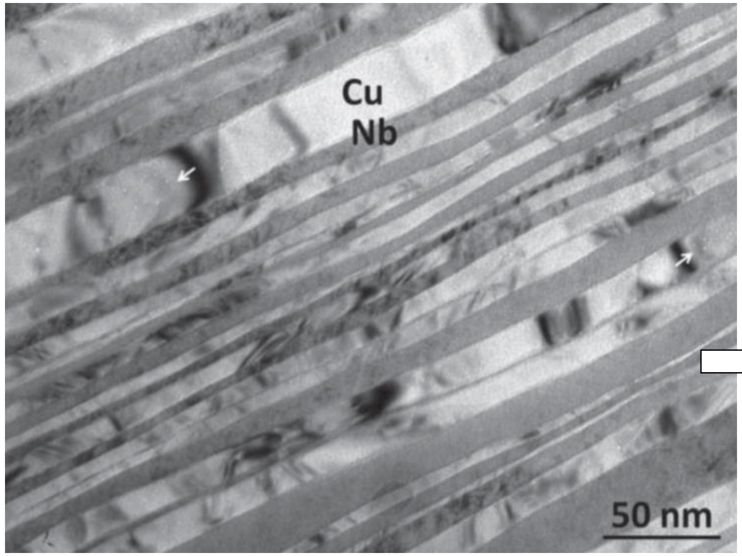
T_A: ambient temperature



Long decay times favor liquid phase disordering

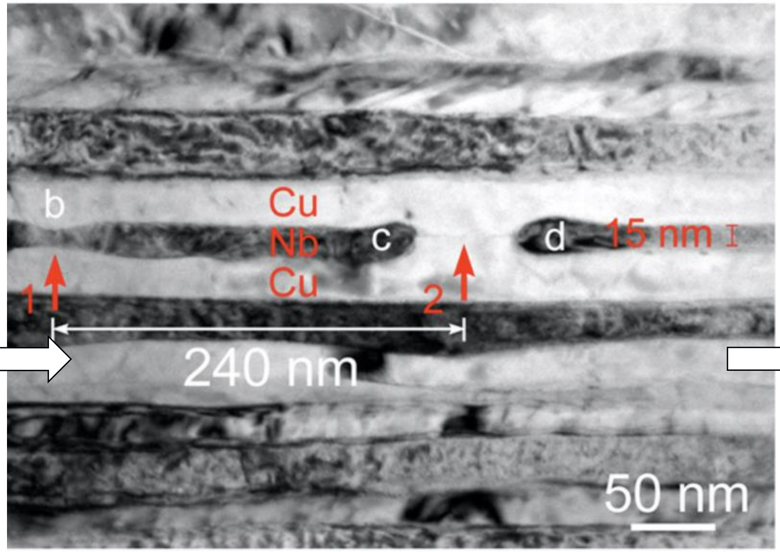
Metal multilayer nanocomposites: radiation resistant, but metastable

Nearly void free after irradiation



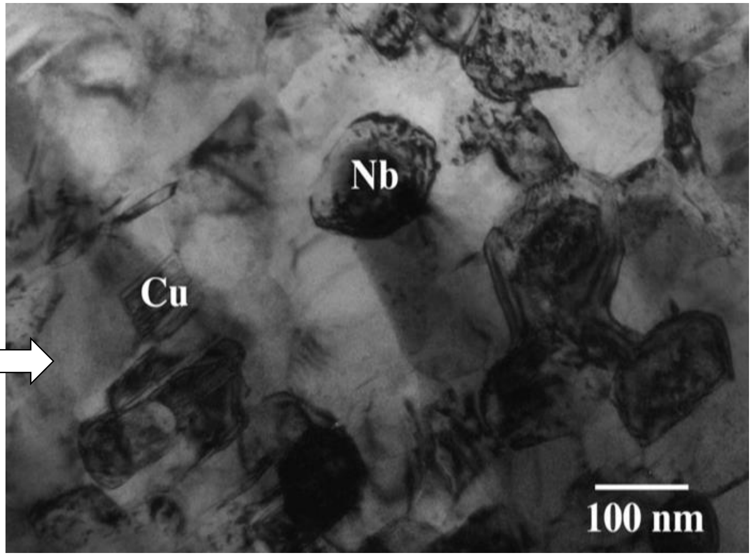
200keV He^+, 450°C, 3 dpa

$\Delta F \uparrow$



$\Delta F \downarrow$

Spherodization after annealing



Anneal at 700°C, 1hr

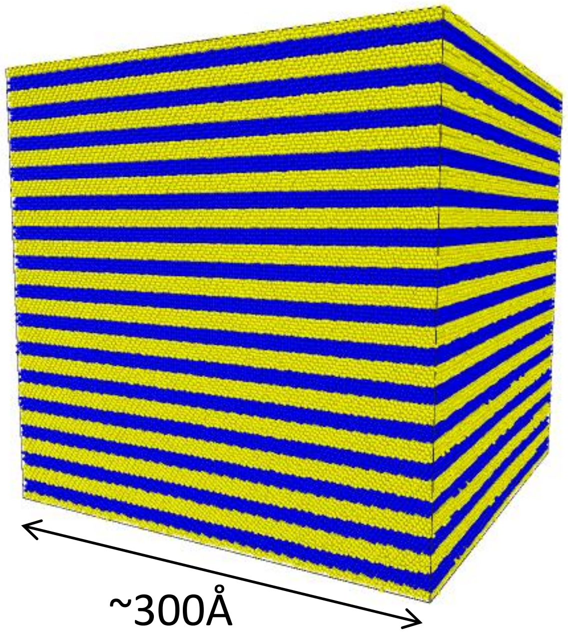
Can thermal spikes initiate layer pinchoff?

W. Han *et al.*, Adv. Mater. **25**, 6975 (2013)

S. Zheng *et al.*, APL **105**, 111901 (2014)

A. Misra and R. G. Hoagland, J. Mater. Res. **20**, 2046 (2005)

Molecular dynamics of metal multilayers

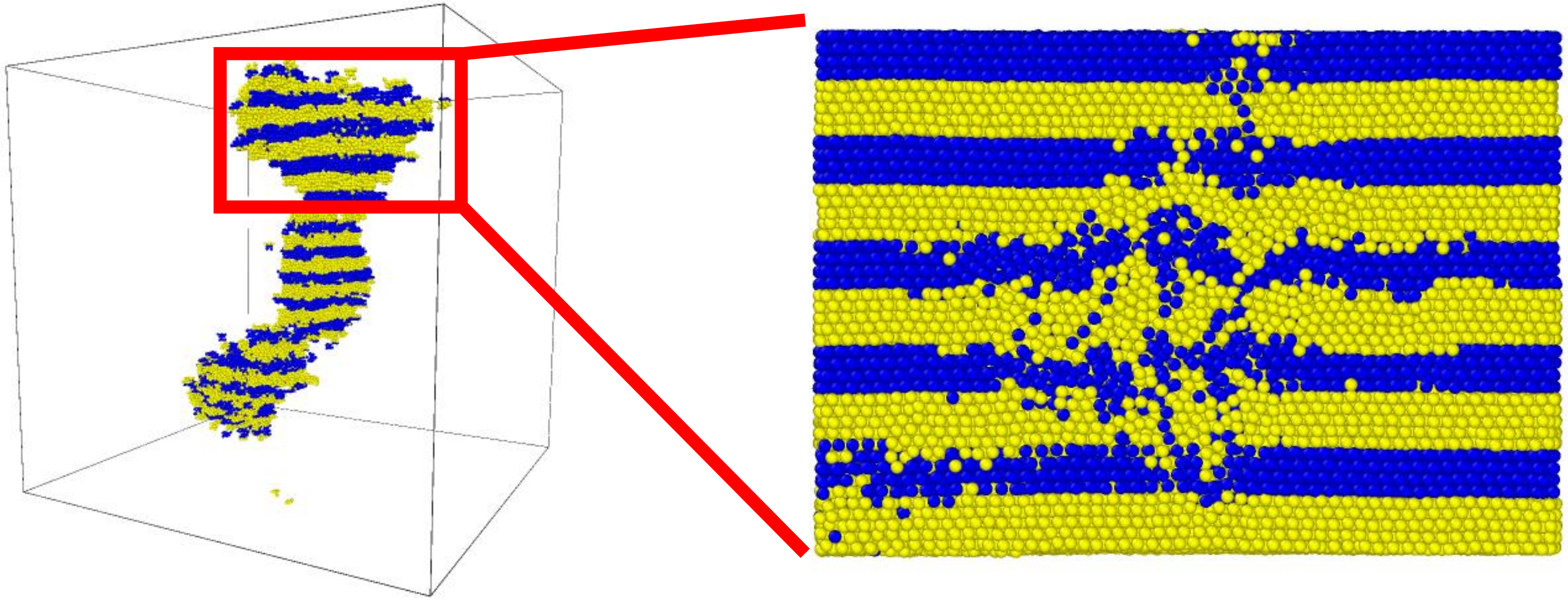


~300Å
~2 million atoms
Layer thickness: 10Å

Cu: Yellow
Nb: Blue

Thermal Spike (~0.5 ps)

After thermal spike (~200ps)



PKA energy: 100keV

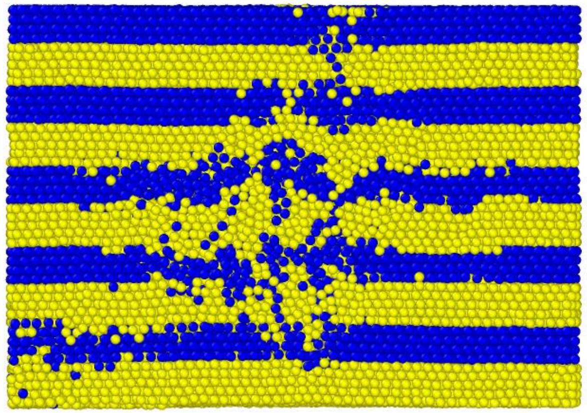
Liquid phase mixing
within thermal spikes

L. Zhang and M. J. Demkowicz, APL **103**, 061604 (2013)

Phase field modeling of final microstructure

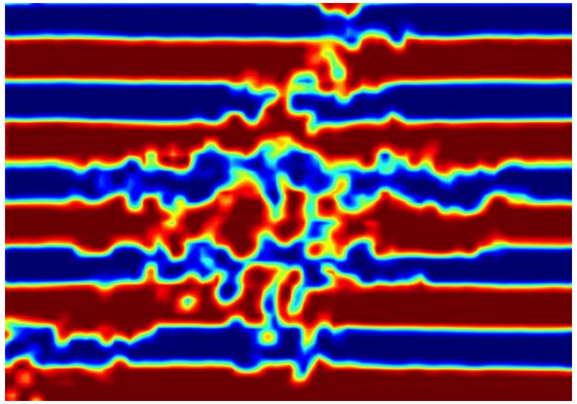
Structure from MD

Cu
Nb



Phase field model

Cu
Nb

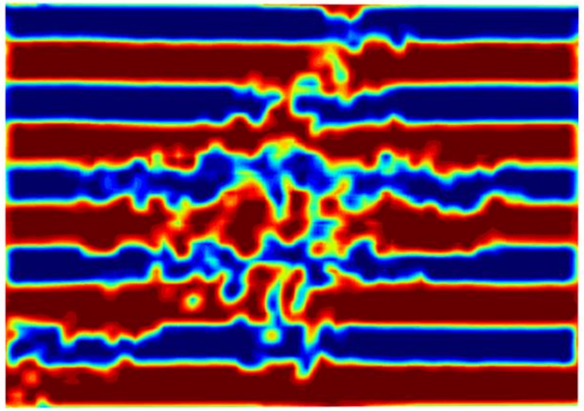


- Cahn-Hilliard equation
- Local free energy density function:

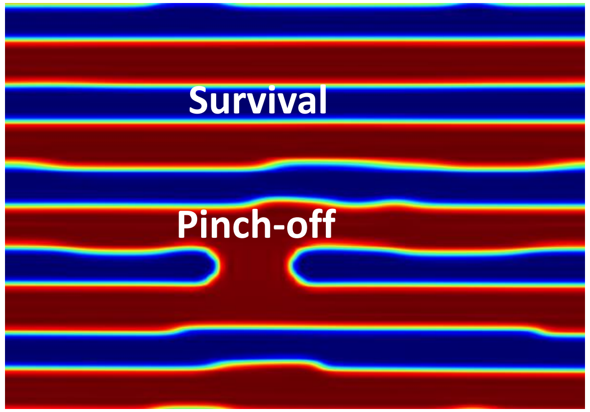
$$f(\phi) = 4\Delta f \phi^2 (1 - \phi)^2$$

- Interfacial thickness is set to be ~2Å

Extensive
de-mixing in
solid state

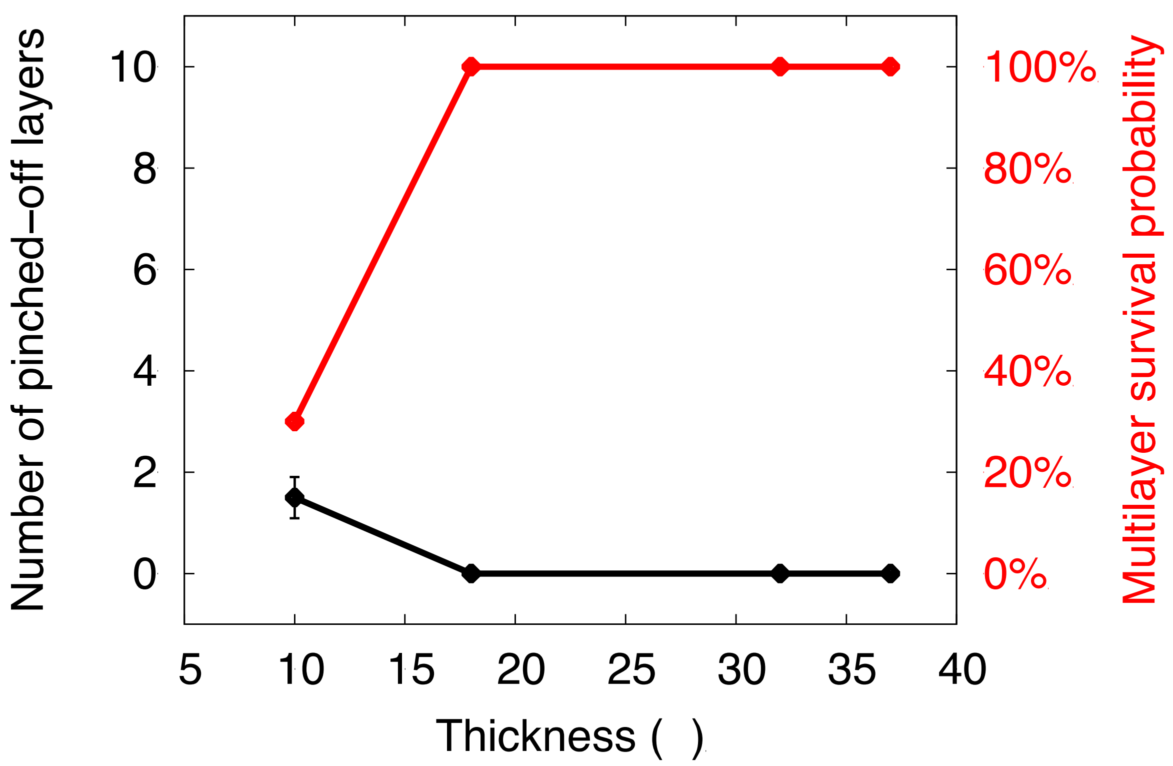


Cu
Nb

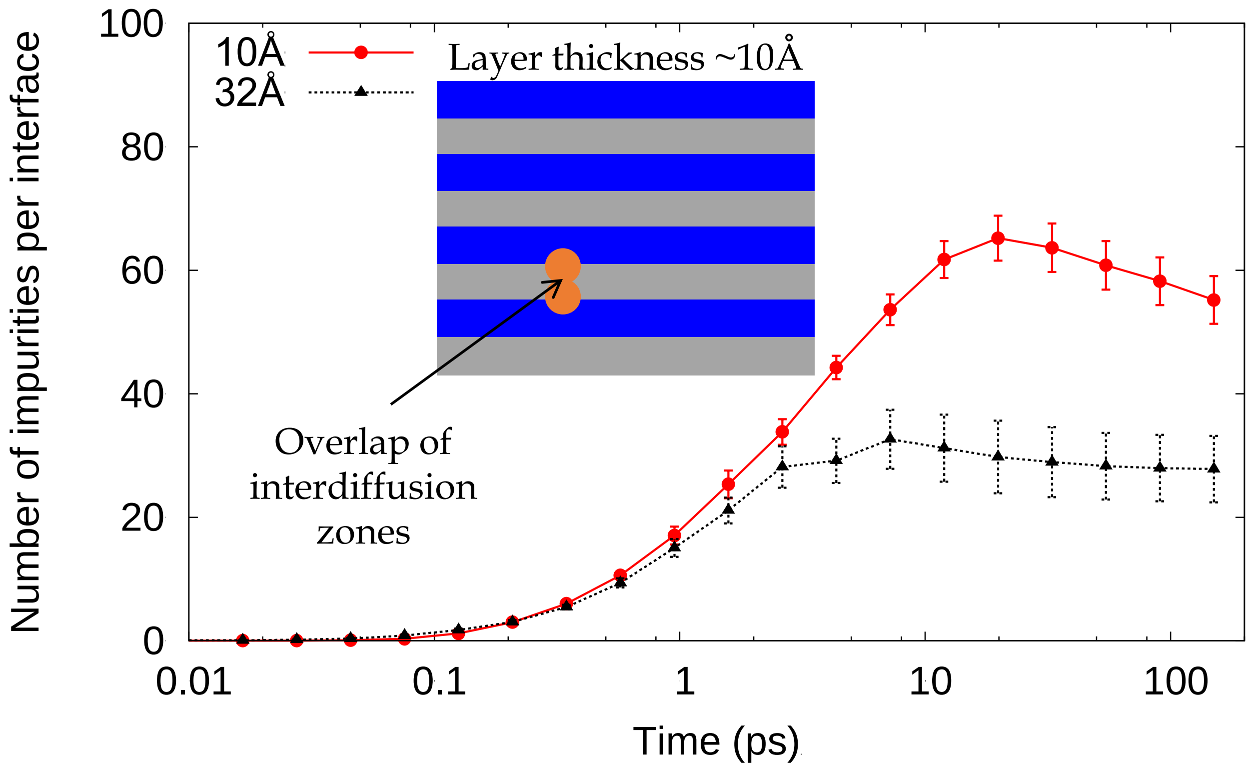


We are interested in the
final state of multilayers
(pinched off or not?)

No pinchoff above a layer thickness of 2nm



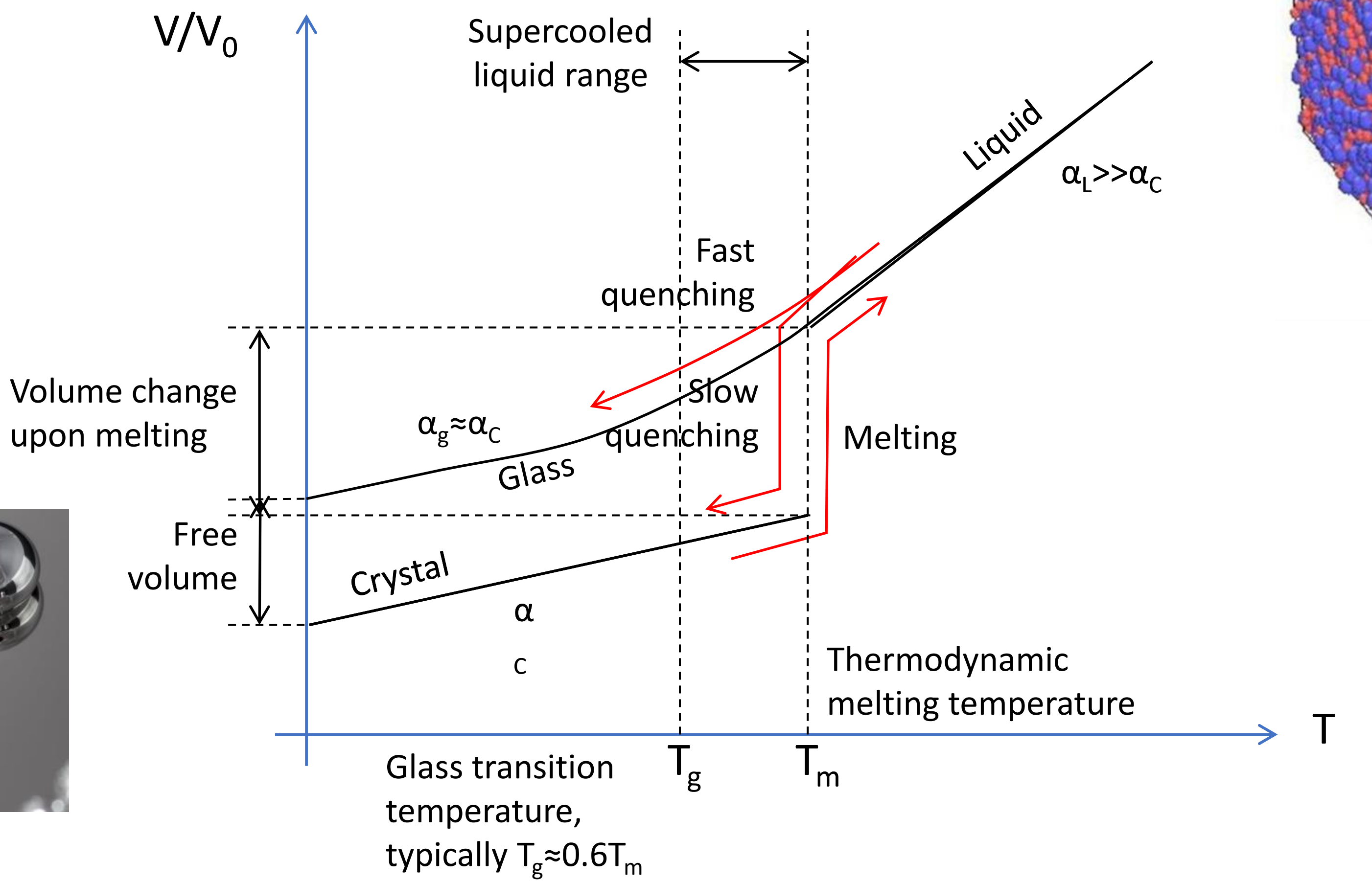
10 independent simulations were performed for each layer thickness



Over a typical thermal spike lifetime, the liquid-phase interdiffusion distance is ~1nm

Composites with layer thickness above 2nm remain metastable under irradiation

Metallic glasses



~cm-scale BMG samples

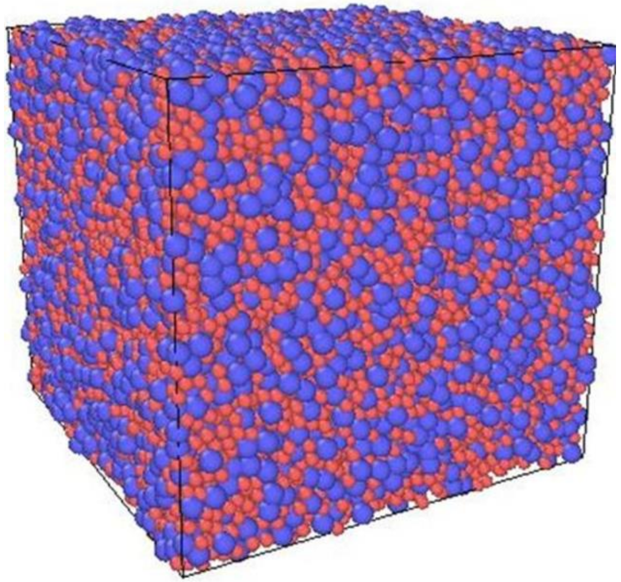
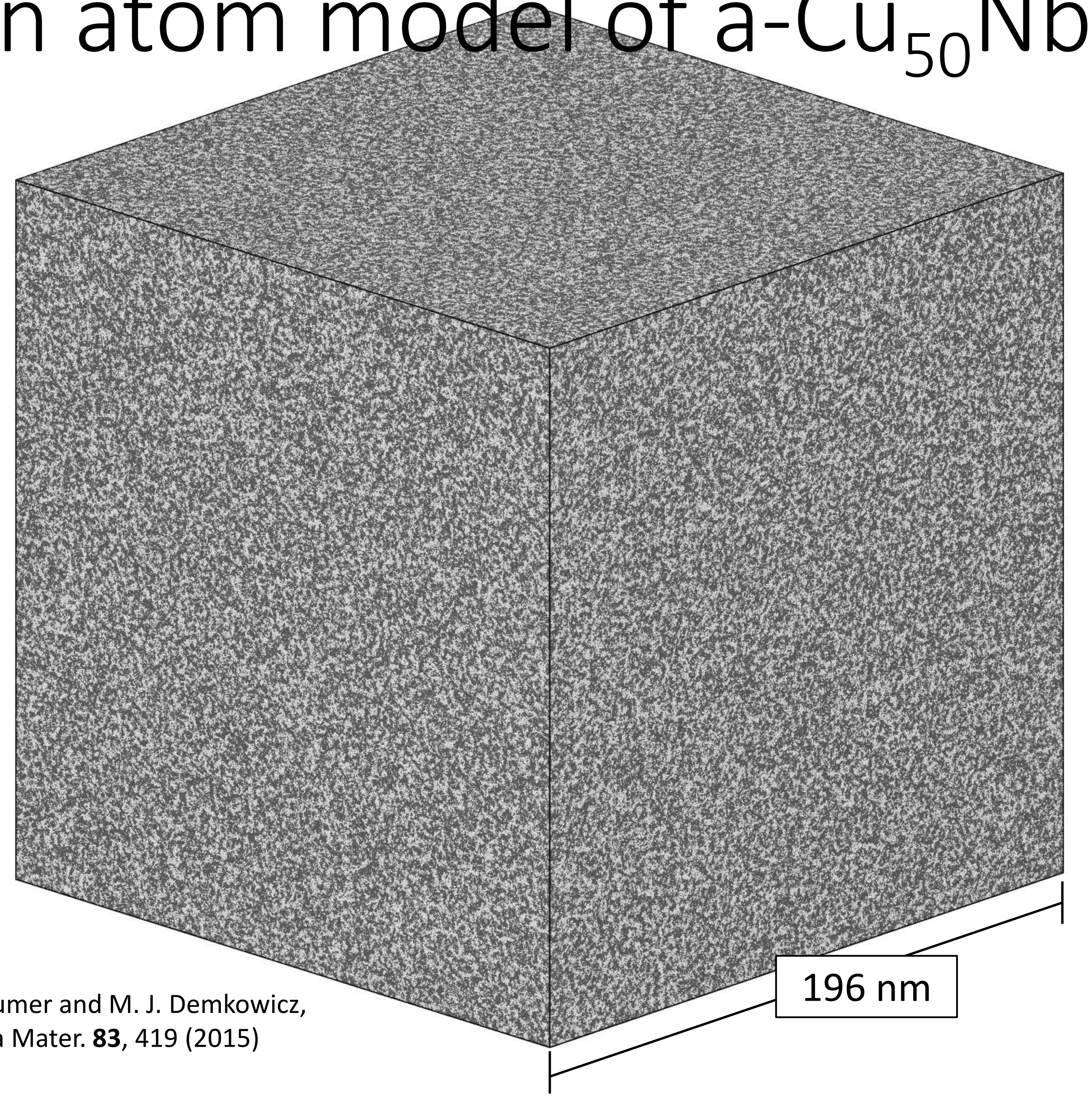


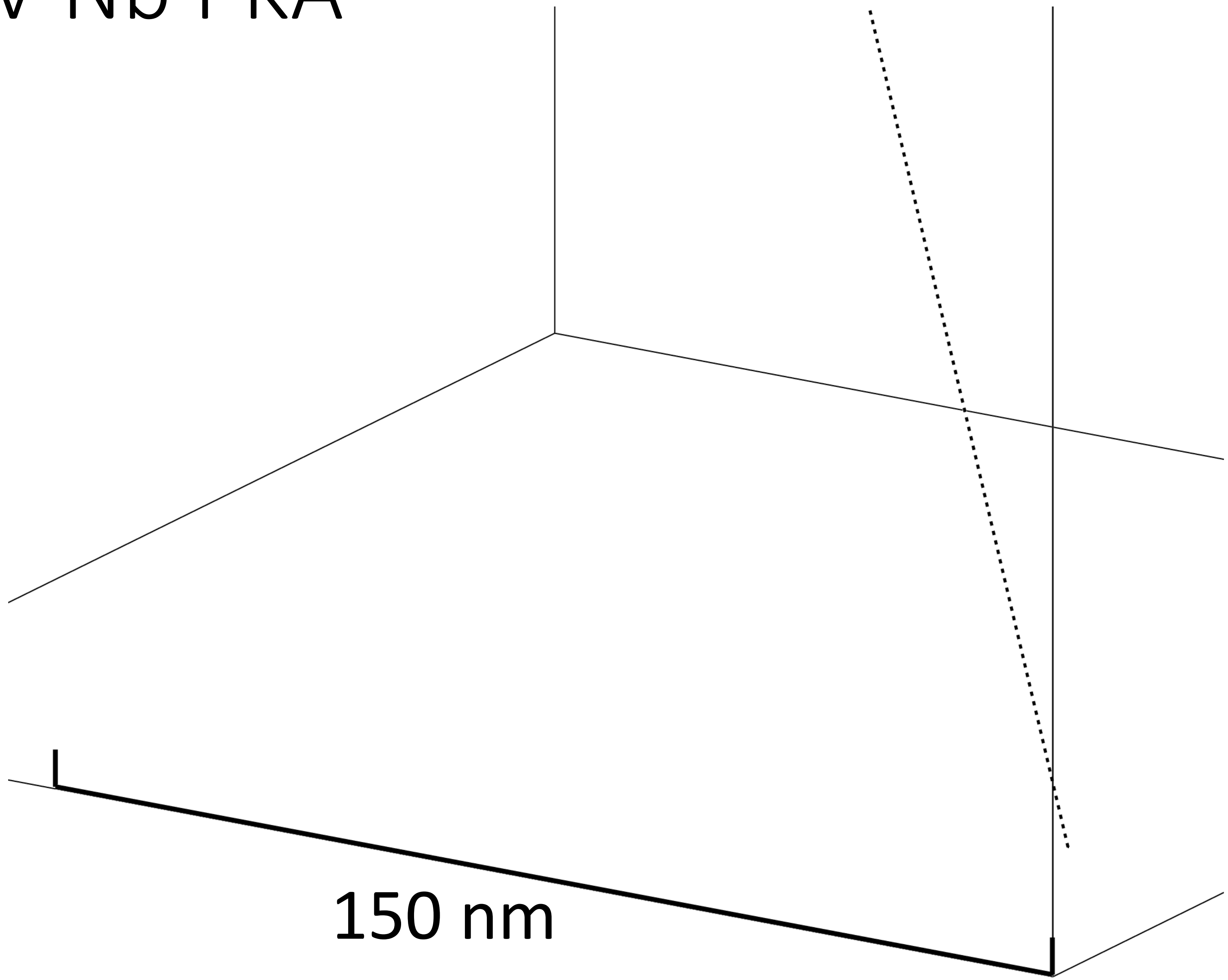
Image credit: K. Flores (WUSTL)

474-million atom model of a-$Cu_{50}Nb_{50}$

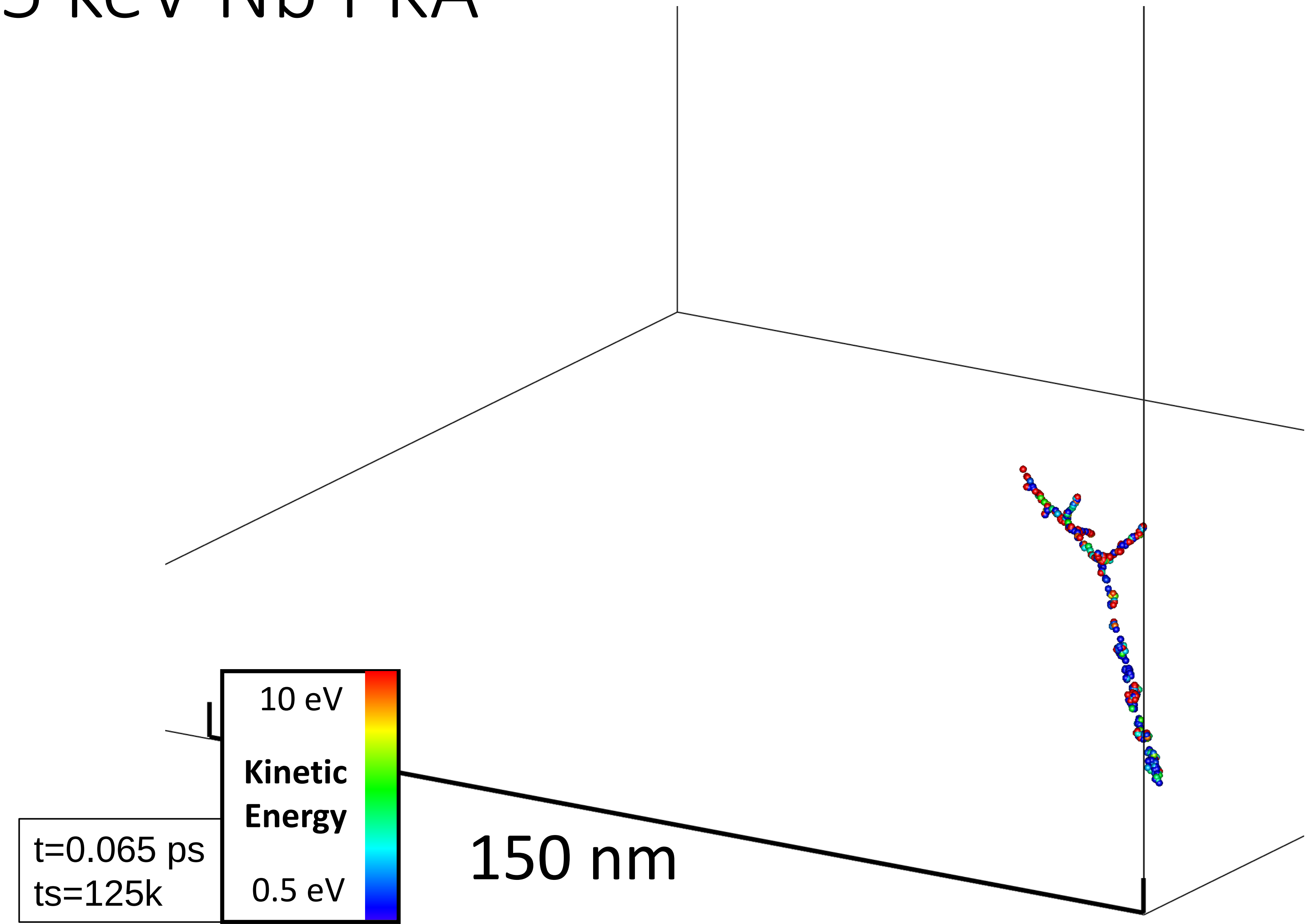


R. E. Baumer and M. J. Demkowicz,
Acta Mater. **83**, 419 (2015)

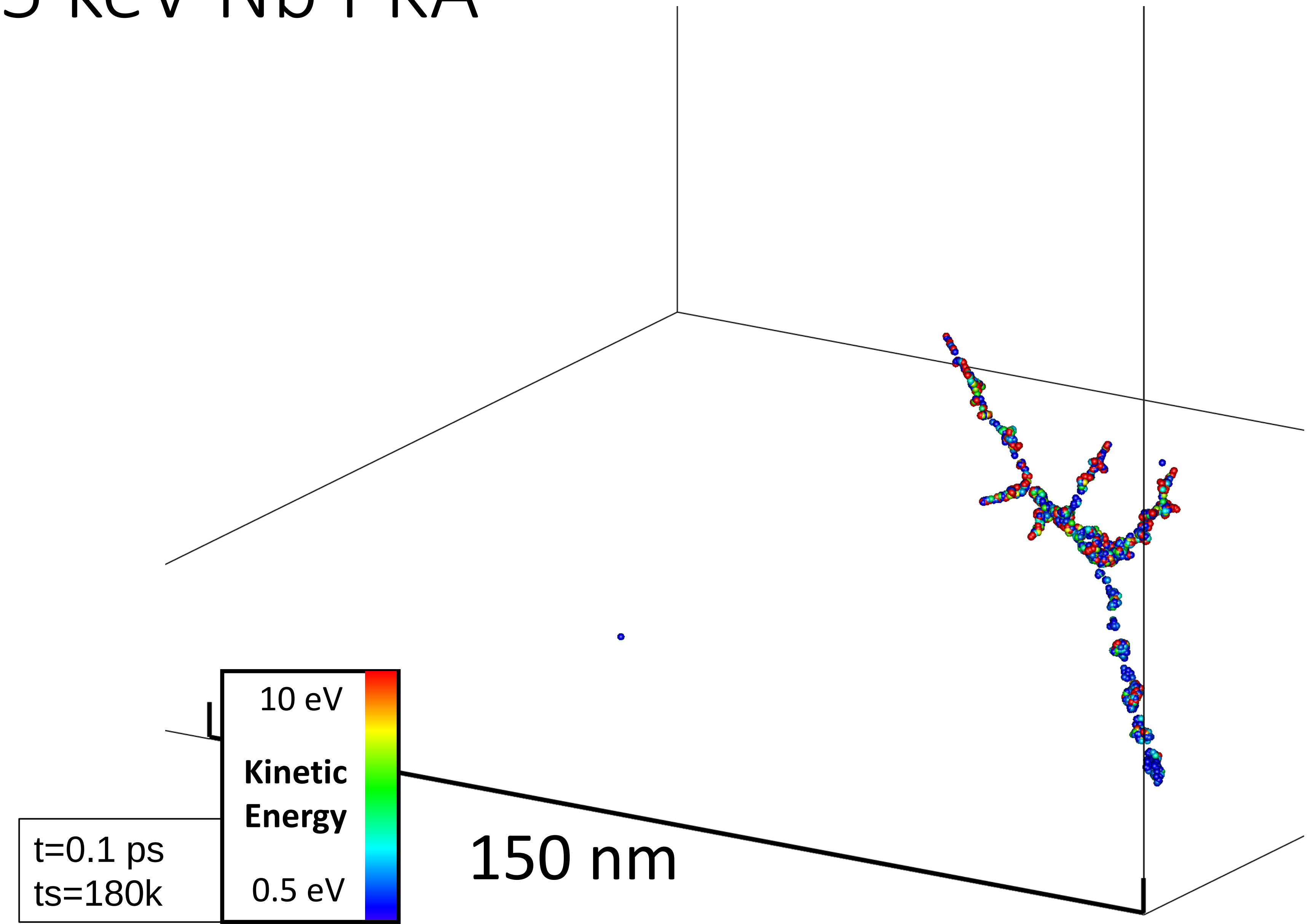
475 keV Nb PKA



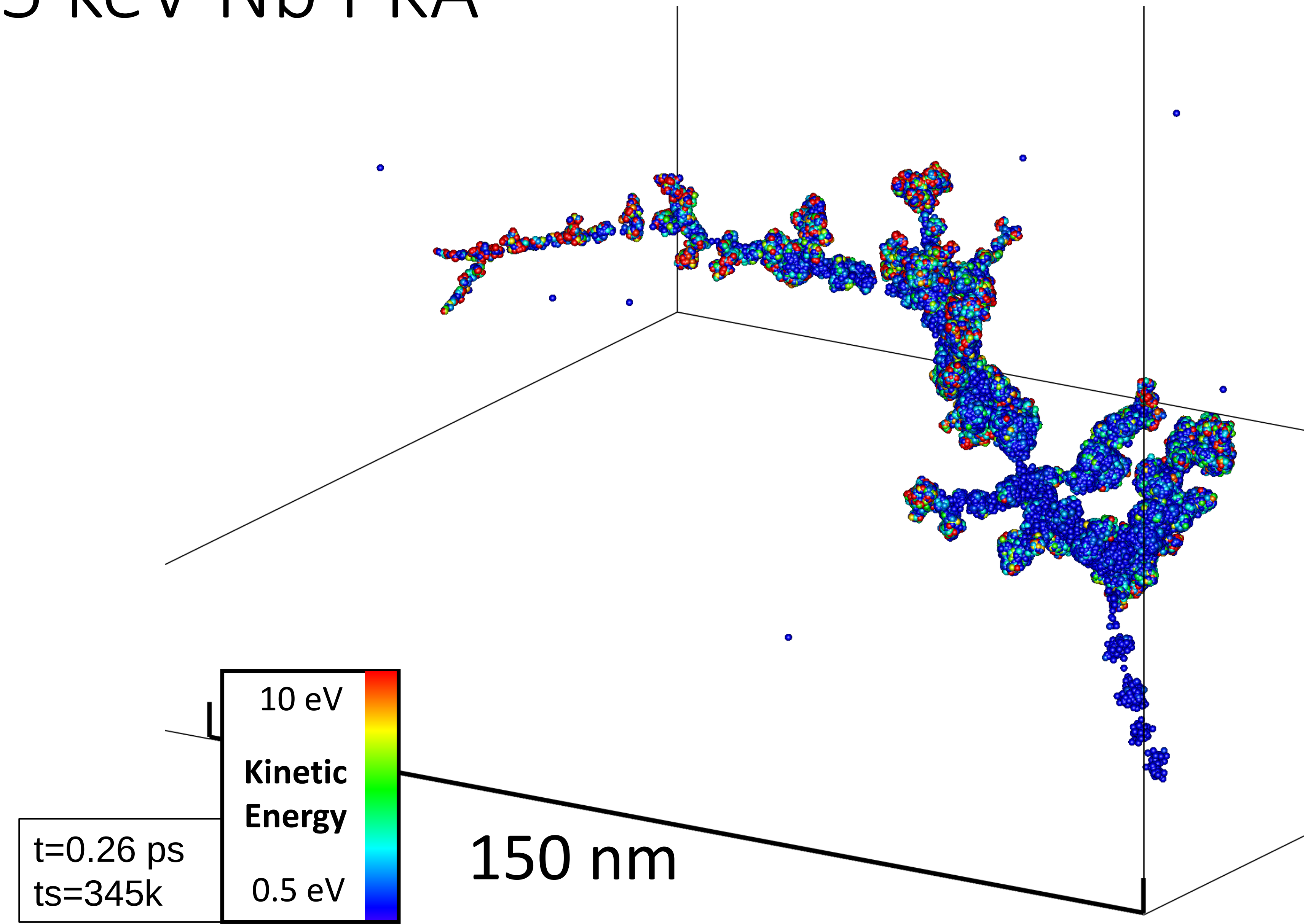
475 keV Nb PKA



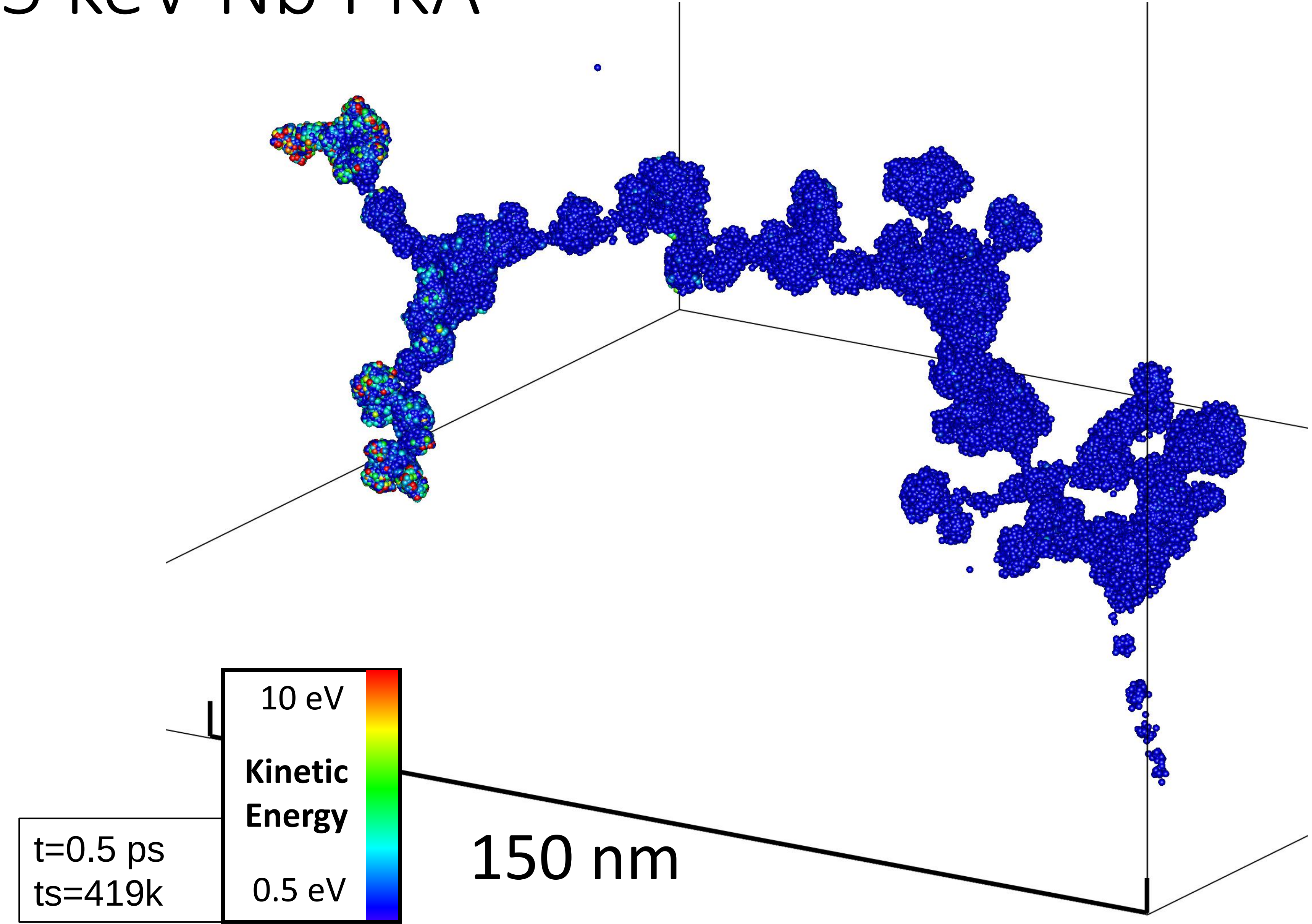
475 keV Nb PKA



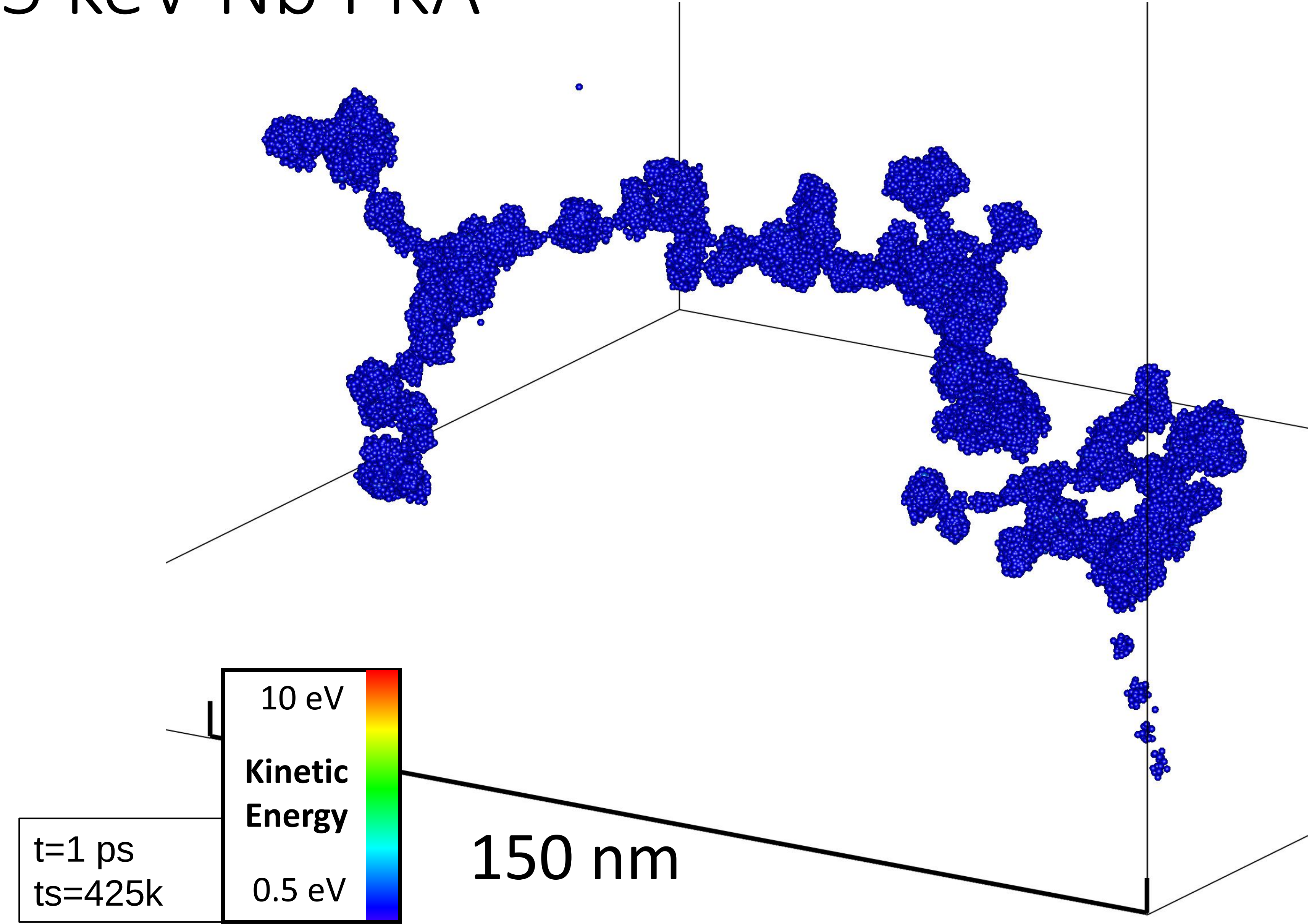
475 keV Nb PKA



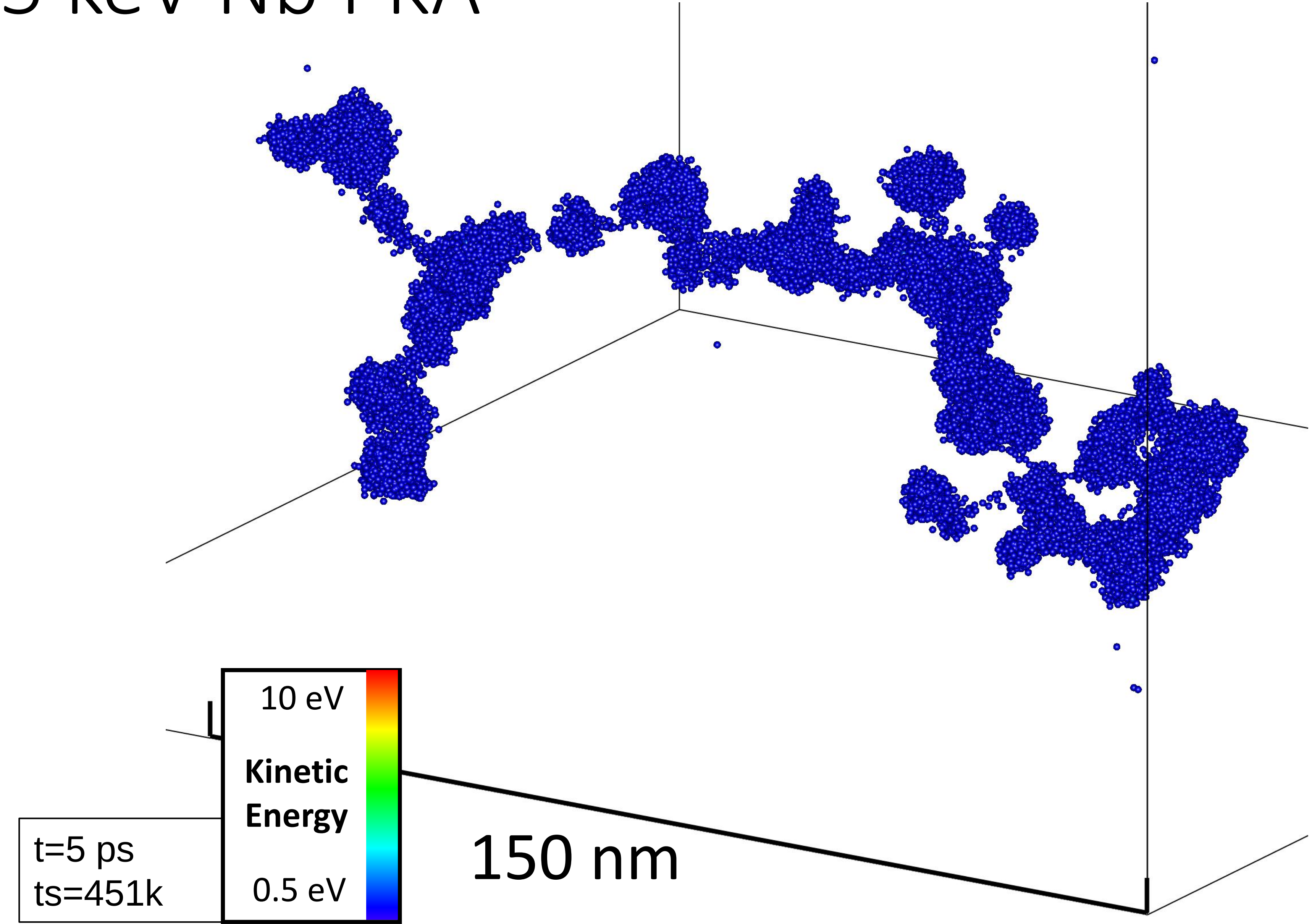
475 keV Nb PKA



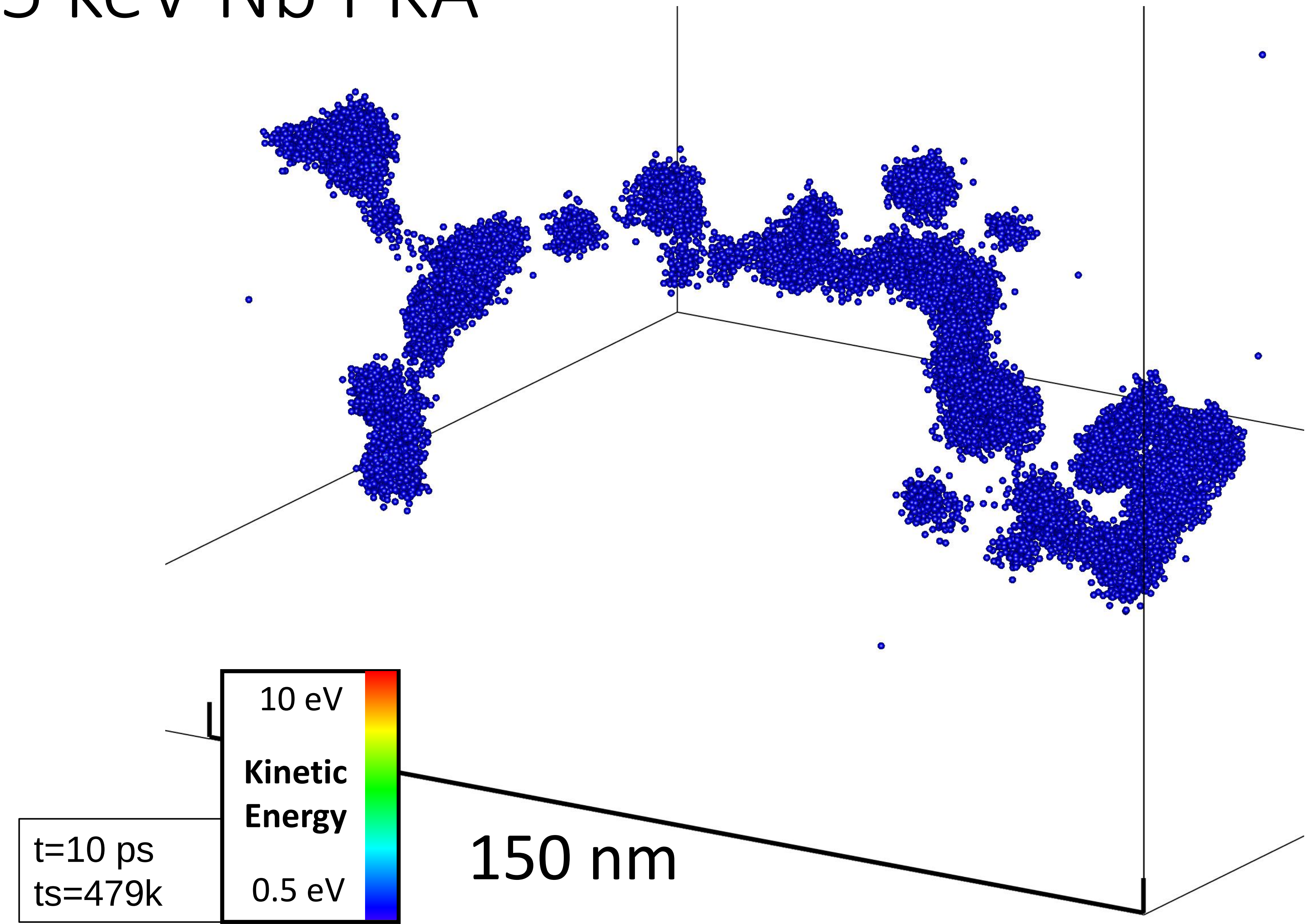
475 keV Nb PKA



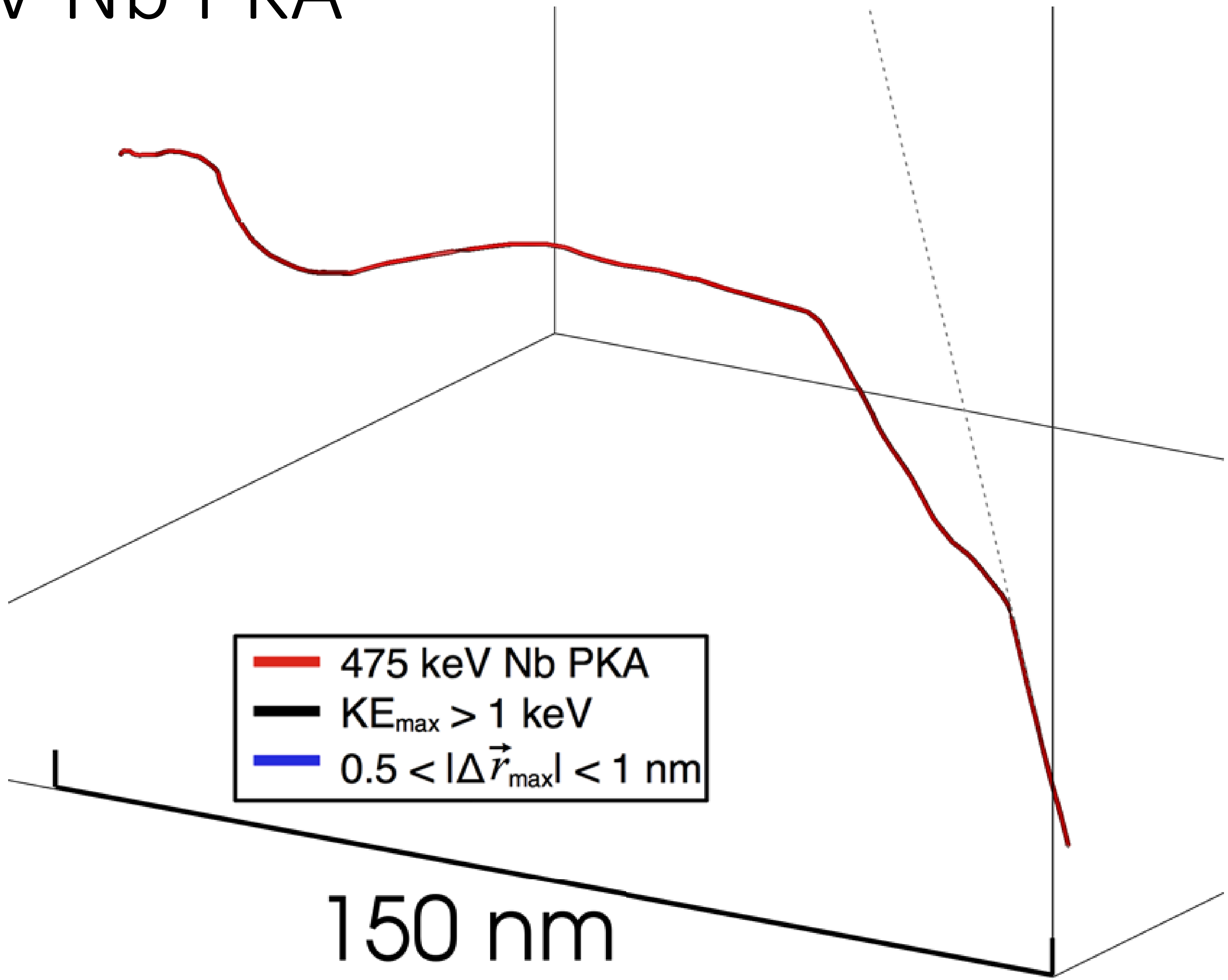
475 keV Nb PKA



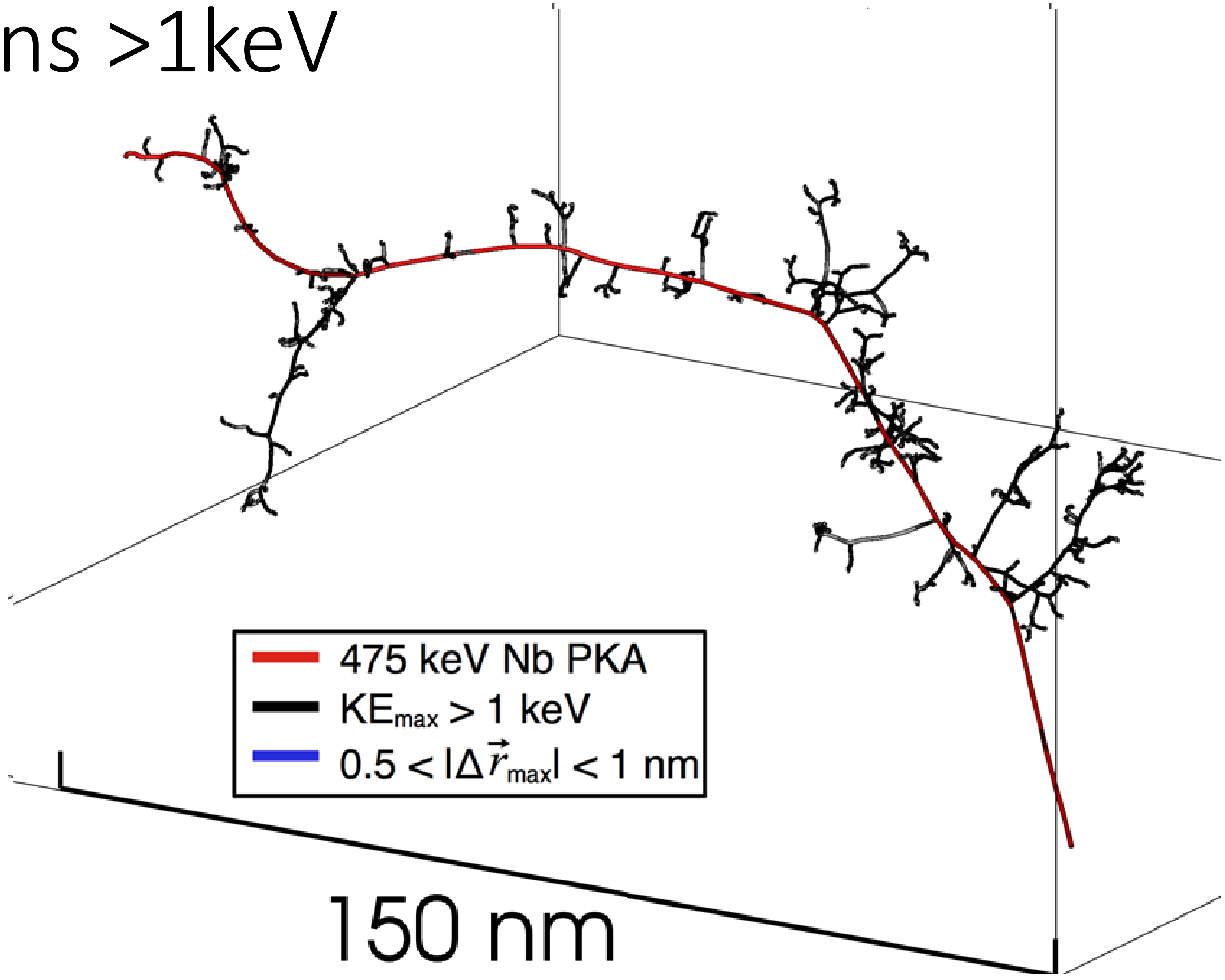
475 keV Nb PKA



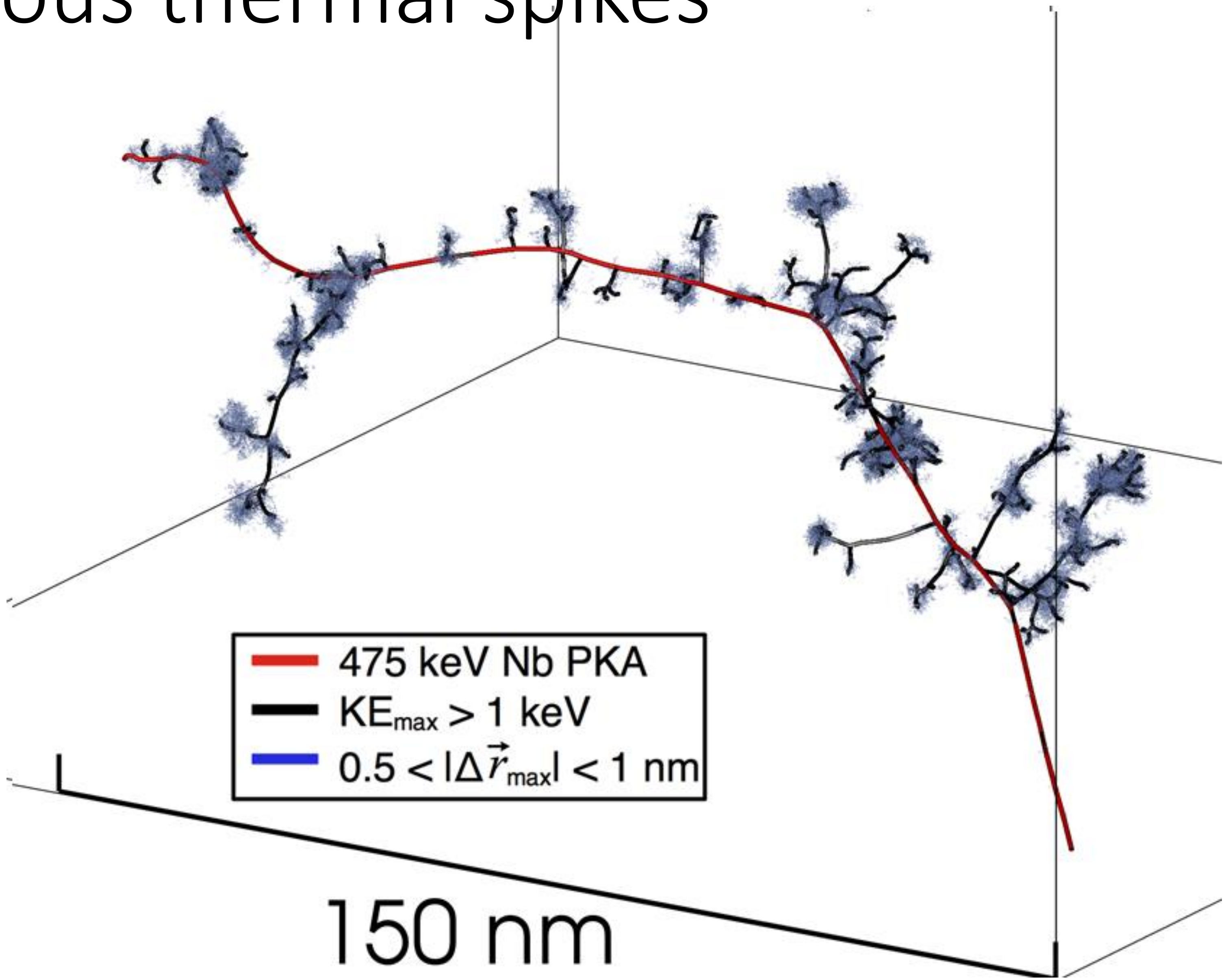
475 keV Nb PKA



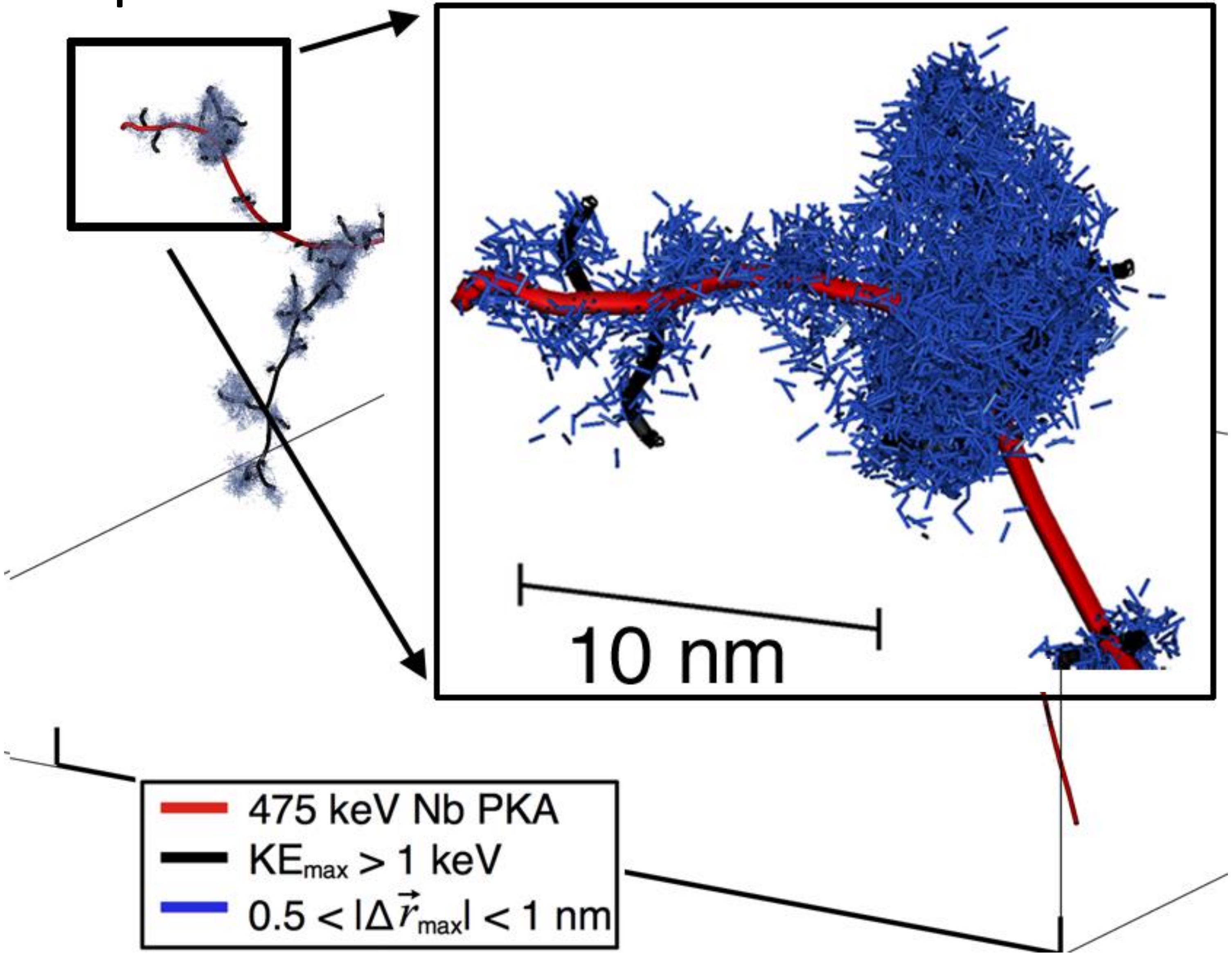
90% of PKA energy dissipated in binary collisions >1keV



Numerous thermal spikes



Thermal spikes are ~10nm wide



Voxel field analysis

- **Averages**: Potential energy, density, and stress fields

$$pe = \frac{1}{N}\sum_{i=1}^{N} pe_i \qquad \rho = \frac{1}{V}\sum_{i=1}^{N} m_i \qquad \sigma_{kl} = \frac{1}{V}\sum_{i=1}^{N} (\sigma_{ab}V^*)_i$$

- **Fitting**: Temperature field

$$p(ke) = 2\left(\frac{ke}{\pi}\right)^{1/2}\left(\frac{1}{k_B T}\right)^{3/2}\exp\left(-\frac{ke}{k_B T}\right)$$

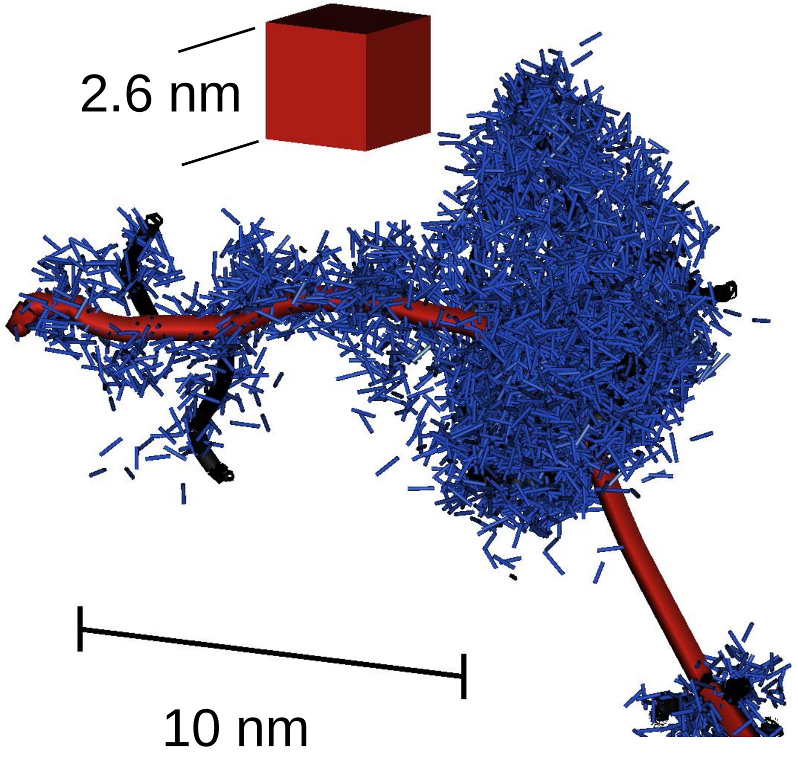
- **Derived:** Strain field

$$\varepsilon_{ij}^{tot} = 1/2(F_{ki}F_{kj} - \delta_{ij})$$

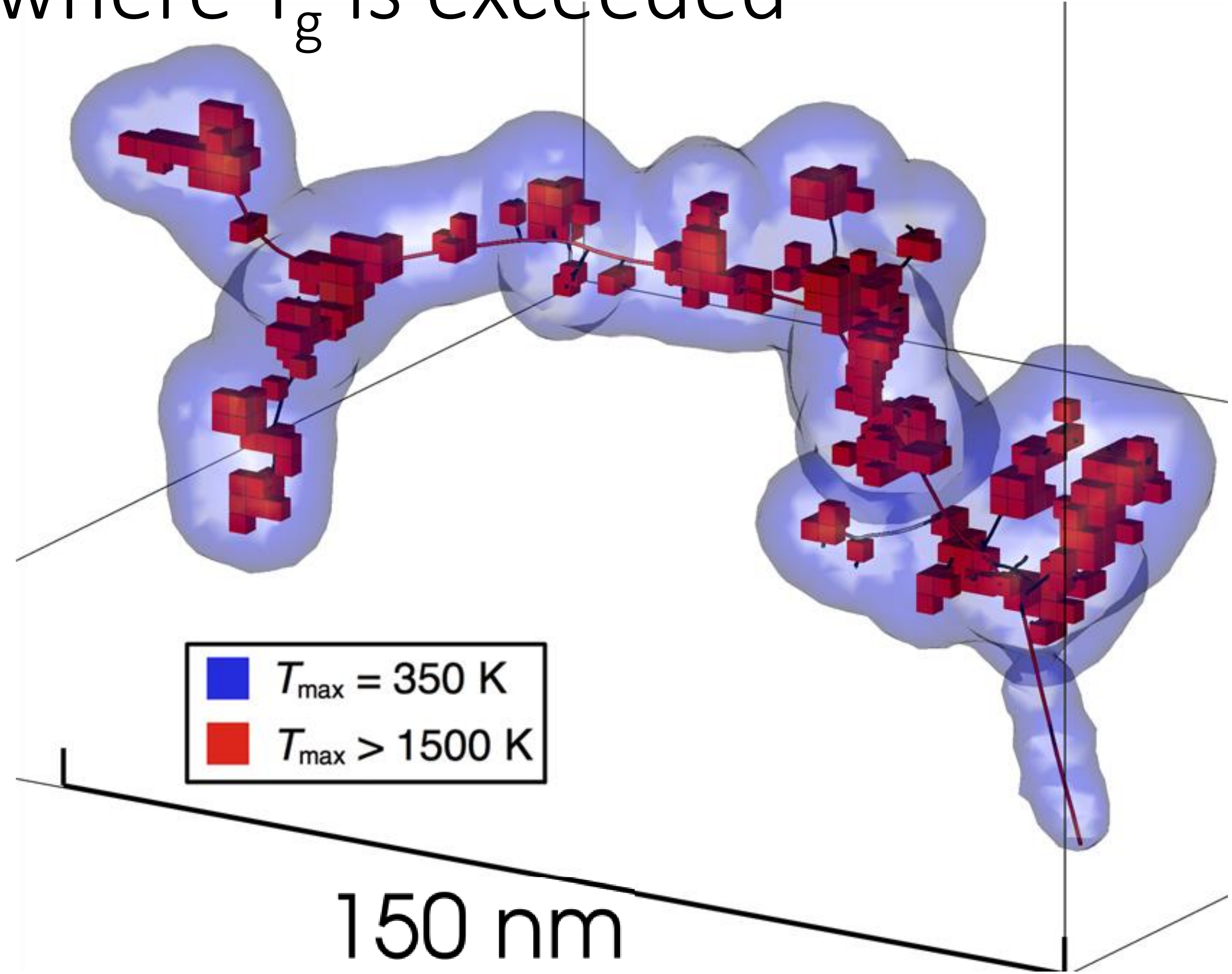
$$\varepsilon_{ij}^{p} = \varepsilon_{ij}^{tot} - \left(S_{ijkl}\sigma_{kl} + \alpha\Delta T\delta_{ij}\right)$$

- **Derived:** Diffusivity field

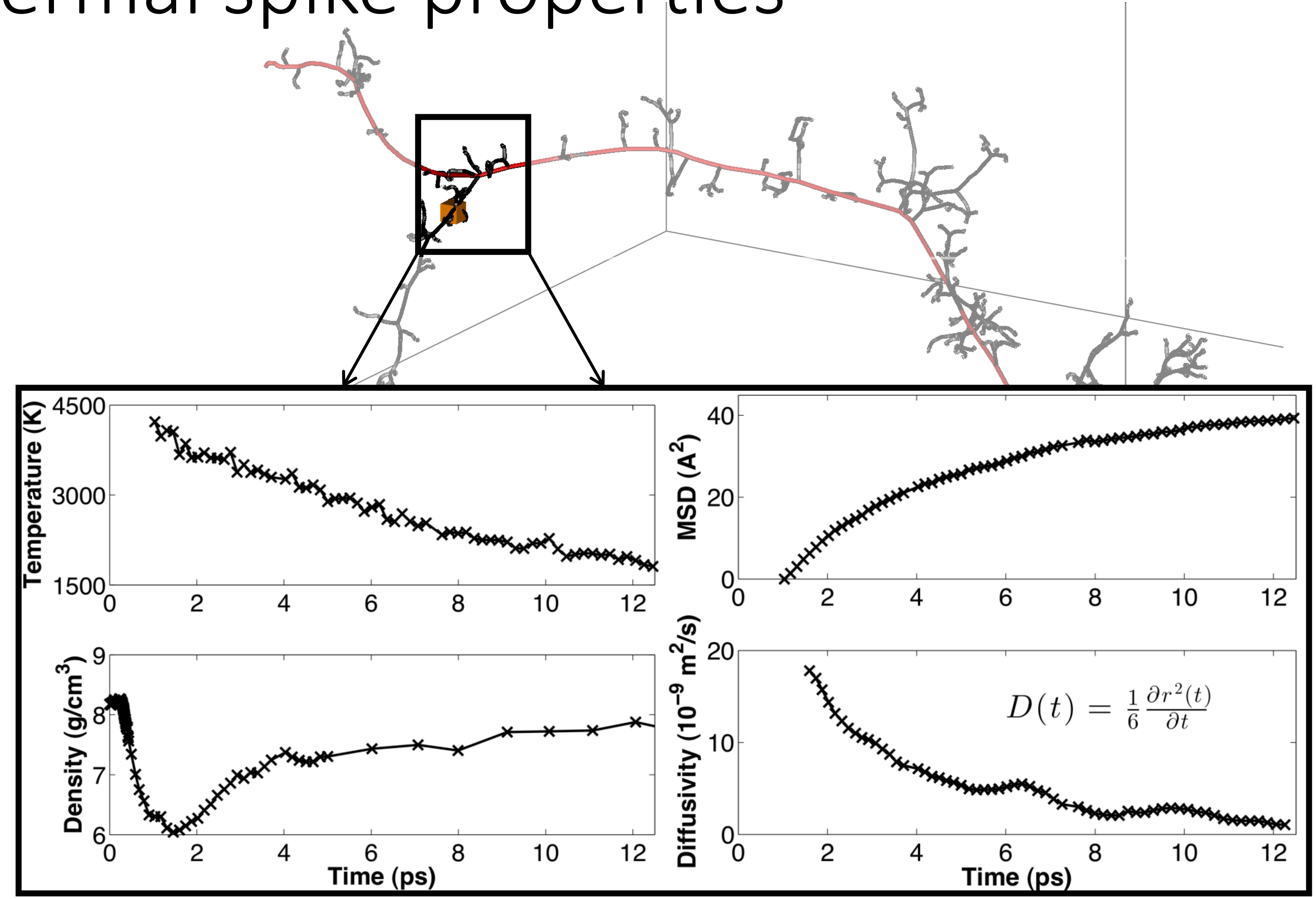
$$D(t) = 1/6\frac{\partial < r^2 >}{\partial t}$$



Zones where T_g is exceeded

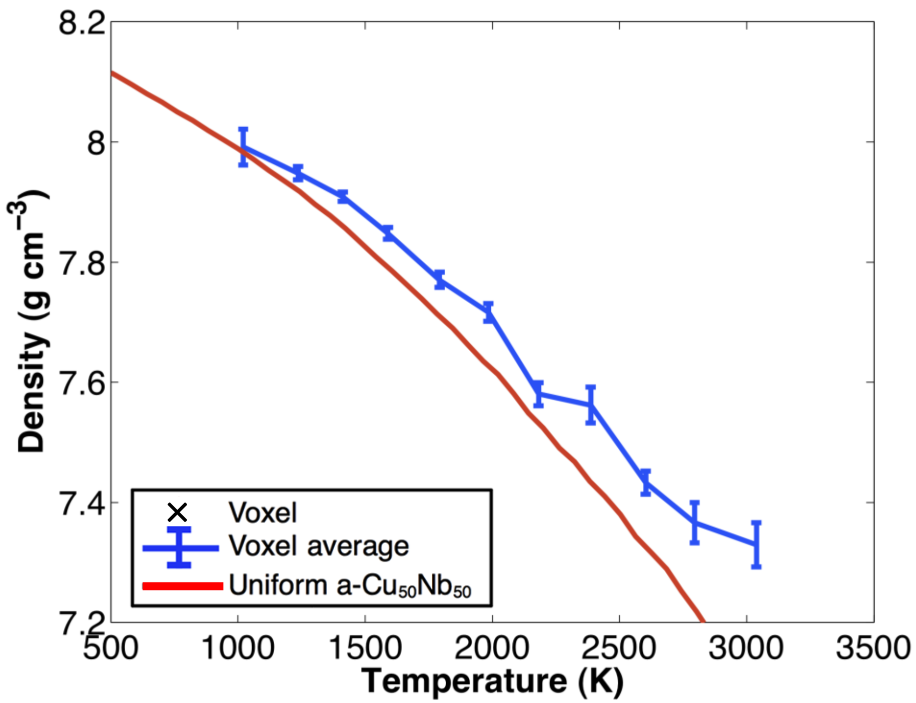


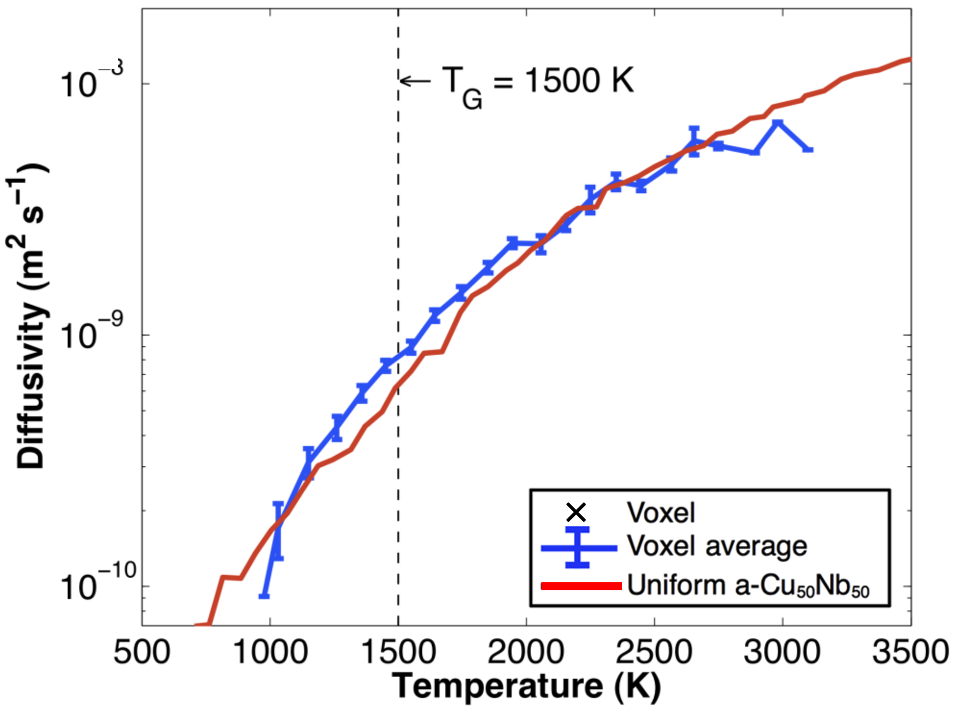
Thermal spike properties



Density and diffusivity within thermal spikes

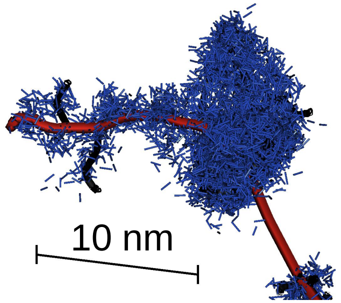
Properties of voxels with T_{max} > 1500 K at a single time, t = 5 ps

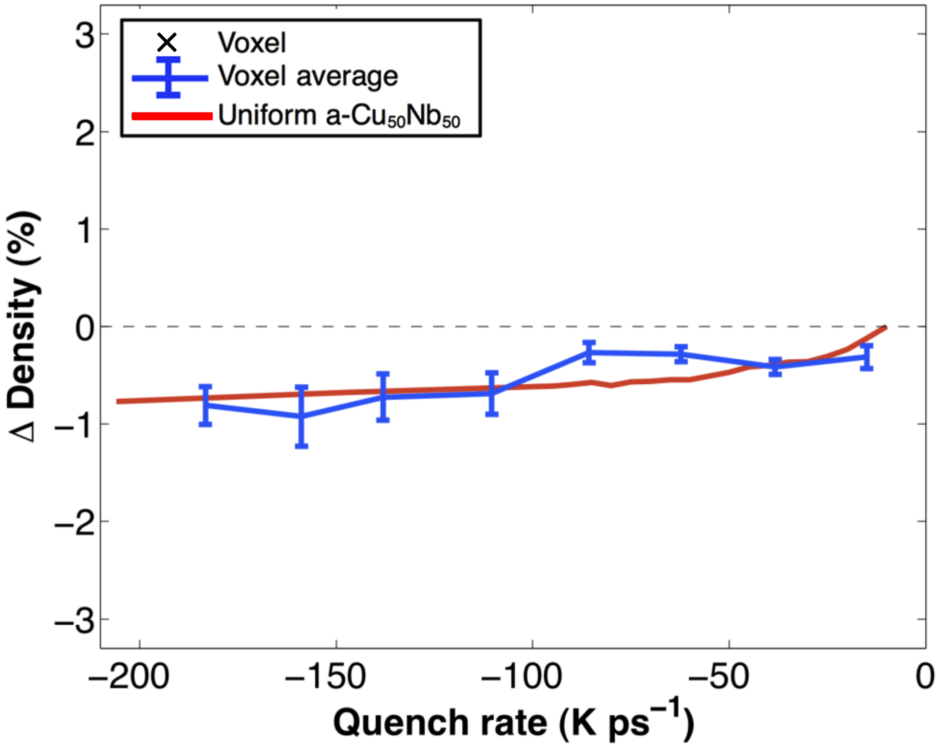


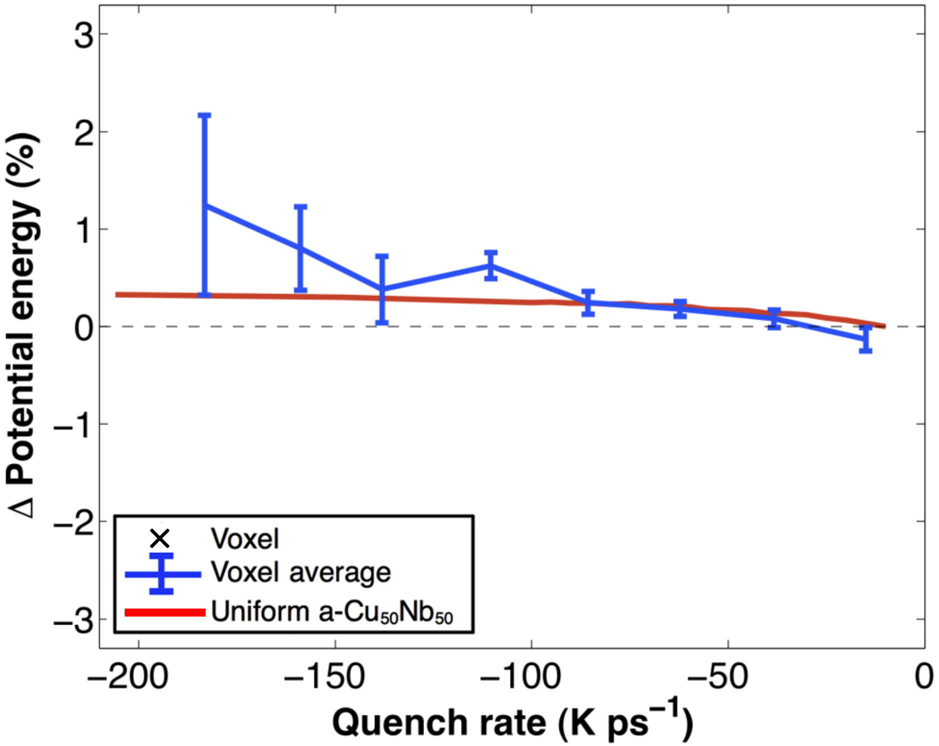


Rapid quenching locks in free volume, excess energy

Properties of liquid voxels are determined by quench rate



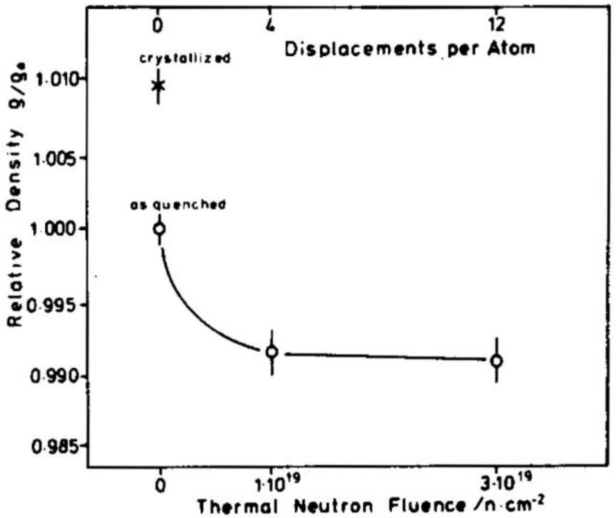


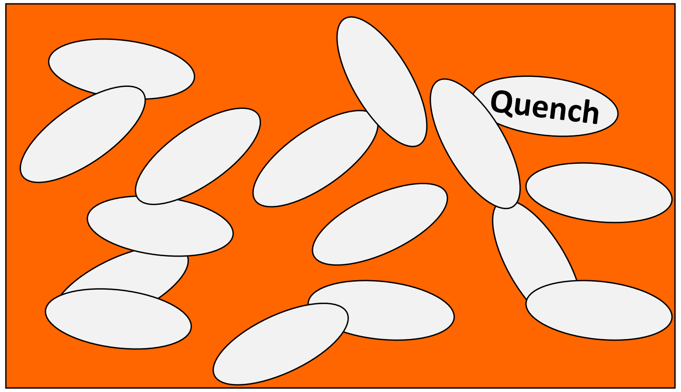


Radiation swelling of metallic glasses

Irradiated a-$Fe_{40}Ni_{40}B_{20}$

Gerling, Scripta Metall. **16**, 963 (1982)

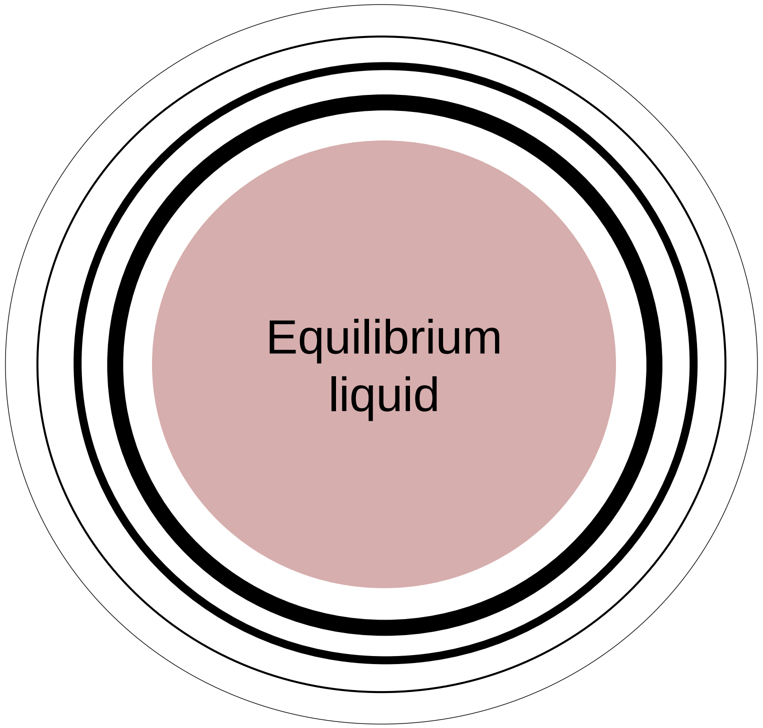




	Density [g cm^{-3}]	Yield stress [GPa]	Yield strain [%]	Young's Modulus [GPa]
Matrix QR=1×10^{13} K/s	8.199	2.38	0.0253	101.8
Quenched thermal spikes <QR>=6.5×10^{13} K/s	8.154 (~0.5%↓)	3.02	0.0361	88.6

Similar reasoning may explain radiation-induced ductilization

Shock waves emitted by thermal spikes



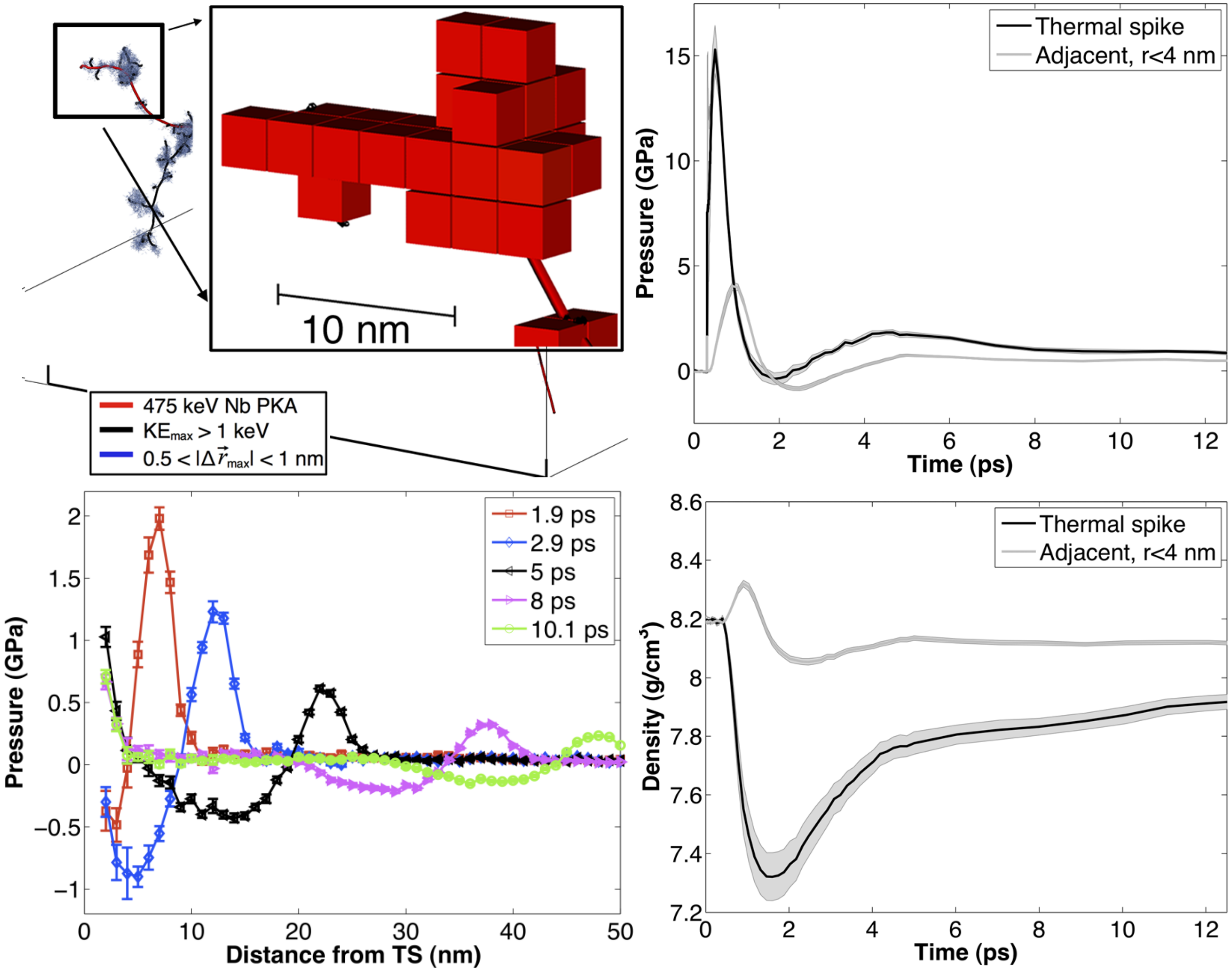
Elastic pulse

Time scale:
0.1-10ps

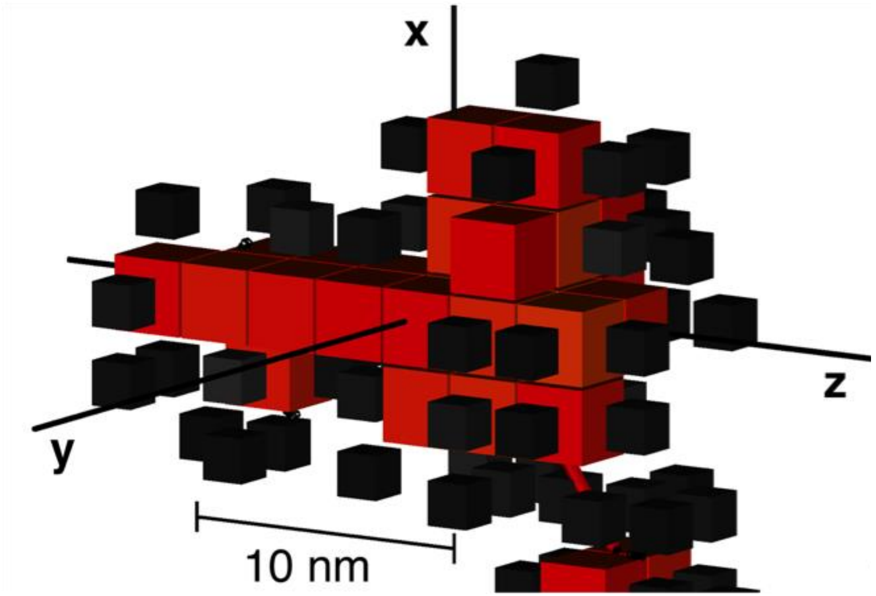
Length scale:
~10nm

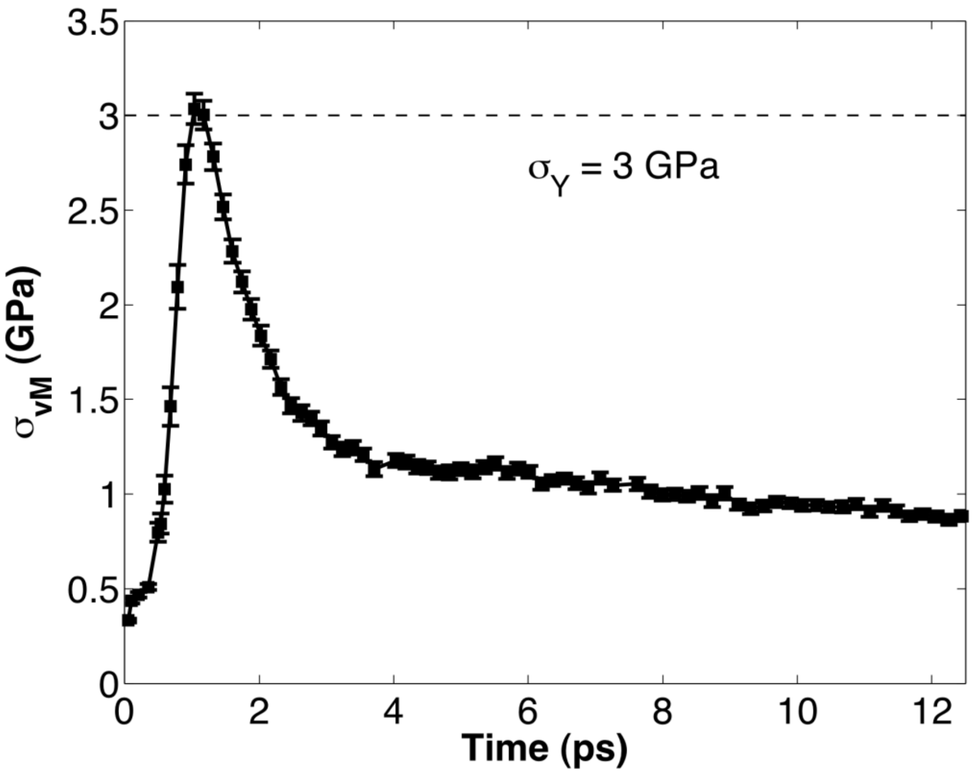
The high pressure liquid in a thermal spike acts as an inclusion with a transient misfit => emits an elastic pulse

Pressure excursions in thermal spikes



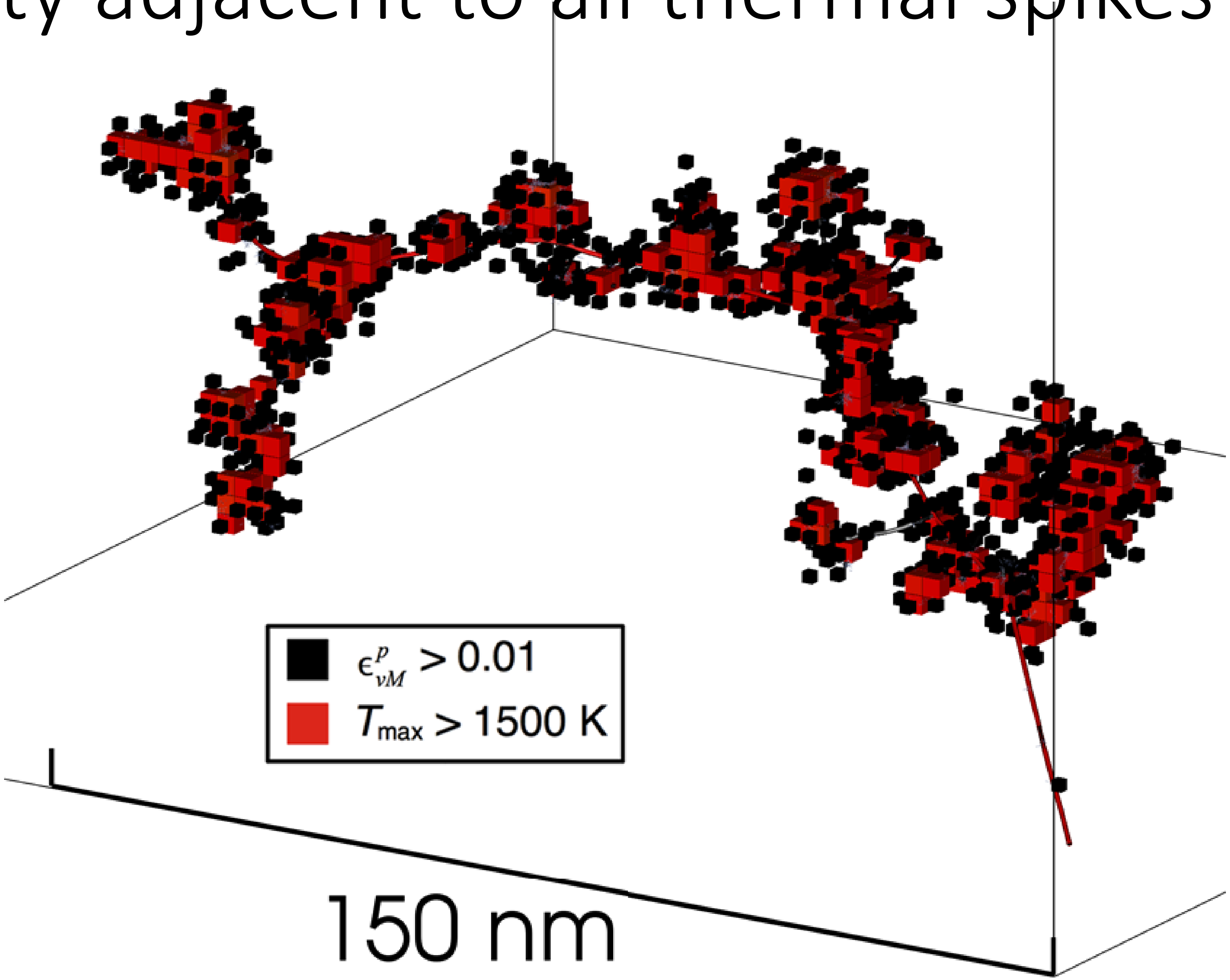
Plasticity adjacent to thermal spikes



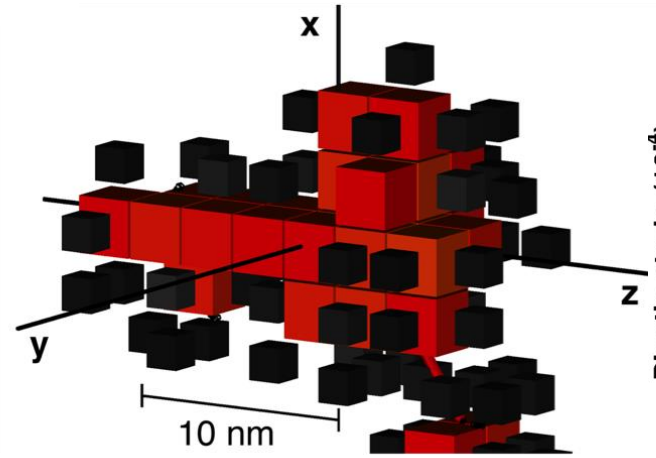


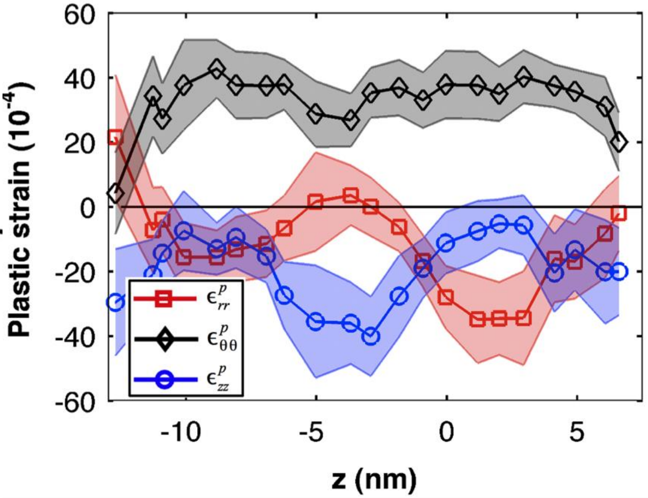
Stress pulse exceeds material yield stress adjacent to thermal spike, leading to plastic flow

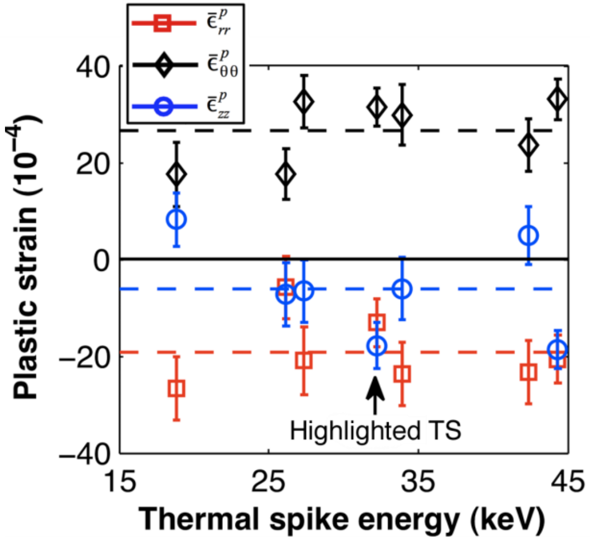
Plasticity adjacent to all thermal spikes

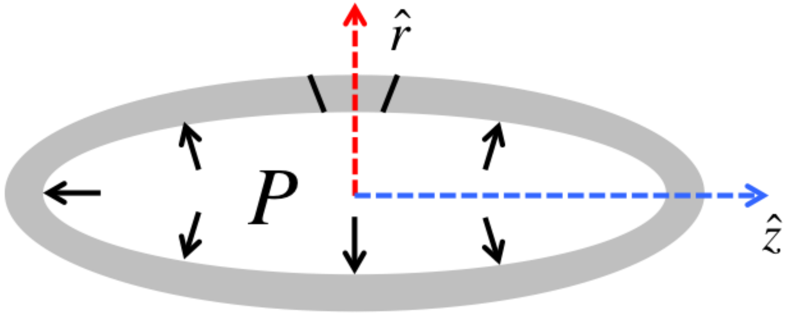


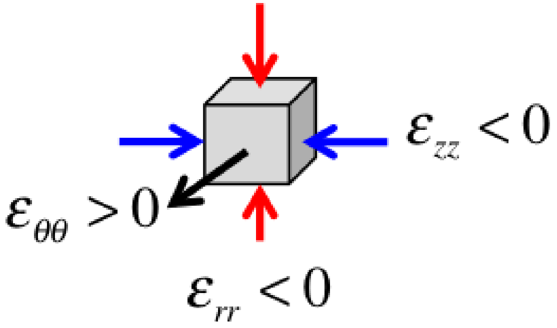
Thermal spike plasticity is polarized





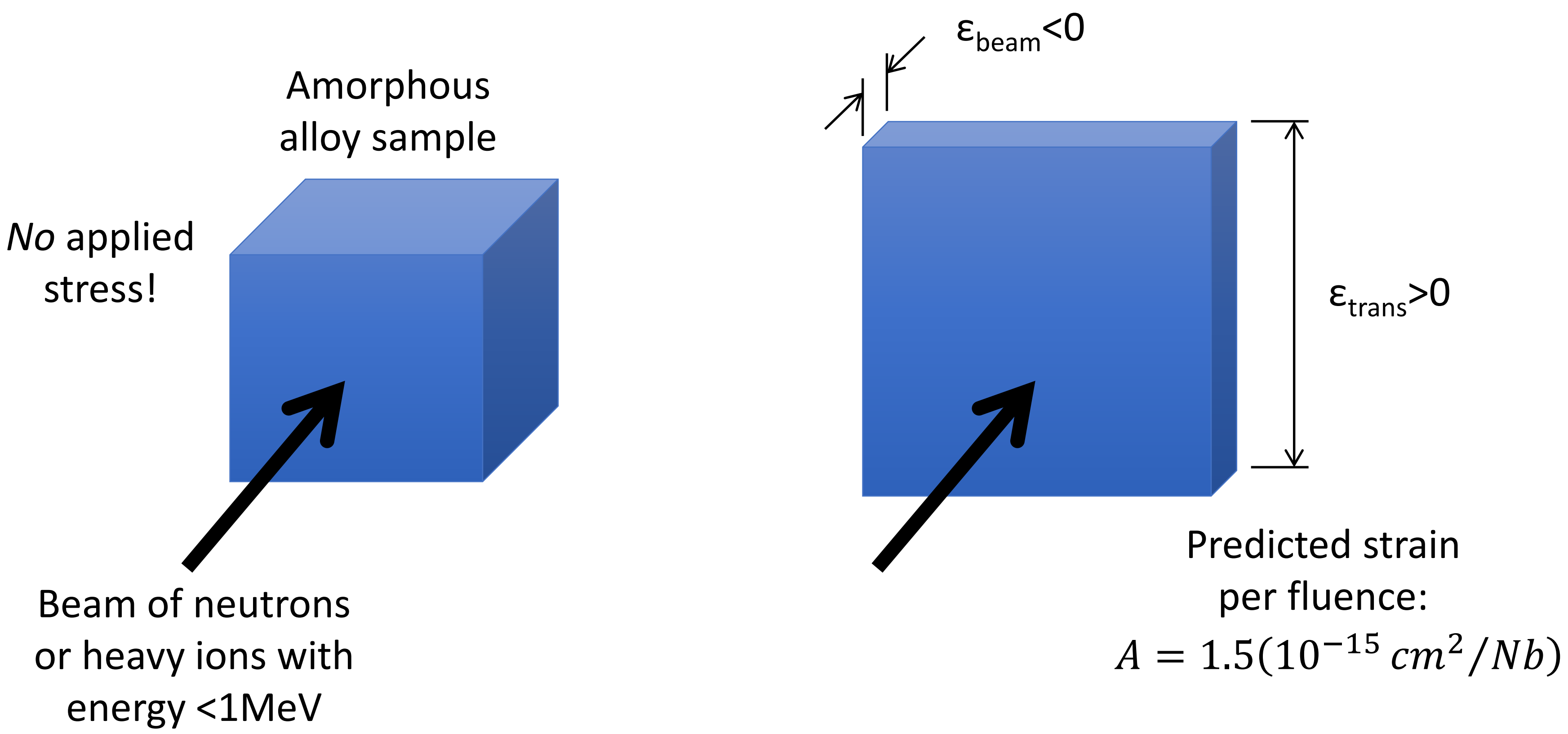






Consistent with a pressurized ellipsoidal inclusion

Prediction: a directed particle beam causes metallic glasses to deform plastically



This prediction still awaits experimental validation

Summary

- Thermal spikes play a major role in radiation response of solids
  - Their shape (compactness) affects defect formation
  - Liquid phase interdiffusion in thermal spikes limits the thermal stability of nanocomposite metals
  - High-rate quenching of thermal spikes reduces the density of amorphous metals
  - In amorphous metals, stress pulses emitted from thermal spikes cause anisotropic plastic deformation in the surrounding solid material

- Much is known about thermal spikes, yet much remains to be discovered

- We are beginning to use our understanding of thermal spikes to engineer materials for radiation resistance

CREDDS

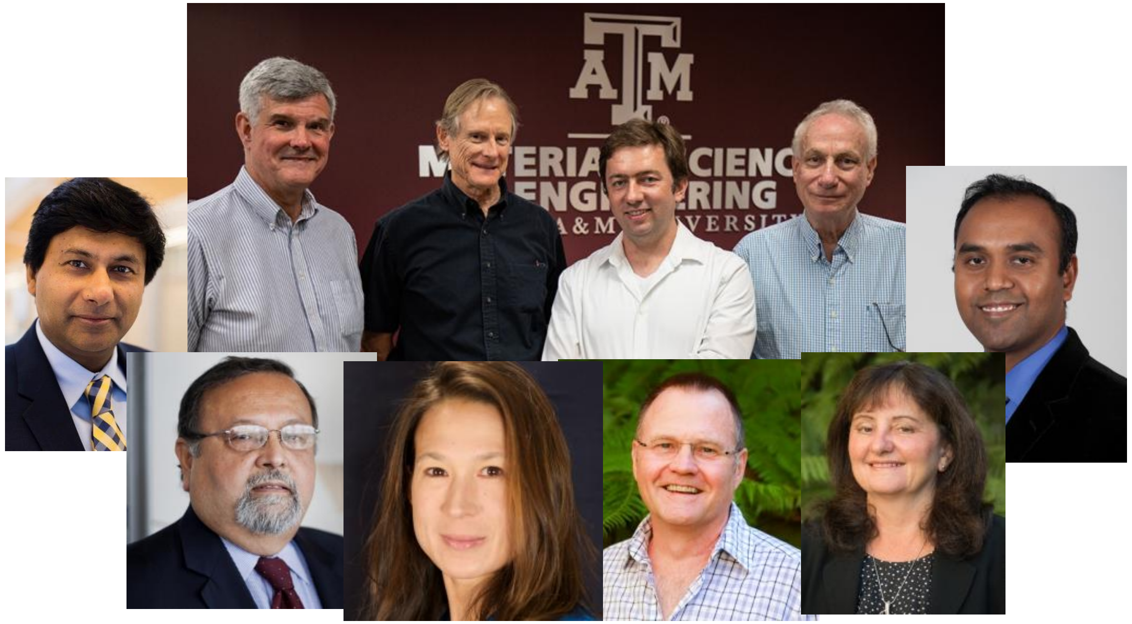
Center for Research Excellence on Dynamically Deformed Solids

NNSA/SSAA center of excellence, Est. June 2018

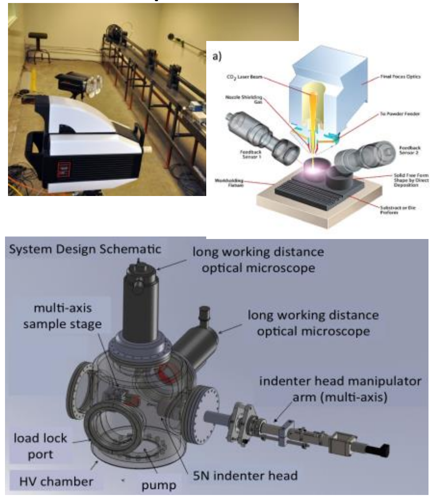
credds.tamu.edu

- Lead: Texas A&M University, director: M. J. Demkowicz (MSEN)
- Collaborating institutions: UCSB, U. Michigan, U. Connecticut
- Goals:
    - ***Discover, understand, and predict the influence of microstructural heterogeneities—such as interfaces, inclusions, and porosity—on the high strain rate ($>10^4$/s) mechanical response of additively manufactured, multiphase materials***
    - ***Train the next generation of leaders in stockpile stewardship through close collaboration with partners at NNSA labs***
- Advisory committee with members from NNSA labs and academia
    - From LLNL: Mukul Kumar

Visionary leadership team



Developing new capabilities



Training the next generation of leaders