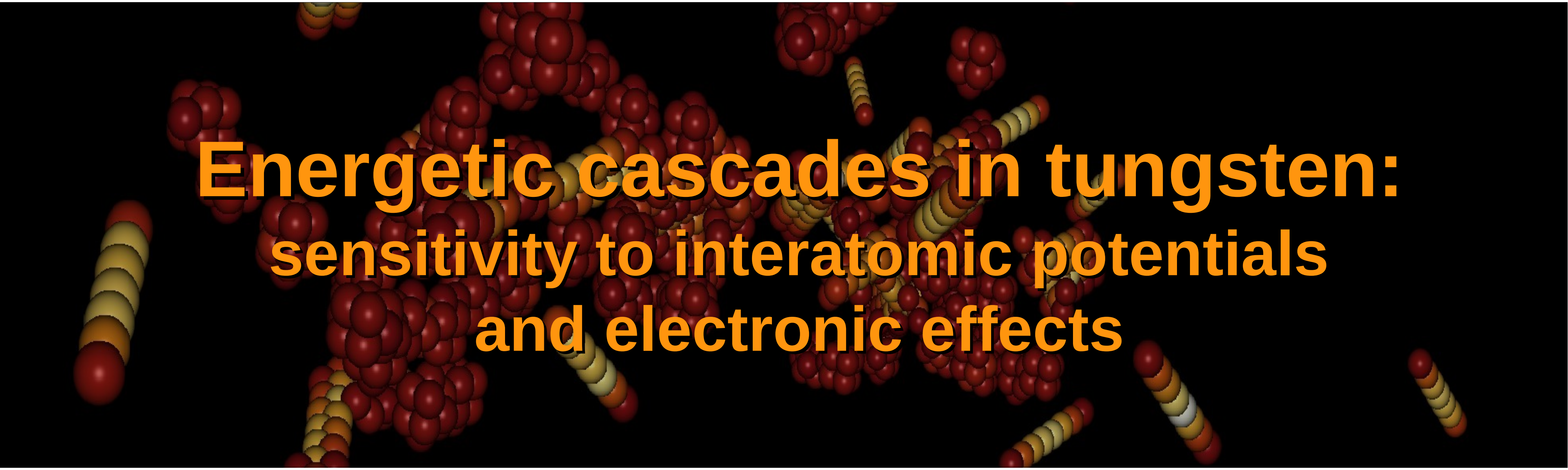

Energetic cascades in tungsten: sensitivity to interatomic potentials and electronic effects

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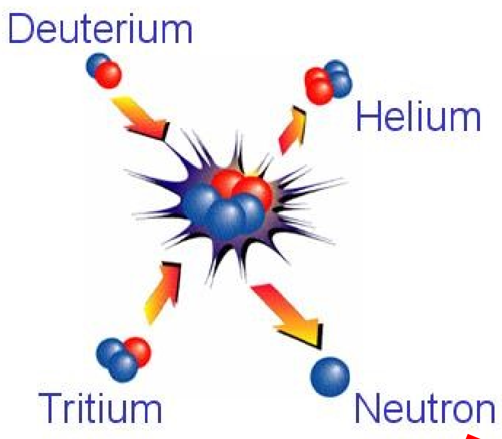
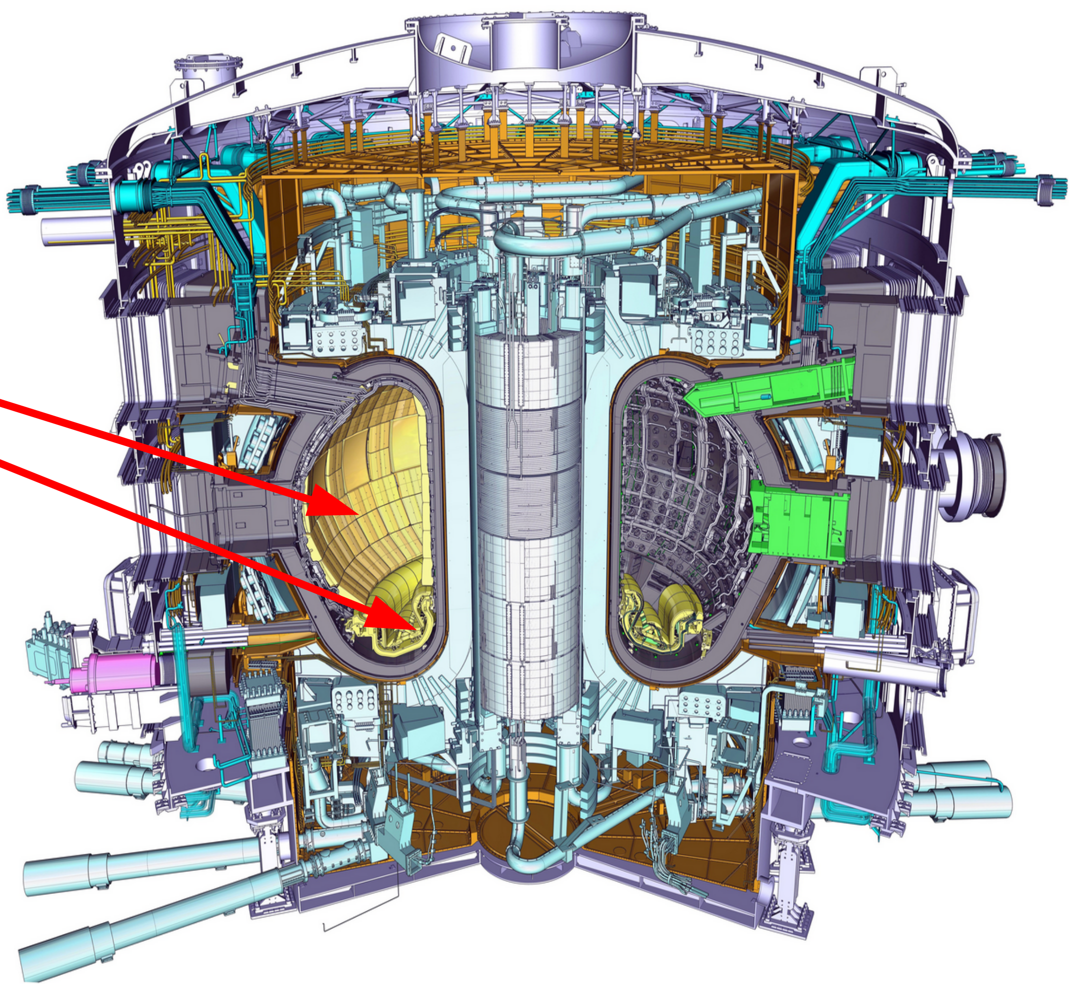
Motivation: Fusion reactors

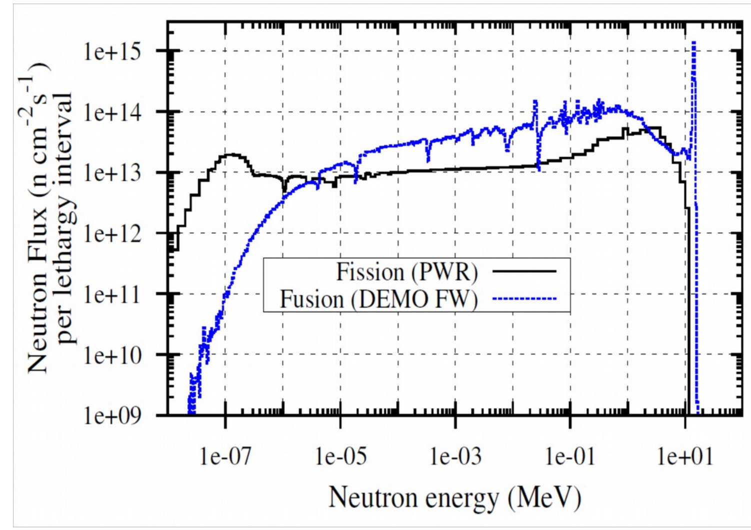




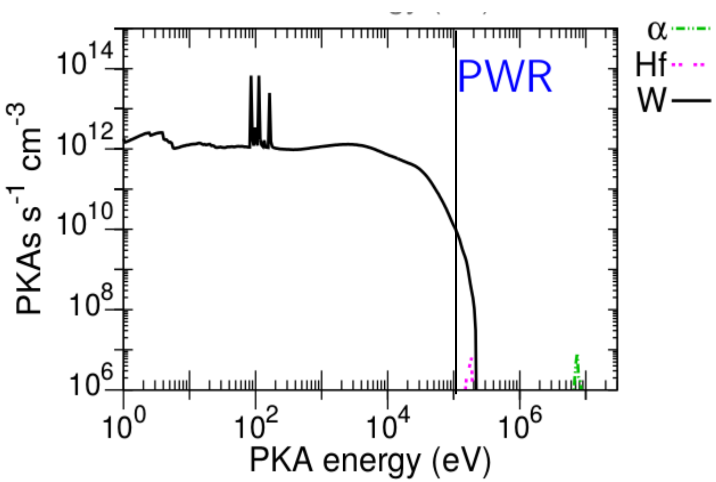
- Materials in future fusion reactors must withstand high levels of neutron irradiation
  - 14 MeV neutrons will produce energetic recoils
- Current experimental facilities *cannot* reproduce conditions in future fusion reactors
  - Modelling offers the only alternative for predicting materials response

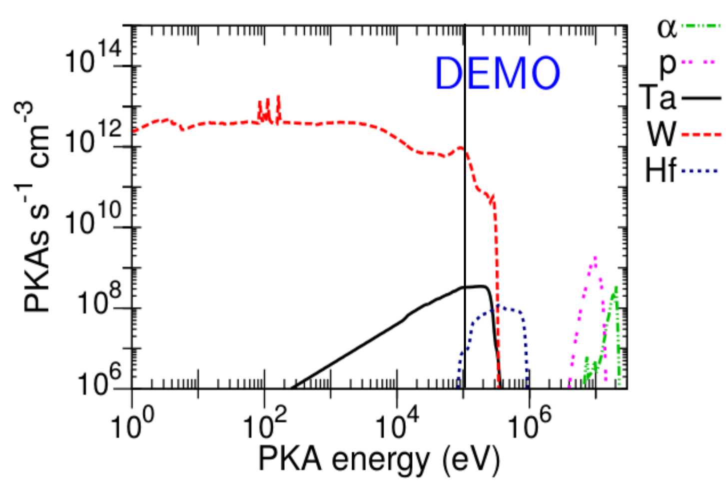
Fusion vs. fission neutrons

- Fusion neutron spectrum peaks at 14 MeV
- Energetic W recoils from DEMO neutron spectrum orders of magnitude more frequent than from PWR spectrum
  - Recoil spectrum in W reaches 300 keV
  - 2 orders of magnitude higher frequency of 100 keV recoils in DEMO
- Energetic PKAs also important in ion irradiation experiments used as proxy to study neutron irradiation effects



[M. R. Gilbert et al, Nucl.Fusion 52 (2012) 083019]

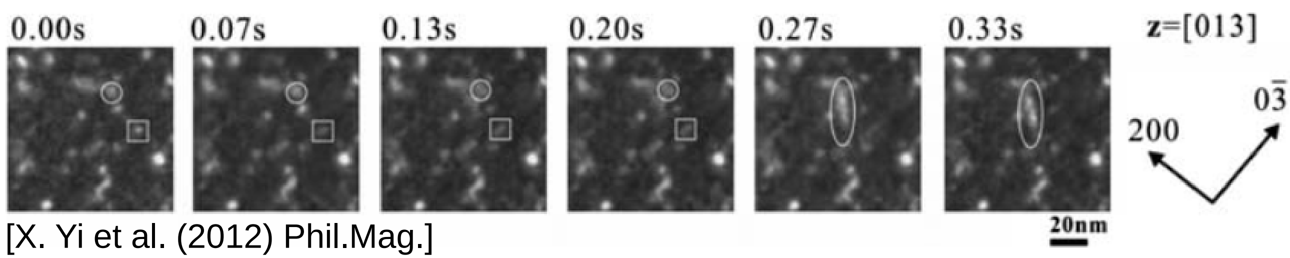




[M. R. Gilbert et al., Tech. Rep. CCFE-R(16)36]

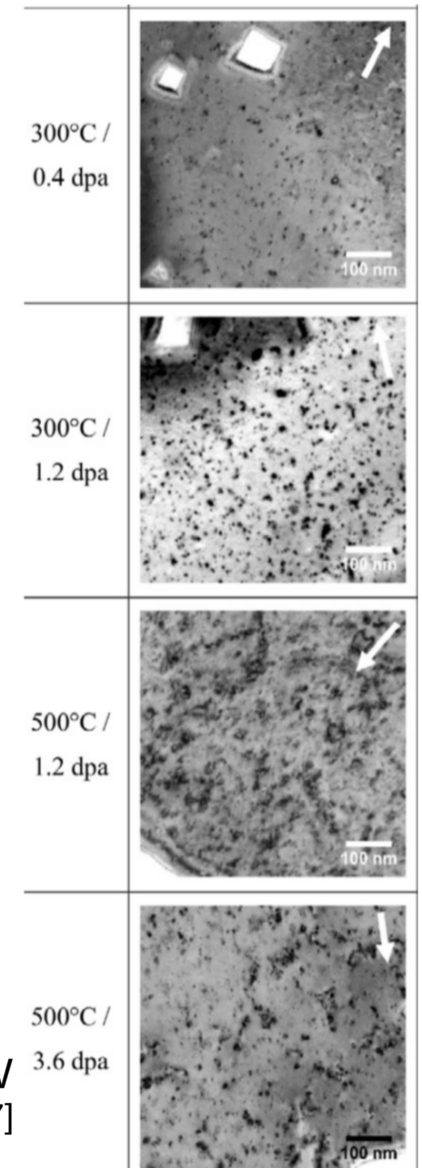
Motivation: Spatial ordering of defects

- Experimental evidence in irradiated material of spatial ordering of nano-dislocation loops
- Experimental observation of coordinated motion of loops



[X. Yi et al. (2012) Phil.Mag.]

- Explanation: elastic forces between dislocation loops very strong at short range
  - Enough to pin loops together
- To understand and predict the evolution we need to know the initial damage state
  - *Energy of interaction depends on size and separation*

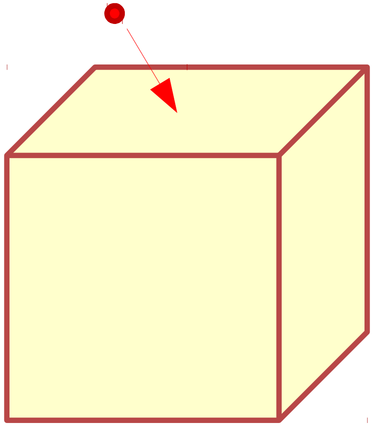


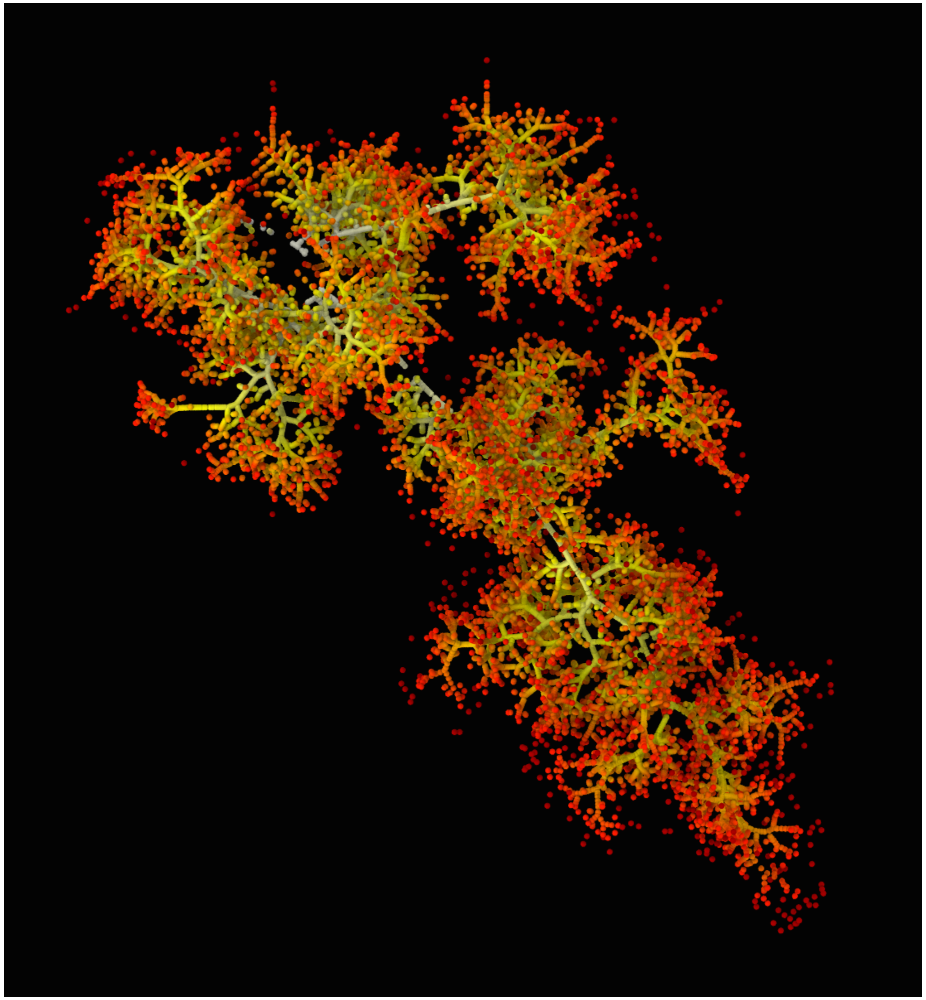
99.996 wt.% W

[Yi, X., *et al*., Acta Materialia **92** (2015) 163-177]

Method: Cascade simulations

- PARCAS classical MD code
  - Open boundaries in one direction for foil
  - Periodic boundaries in other directions
  - Thermostat (Berendsen's to 0 K) at periodic boundaries
  - Electronic stopping on atoms with Ekin > 10 eV
  - Dynamically varying time step





Energetic ions during ballistic phase in 200 keV collision cascade in W (colored according to time)

HELSINGIN YLIOPISTO
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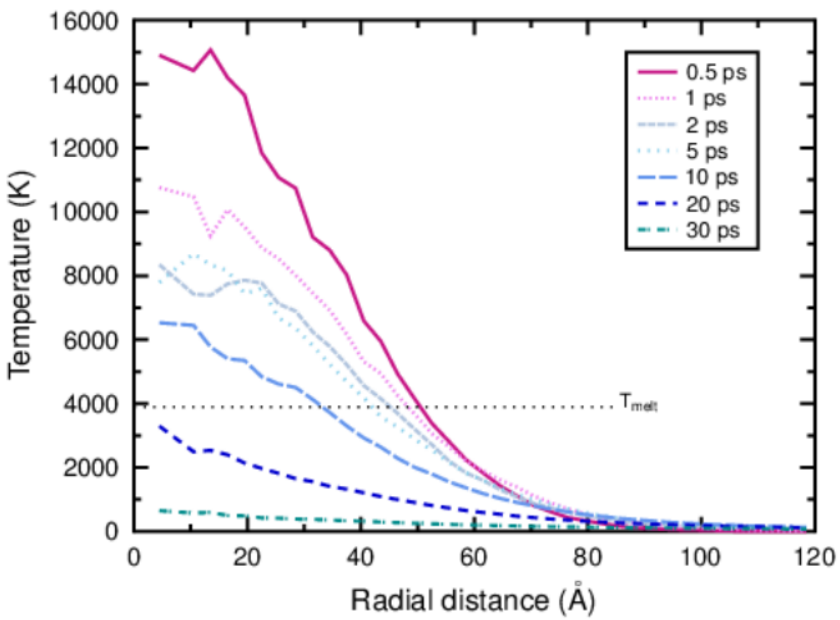
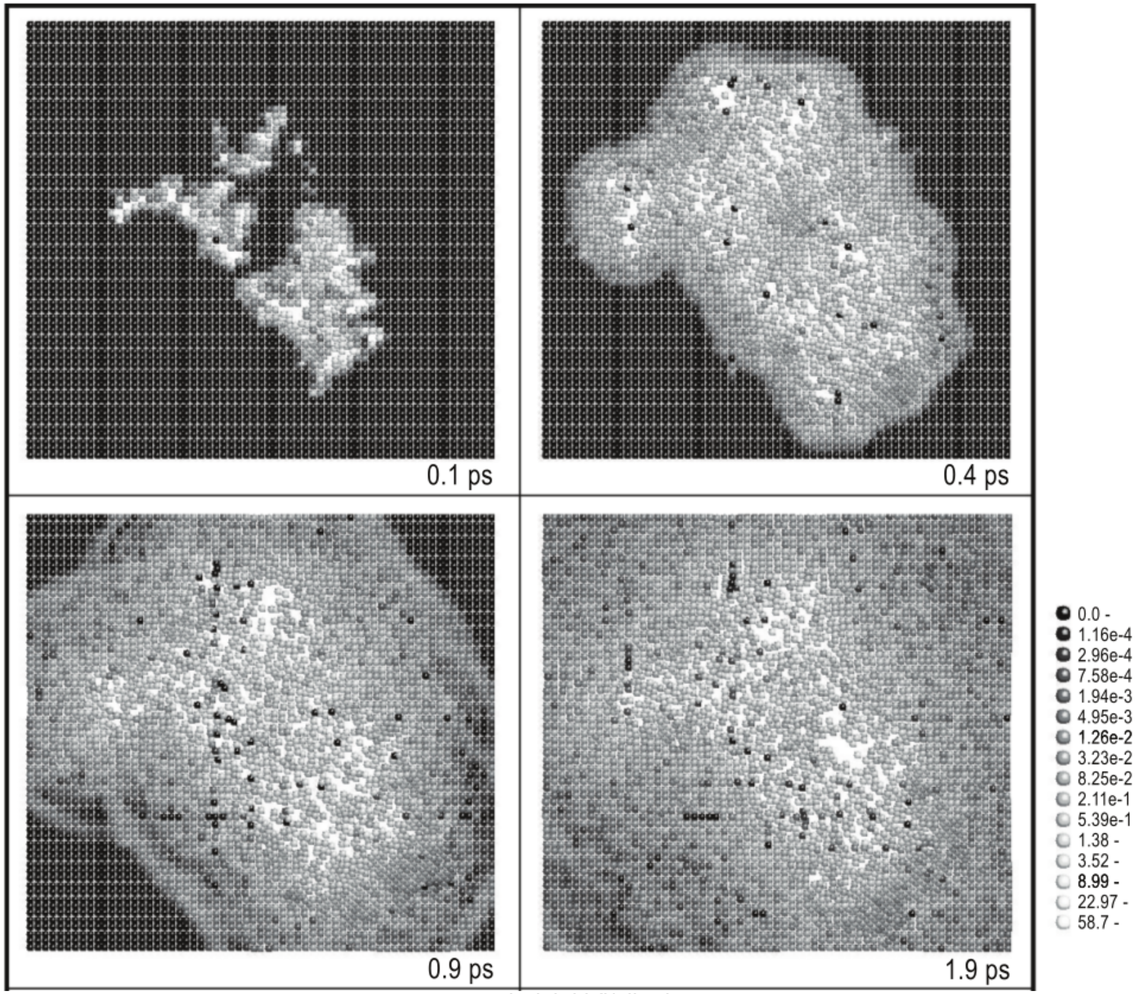
Department of Physics
Andrea Sand

Heat spikes in W

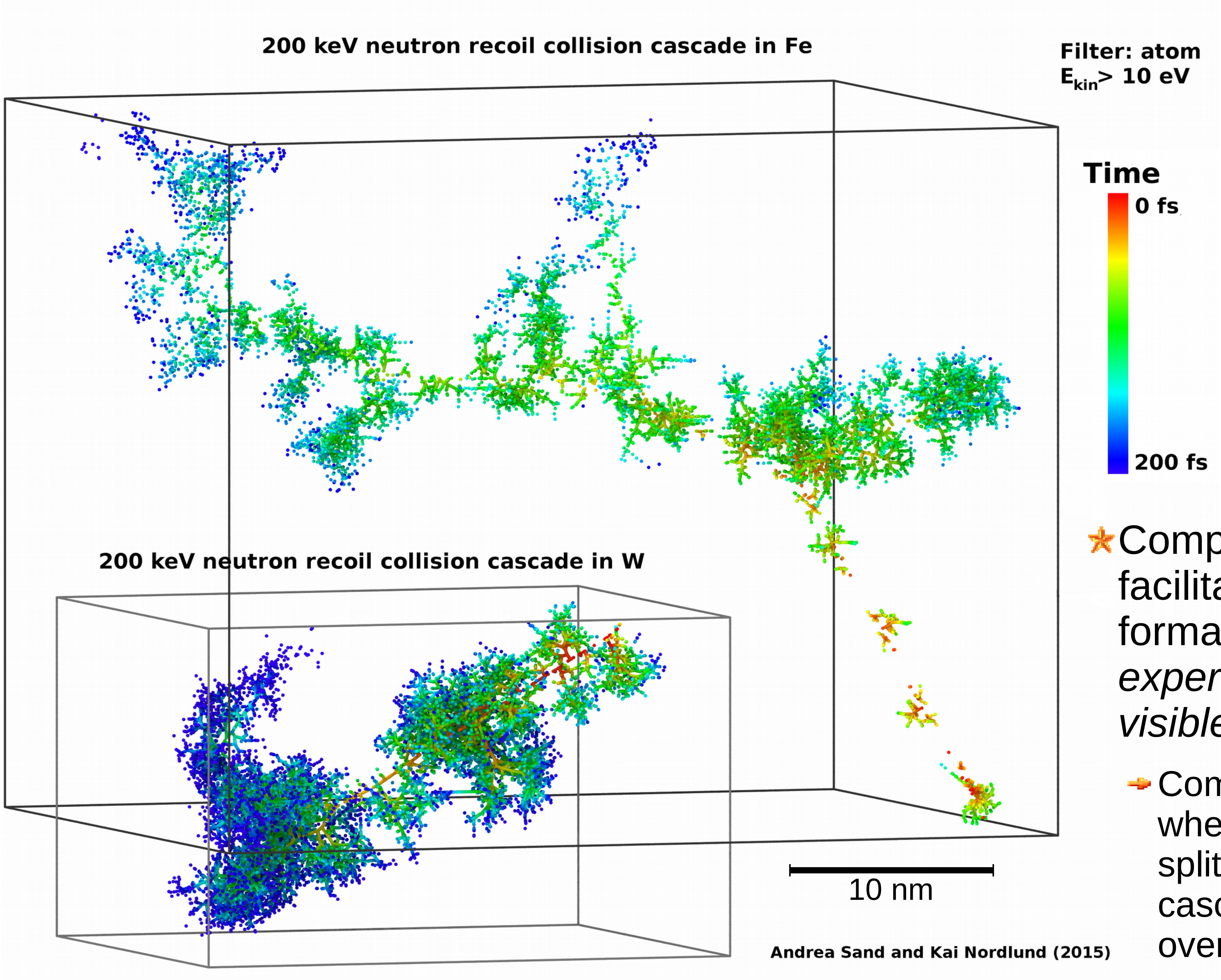
- In W, cascade splitting starts at ~ 150 keV
- Heat spike survives for ~ 10 ps
  - High effective core temperature



150 keV cascade in W, unit cell thick slice



Collision cascades: W vs. Fe

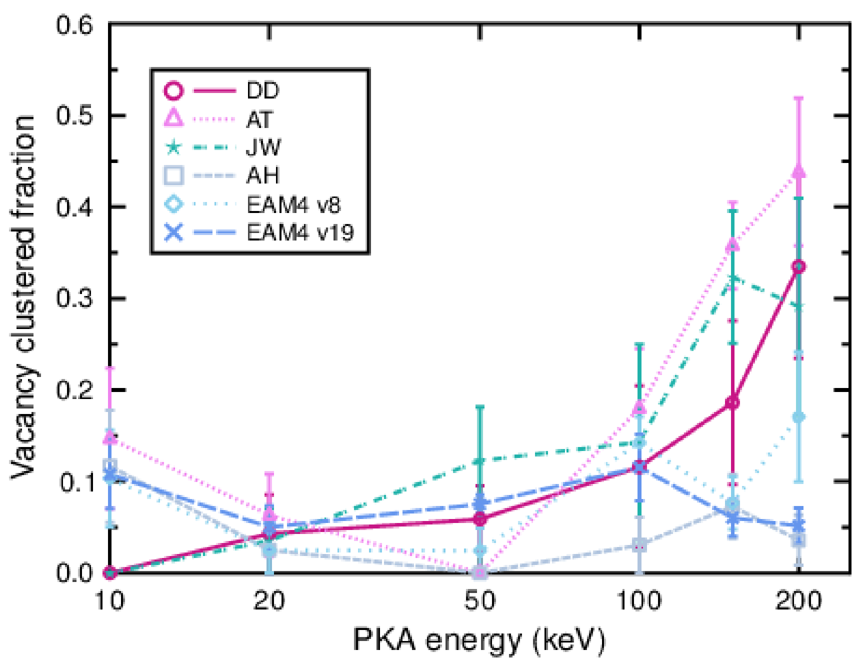


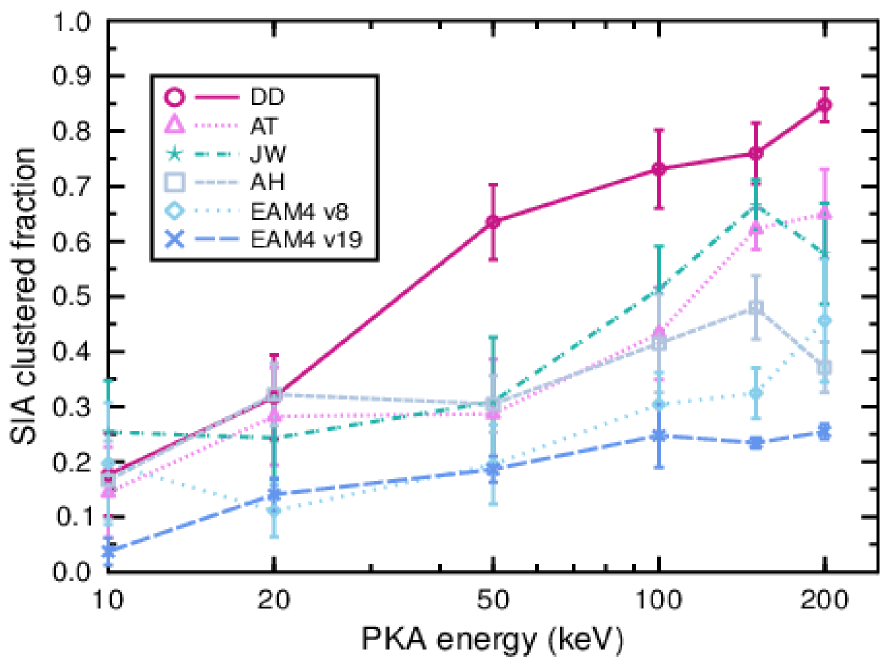
- Compact cascades facilitate in-cascade formation of large, *experimentally visible* defects in W
  - Compare to Fe, where subcascade splitting disperses cascade energy over a wider area

Defect production

- dependence on interatomic potential

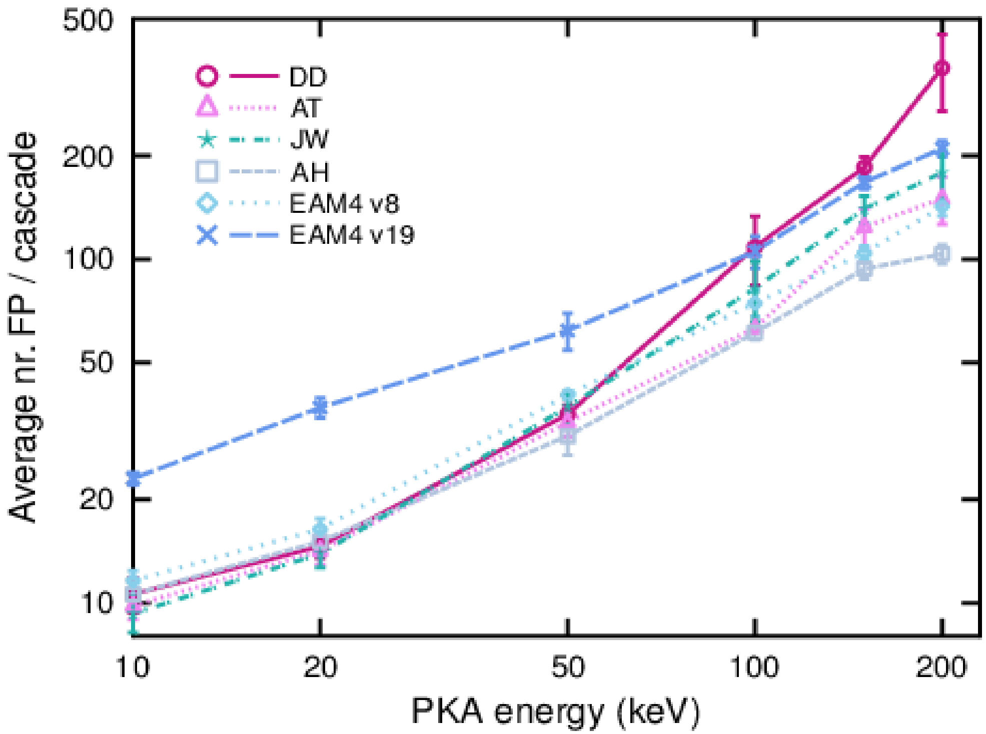
- Potentials with largely *similar point defect formation and migration energies* disagree regarding clustered fraction of defects for high PKA energies
- Some potentials predict only very small clusters, others show formation of clusters of > 100 point defects





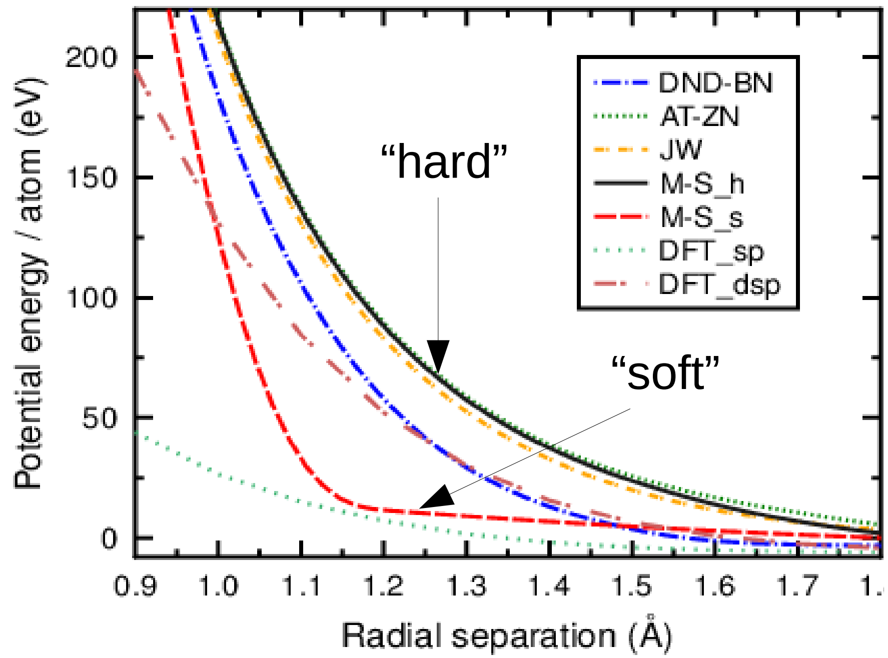
Defect numbers

- Defect clusters inhibit recombination, so with larger clusters more defects survive
- → For energetic PKAs, predictions of defect numbers diverge
- *Why the different predictions, despite extensively fitted "good" potentials??*



Stiffening of potentials

- Energetic impacts involve close interactions
  - "Equilibrium" potentials not constructed to handle this
- Close interactions well described by, e.g., the universal Ziegler-Biersack-Littmark (ZBL) potential
- The two potentials must be smoothly joined
  - "smooth" can be done in many ways!
  - Affects threshold displacement energy (TDE)



- Picture shows the intermediate range of a number of potentials for W dimer
  - Including a recent potential (equilibrium part by Marinica *et al.),* purposely stiffened in two ways, identical in short and long range, both with reasonable TDE

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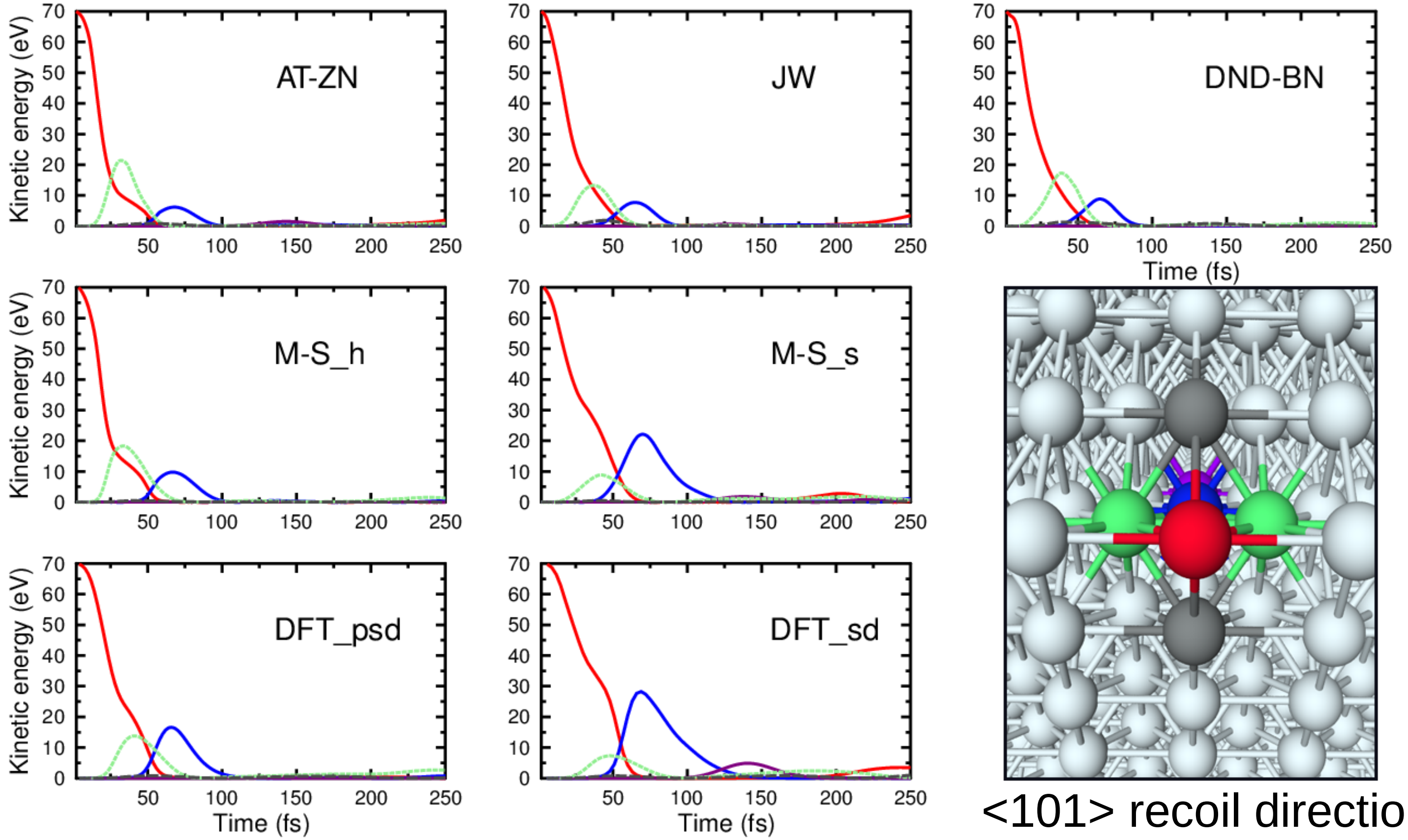
Impact of intermediate range
- energy transfer

- Harder potential → more energy transfered to neighboring atoms; less to head-on atom
  - In general, DFT results including semi-core electrons agree with harder potentials



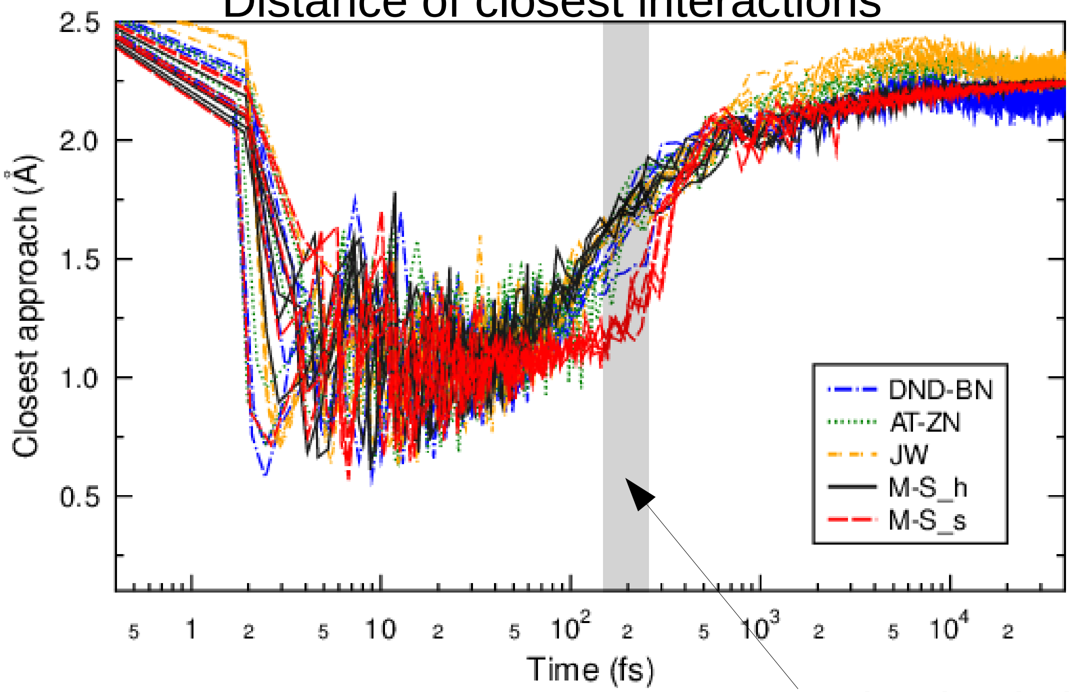
<101> recoil direction

[A. E. Sand et al., JNM **470** (2016)]

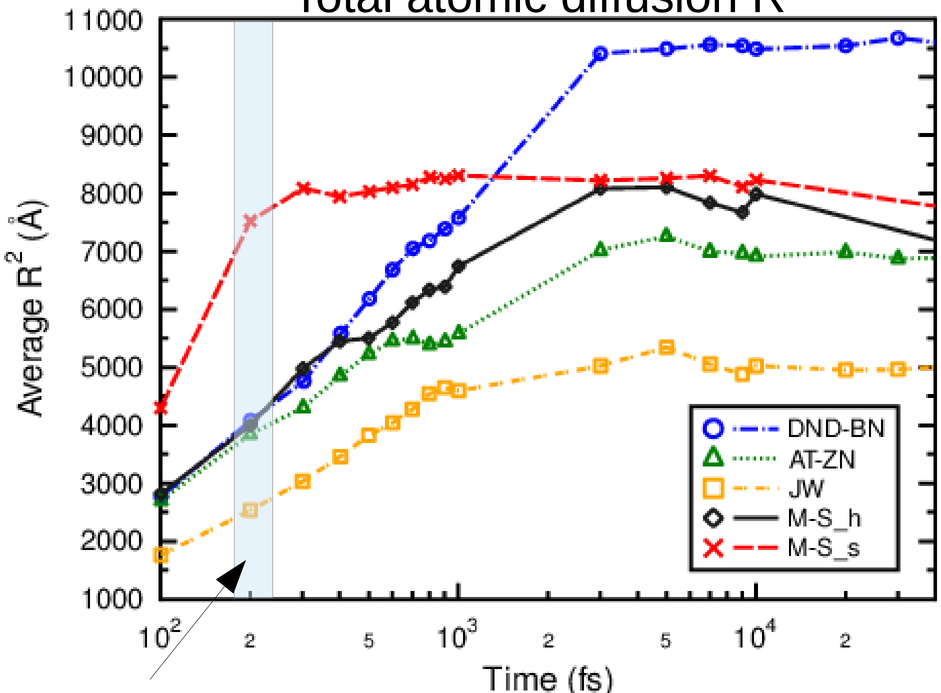
Impact of intermediate range
- full cascades

- Less heat spike diffusion with soft potential (less energy transfer to neighbors)
  - Transition from ballistic phase to thermal phase at ~ 200 fs
  - Heat spike diffusion comes from atoms moving *after* ballistic phase
  - No such diffusion (no heat spike) with the softest potential (M-S_s) in 10 keV cascades!

Distance of closest interactions



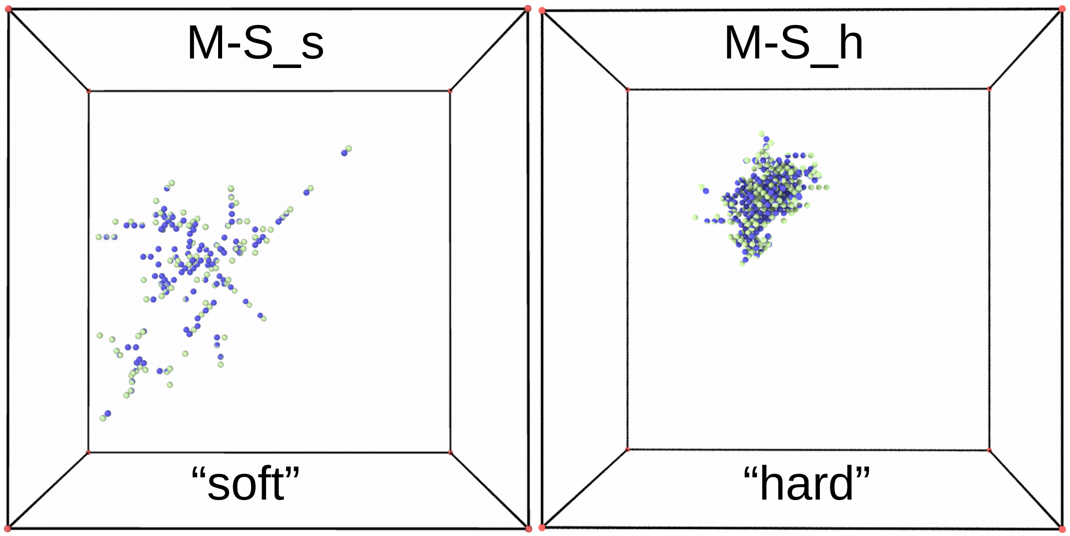
Total atomic diffusion R^2



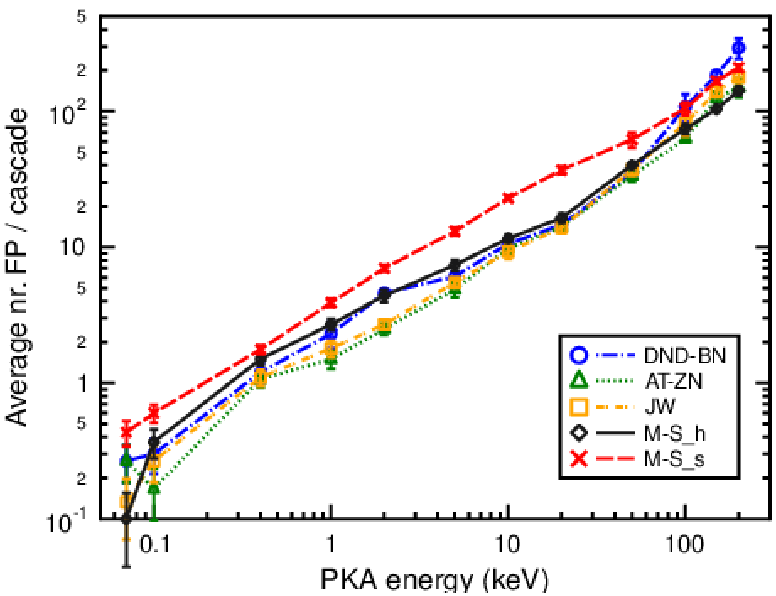
end of ballistic phase

Impact of intermediate range
- final defects

- Heat spike diffusion facilitates recombination
- Also, with softer potential individual recoils travel further (lose less energy to surroundings)
- → more (too many) final defects with softer potential in mid energy range
- ***But...** doesn't explain deviation for higher PKA energies!*

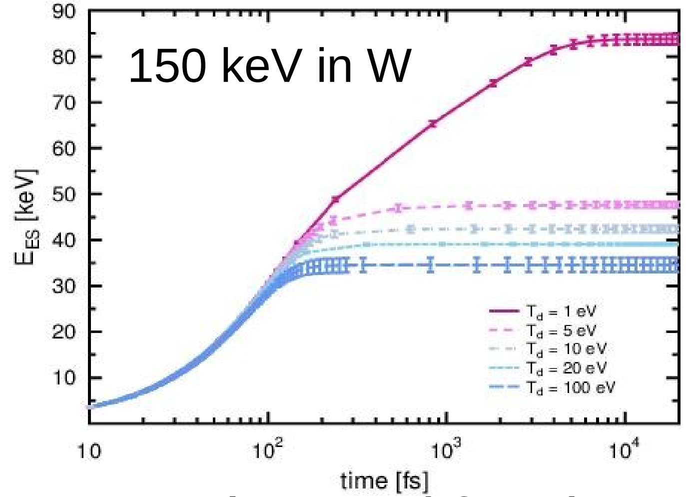


10 keV cascades, displaced atoms

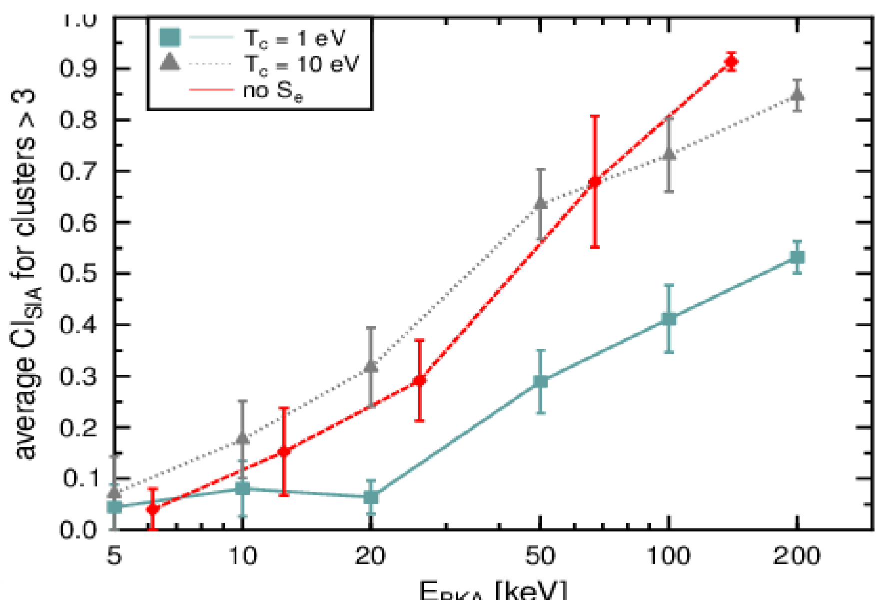


Electronic stopping

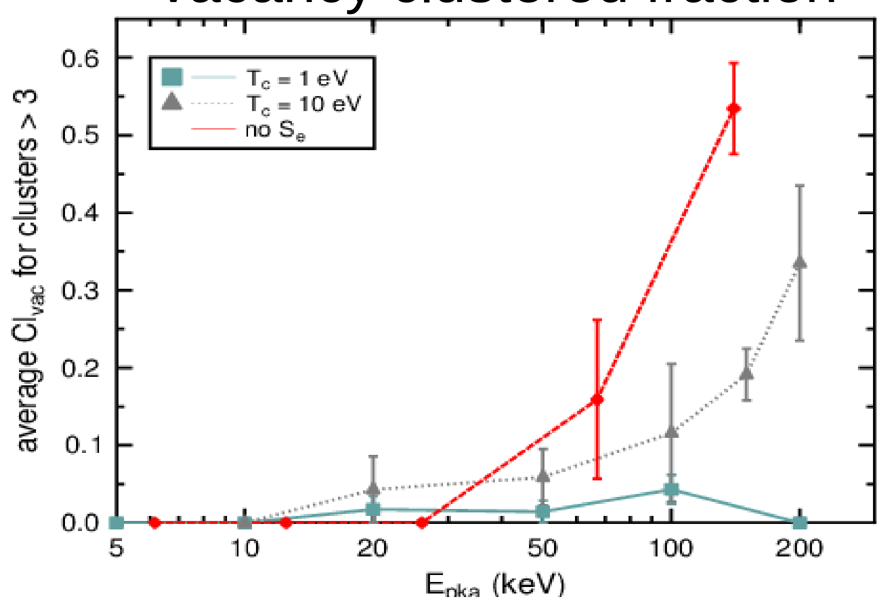
- Electronic stopping applied as friction term above certain K.E. cut-off
- Different clustering behaviour without stopping
- A low cut-off energy (1eV) results in large energy losses from thermal phase of cascade
  - Electronic stopping theories do not necessarily apply in heat spike
- Large effect on final defects!



SIA clustered fraction

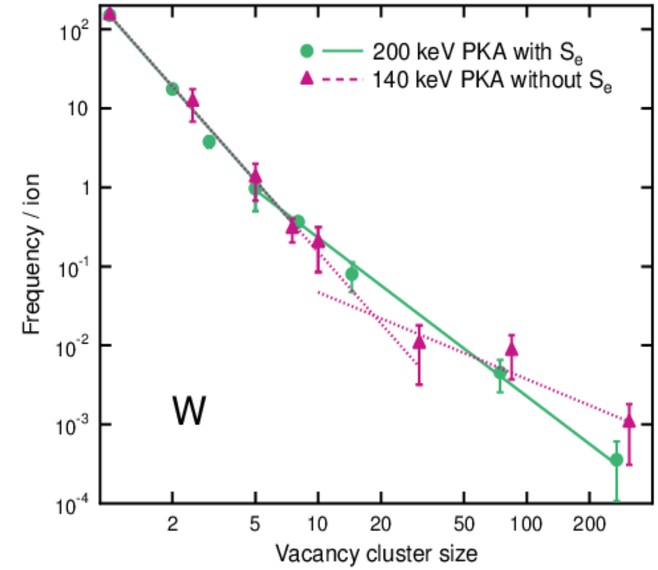


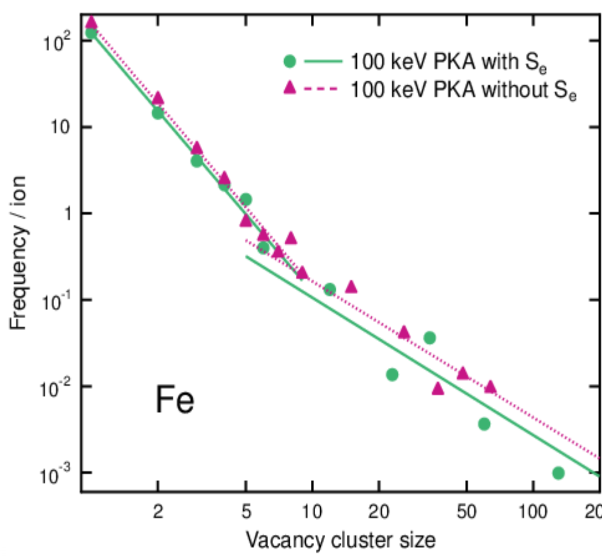
Vacancy clustered fraction

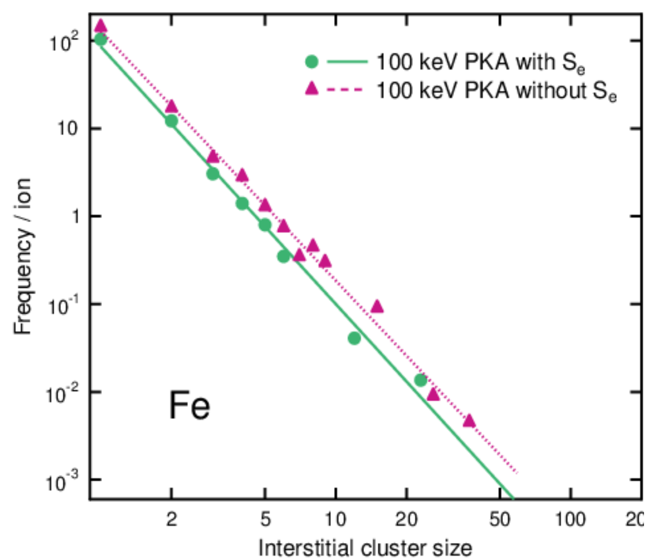


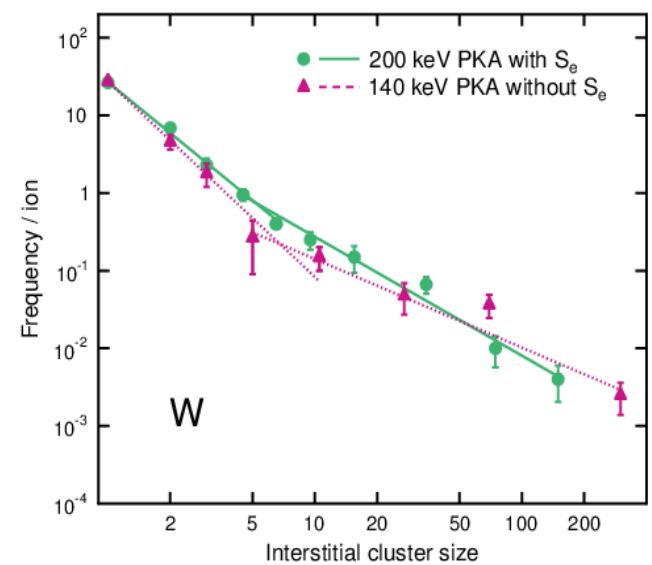
Electronic stopping in Fe vs. W

- Electronic stopping affects results in W, but not so much in Fe
  - Fe cascades compared here have different damage energy → difference in total numbers of defects but same slope in size distribution
  - W cascades have same damage energy (140 keV) → should produce same damage



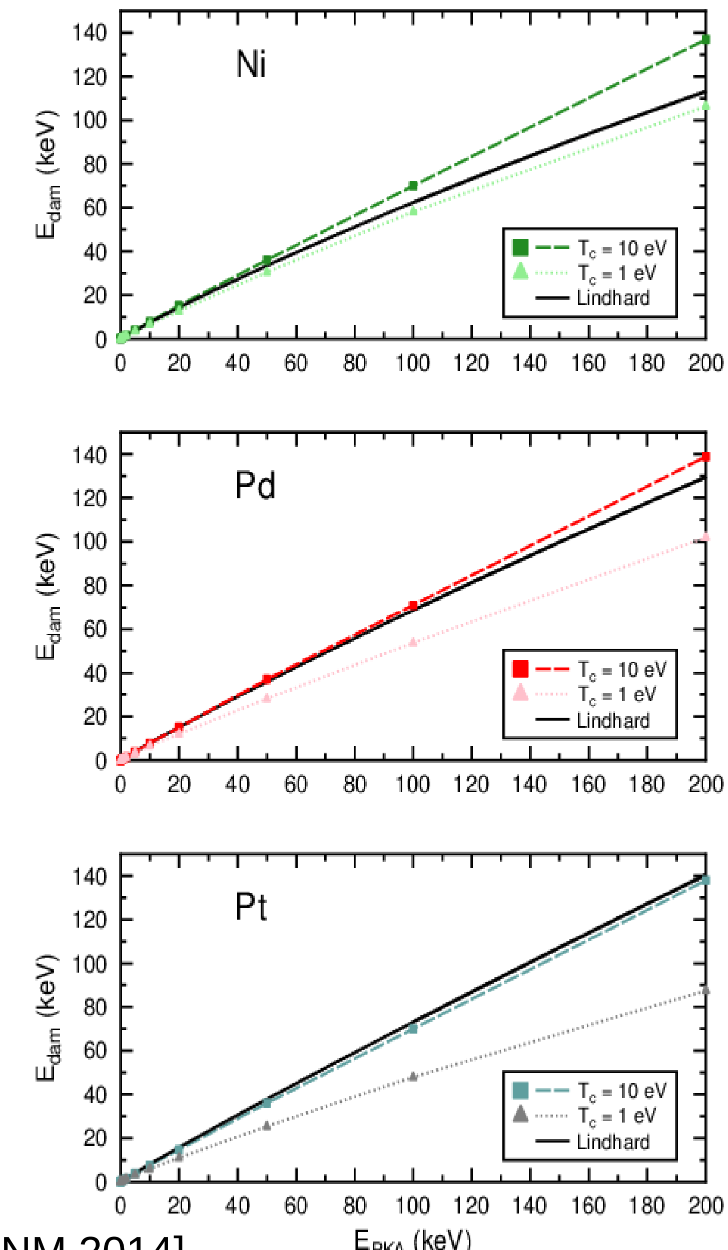






Comparison to experiment

- Choice of cut-off energy arbitrary (no motivation from theory)
- Determines rate of cooling of heat spike
- Cooling rate of heat spike affects level of atomic mixing in cascades
  - Mixing is dominated by the relatively rare energetic cascades
  - Directly comparable to experimental ion beam mixing



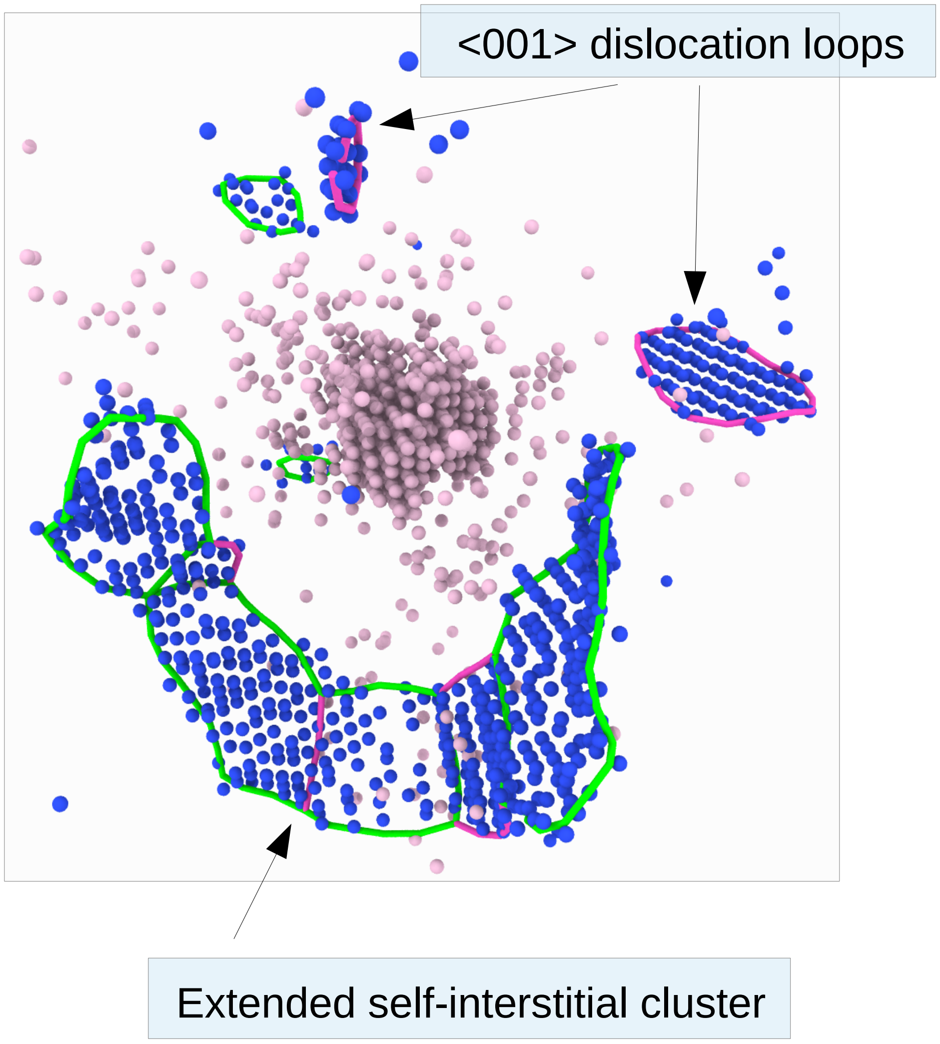
Material	Beam	Q_{sim} (Å^5/eV) $T_c = 1$ eV	Q_{sim} (Å^5/eV) $T_c = 10$ eV	Q_{exp} (Å^5/eV)
Ni	600 keV Kr	2.9 ± 0.1	4.7 ± 0.1	4.8 ± 0.5^a
Ni	650 keV Kr	3.1 ± 0.2	5.1 ± 0.1	5.0 ± 0.7^b
Pd	600 keV Kr	6.2 ± 0.2	14 ± 1	8.4 ± 0.8^a
Pd	400 keV Kr	6.1 ± 0.2	13.7 ± 0.5	9 ± 1^c
Pt	1 MeV Kr	6.1 ± 0.3	20 ± 3	14 ± 2^b

[A.E. Sand and K. Nordlund, JNM 2014]

Damage morphology

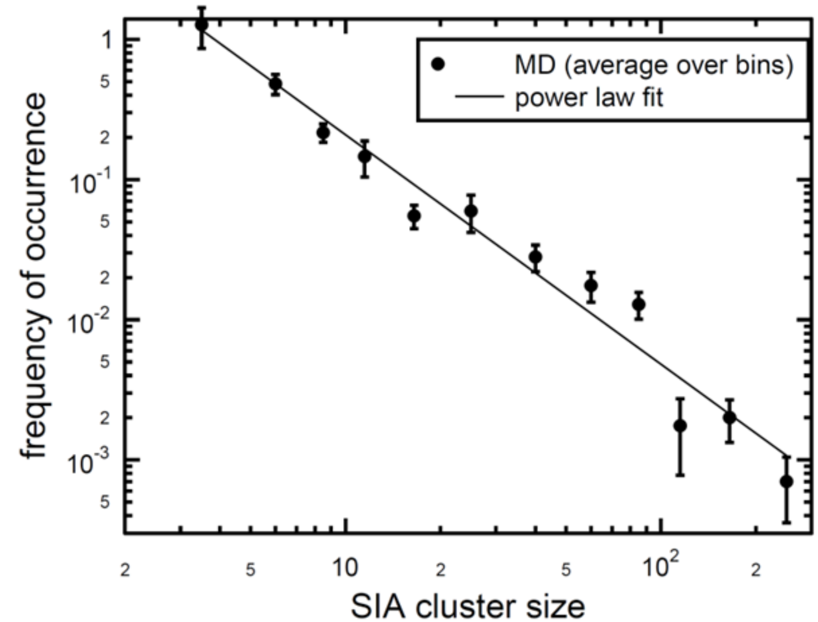
- Defects in energetic cascades occasionally form large clusters
- Strength of elastic interactions depends on size
- Thermal stability depends on size
  - Will the cluster grow or shrink at a given temperature?
- Formation of very large clusters rare events
  - In limited numbers of simulations, not likely to be observed
- *How can we know how often they form?*

200 keV cascade damage in W

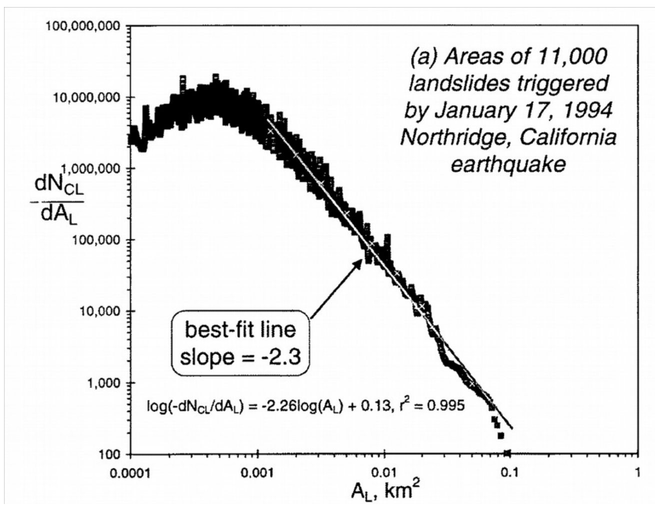


Defect size scaling laws

- The fractal nature of cascades leads to a power law distribution of defect cluster sizes
  - Typical of self-organized critical phenomena
  - Other examples: earthquakes, landslides, forest fires
  - Rare events statistically significant



[A. E. Sand, S. L. Dudarev, and K. Nordlund, EPL **103**, 46003 (2013)]

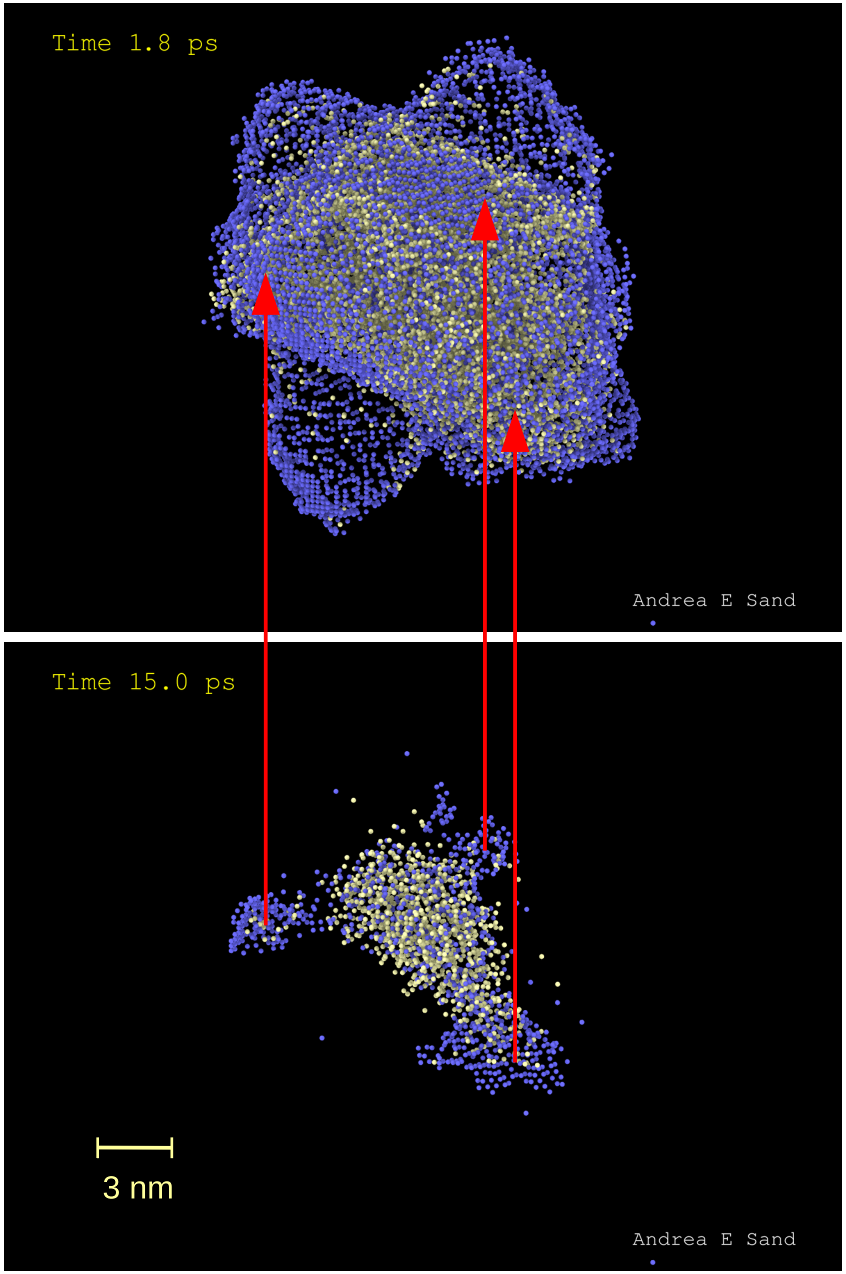


[Turcotte, Rep. Prog. Phys. **62** (1999) 1377–1429]

- Although large clusters form rarely, they contain most of the defects
- Significant impact on defect evolution
- Exponent < 2 : no well defined average size

Formation of clusters

- Large SIA clusters form *inside* perimeter of dense, nearly spherical cascades
  - 150 keV cascades reach a maximum of ~ 30 000 “liquid” atoms → diameter ≈ 10 nm
  - Alternative to spherical geometry is an extended, irregular liquid area → no large defects
- Different formation mechanism of single SIA
  - Product of replacement collision sequences, mainly *outside* liquid area
  - Also formed in irregular, extended cascades
- Pressure wave in “lobes”
  - Does not create damage
  - Neither does it remove single SIAs
- **Size of cascade region gives upper bound on defect size distribution**



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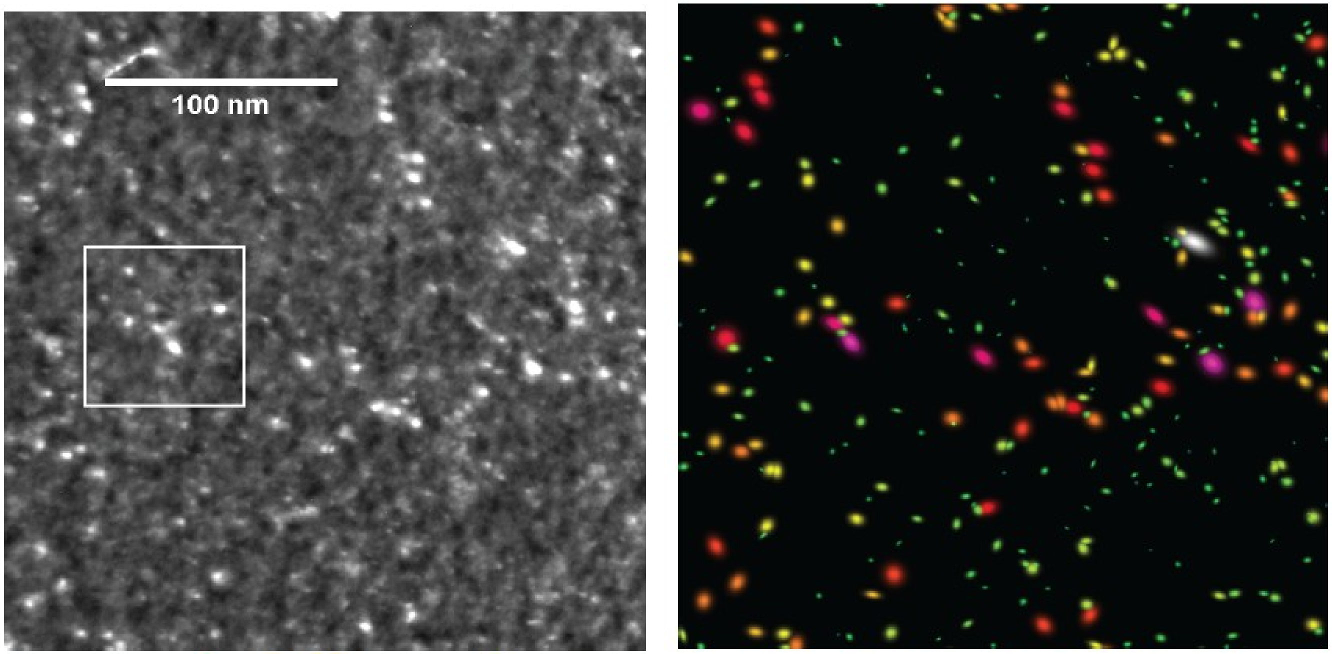
Comparison to experiment



[X. Yi, A.E. Sand, D.R. Mason, M.A. Kirk, S.G. Roberts, K. Nordlund and S.L. Dudarev, EPL **110** (2015) 36001]

- Automated analysis of in-situ TEM, 150 keV $W^{+} \rightarrow W$
- Experiments performed at 30 K
  - Reduced defect mobility
  - Some loop loss
  - No visible growth of defects
- Observing more or less the primary damage

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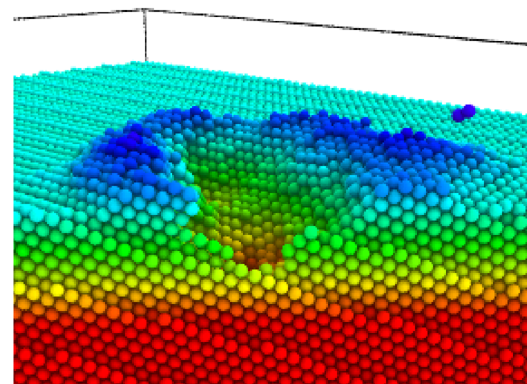
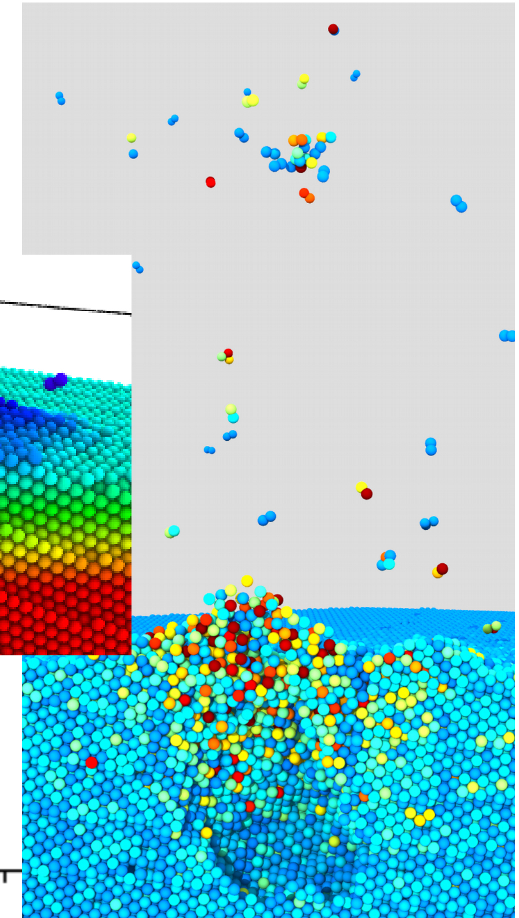
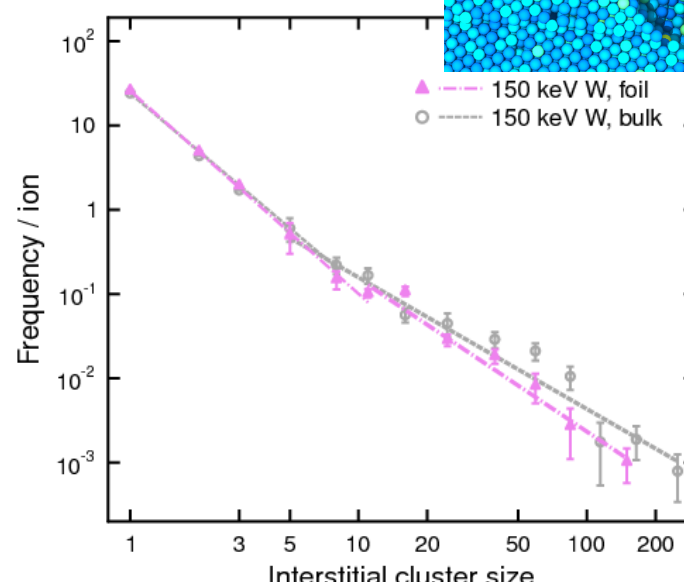
Foil irradiation in MD

- Material flow to the surface in shallow impacts results in sputtering and deposited adatoms on the surface
  - Increase in vacancy-type defects
  - Decrease in SIA clusters
- Deeper cascades strongly resemble bulk cascades
  - Surface effects due to image forces on dislocation loops do not have time to affect deeper defects on cascade time scale (~ 60 ps)
- Formation of large interstitial-type clusters slightly reduced on average
  - Slight change in slope of SIA cluster size distribution





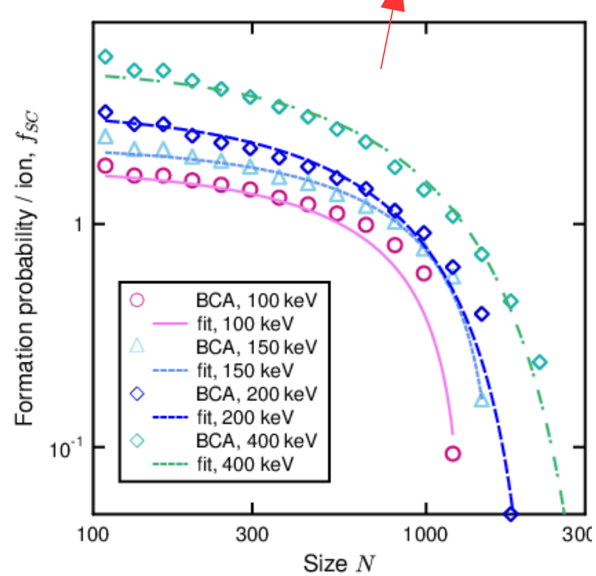


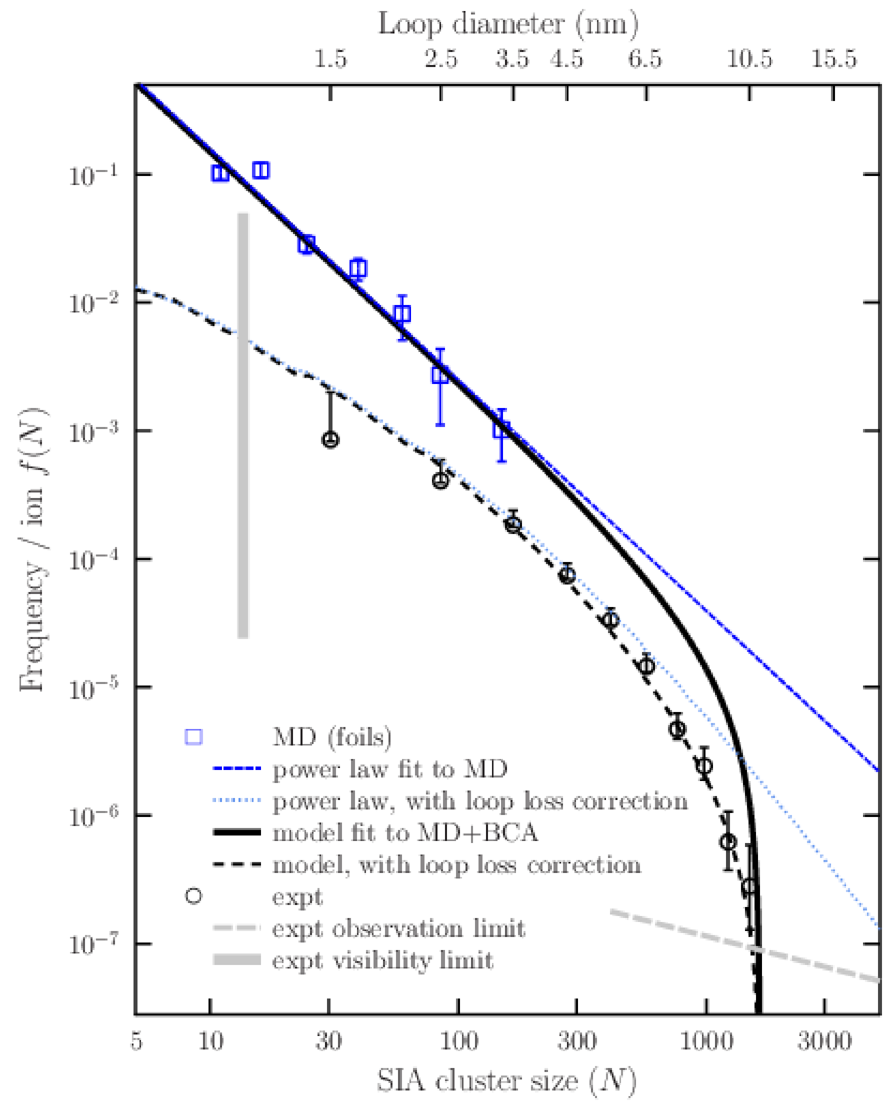
[A. E. Sand, *et al.*, EPL **115** (2016) 36001]

Size distribution

- Combining defect statistics with probabilities for (sub)cascades of various sizes gives an upper limit to the size distribution
  - Error bars for experiment derived from estimate of ”invisible” loops
  - Correction for estimated loop loss gives dotted black line from MD
  - *No fitting to experiment!*

$$f(N) = \frac{A}{N^S} \times B((N_c - N)/N_c)^{\kappa}$$





[A. E. Sand, et al. MRL **5** (2017) 357 - 363]

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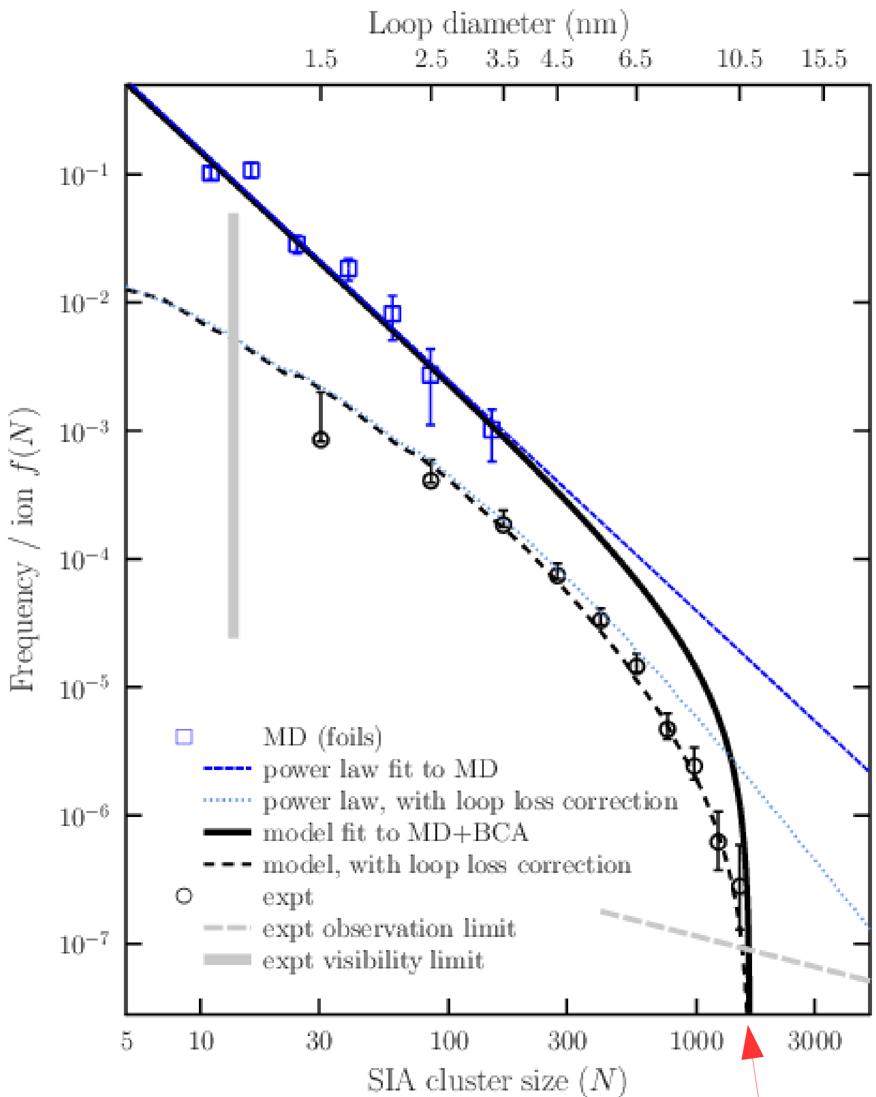
$$f(N) = \frac{A}{N^S} \times B((N_c - N)/N_c)^{\kappa}$$

Maximum cluster size

- Describes the full defect size distribution
  - **Below visible range in TEM**
  - **Beyond direct accessibility by MD**
- Well defined upper size limit
  - Computationally expedient

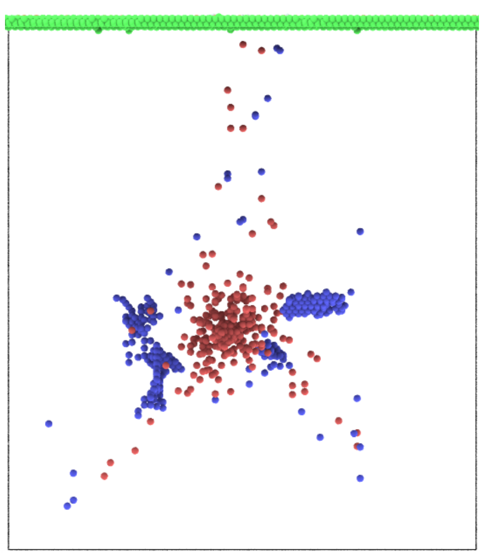


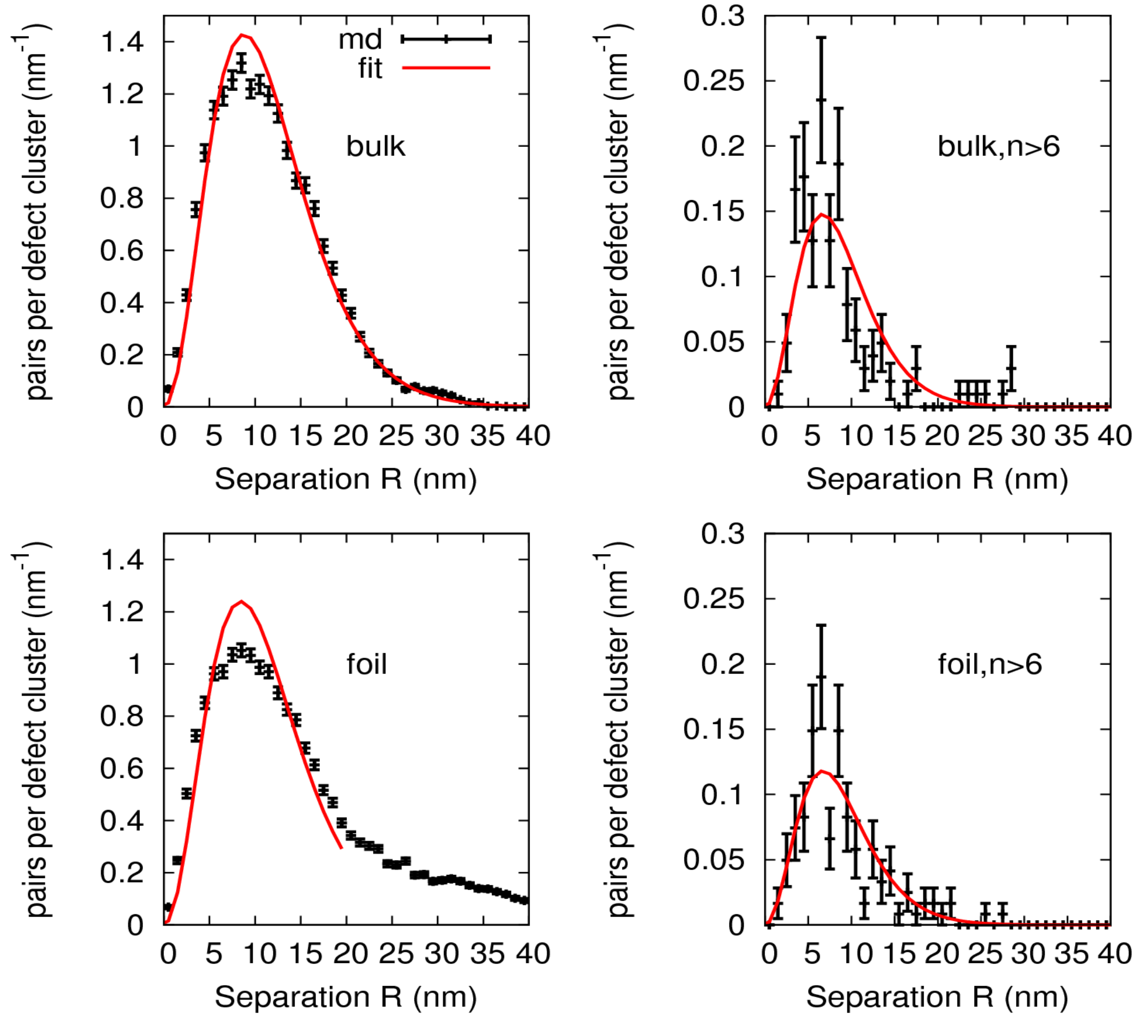
[A. E. Sand, et al. MRL **5** (2017) 357 - 363]

Maximum cluster size

Spatial distributions in MD

- Loops/clusters closer together than single SIAs on average
- A tail at large separation seen in foil
  - Due to trails of point defects along channeled ion tracks

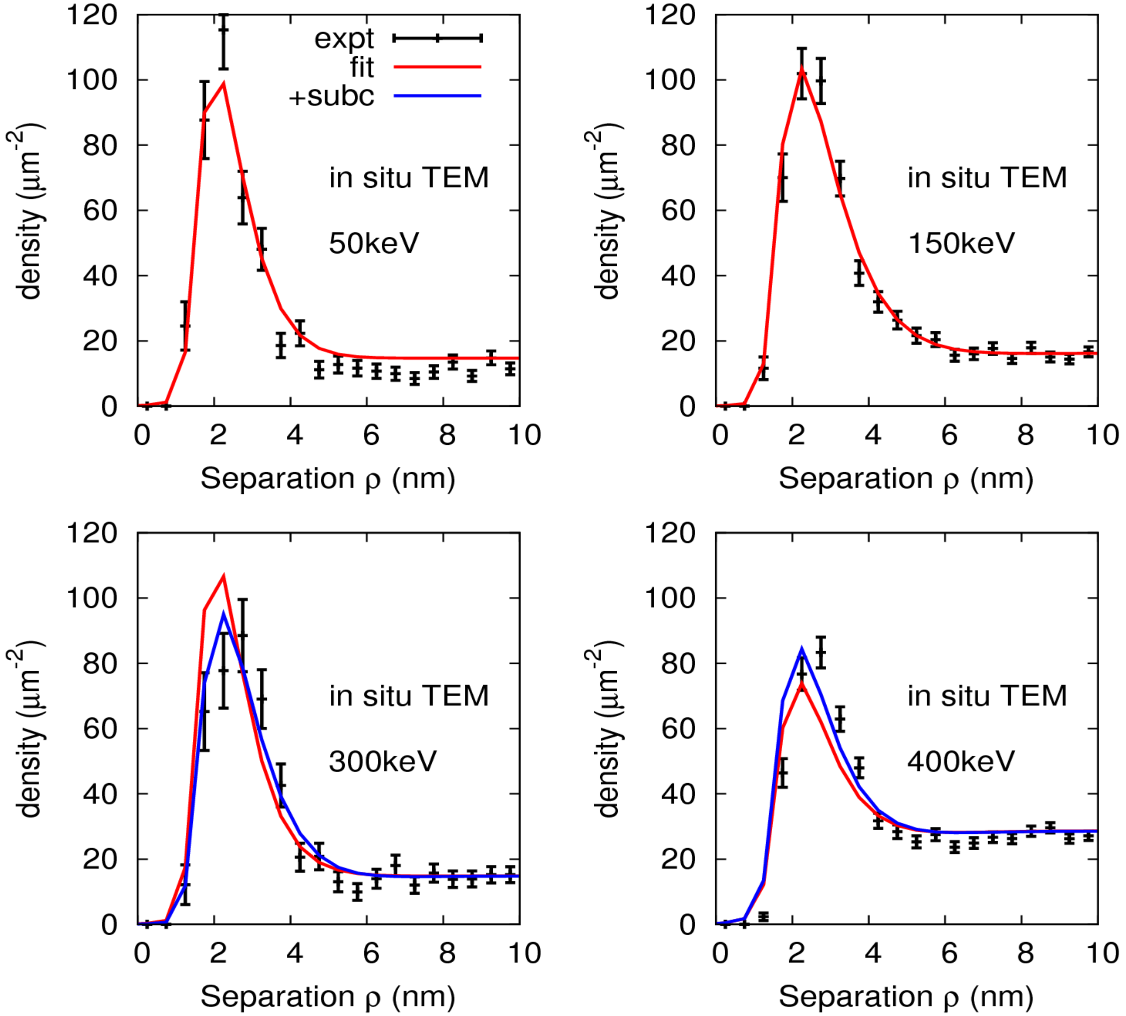




[D. Mason, A. E. Sand, X. Yi, K. Nordlund, S. L. Dudarev, Acta Mat. (2017) *in review*]

Spatial distributions in TEM

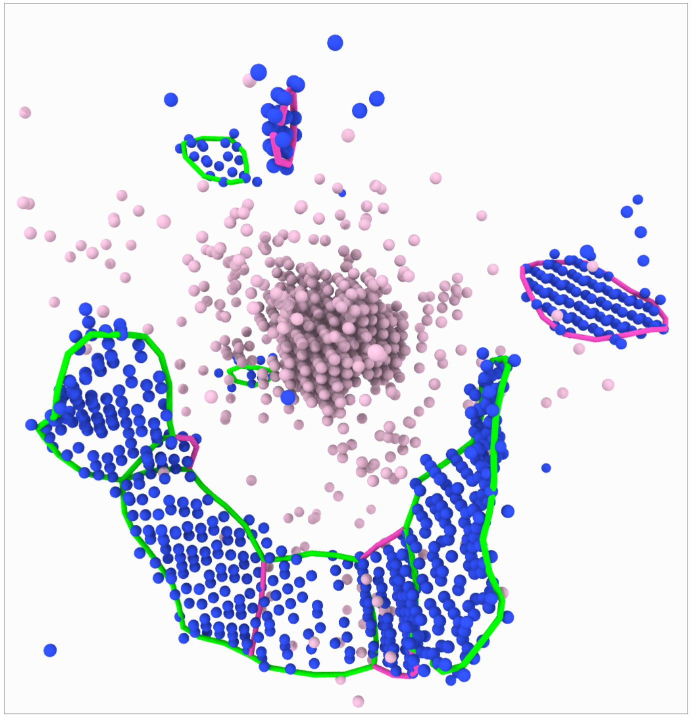
- Radial distributions from TEM micrographs show clear peak for closely spaced pairs of loops
  - Defects from single cascades
- Compared to MD, experiment predicts smaller peak separation
  - Loss of outliers?
    - Preferential trapping of closer pairs
  - Larger clusters closer together?
    - MD cannot give statistics for > 1 nm
  - Elastic relaxation?
    - Pairs will be trapped at point of closest approach of glide cylinders



[D. Mason, A. E. Sand, X. Yi, K. Nordlund, S. L. Dudarev, Acta Mat. (2017) *in review*]

Effect of size

- Simulations of cascades show large loops forming in dense cascades
  - small separation between loops
- Smaller loops and clusters form readily in spatially extended cascades
  - Can have large separation
- BUT dense cascades and large defects occur rarely
  - Seen only in a few simulations
  - In TEM, on the other hand, small defects are invisible
  - Not enough data to determine whether spatial separations have a dependence on size



Conclusions

- Intermediate range of potential (interpolation region) affects results for lower and intermediate PKA energies
  - Harder potentials with reasonable TDE agree best with DFT
- Details of electronic energy losses very important in W
  - Magnitude of energy losses can be estimated by comparison to IBM experiments
- Size distribution of defects in W in good agreement with TEM
- Analysis of TEM shows that multiple “visible” loops are formed in individual cascades
  - Agreement with MD for certain potentials and electronic stopping!
- Prediction of peak separation distances for pairs of loops in slight disagreement with TEM
  - Simulations indicate larger loops may on average have closer separation, but statistics not enough to quantify effect
- **Rare events, not easily captured by MD, are important for microstructural evolution**
  - **Likely larger impact in fusion than in fission due to energetic recoils**

Extra slides