Post-processing MD simulations to identify defects, their trajectories and clustering in Molecular Dynamics Simulations using LAMMPS

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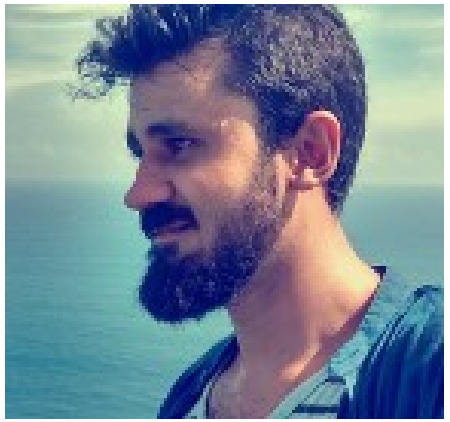


Scope (I) MD simulation data of collisional cascades in materials upon irradiation and (II) Establishment of an internationally agreed library of such data at IAEA.

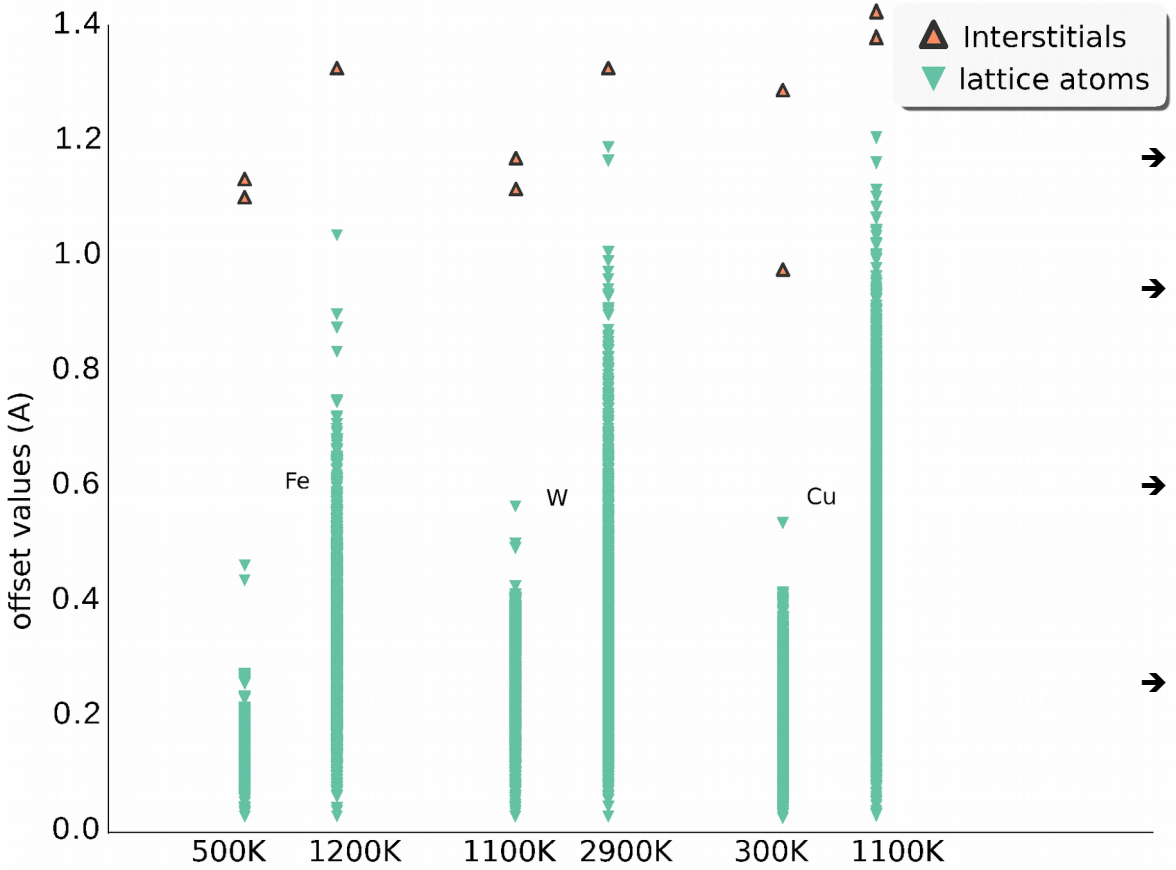
Overview

- Post processing of data to identify:
  i. Interstitials and vacancies.
  ii. Their in-cascade clustering.
  iii. Their trajectories (to gain physical insight – ex: how dominant is the mechanism proposed by Vineyard?), and
  iv. Their distributions – for input to higher scale simulations.

- MD simulation standards / protocols:
  i. Ensembles, boundary conditions, size of simulation, etc
  ii. Stiffening/Validating potentials, handling electronic loss, etc
  iii. Extending scope – defect diffusion, displacement energy, etc.

Max-space clustering method to identify point defects

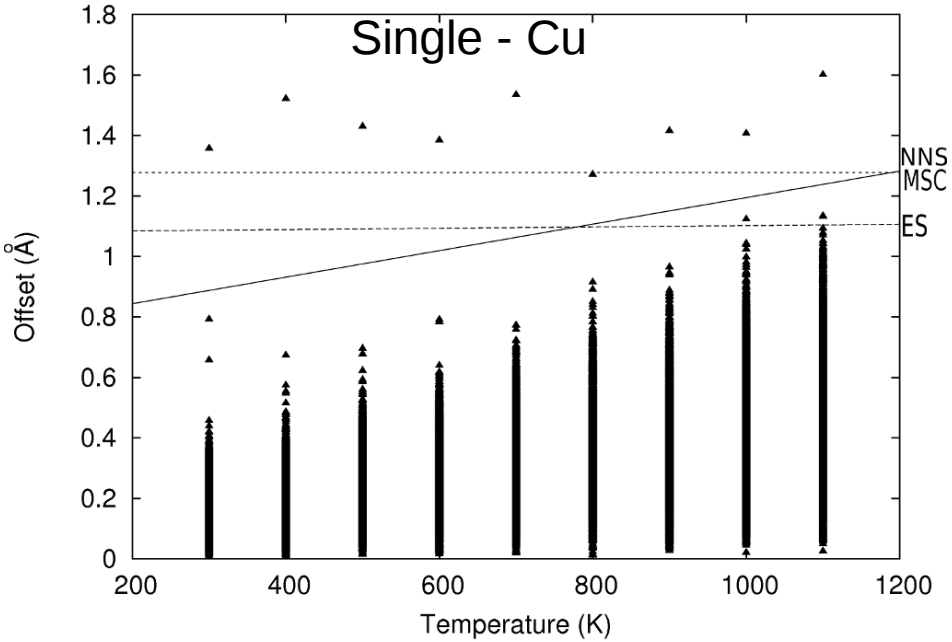
- Create a perfect crystal with a single interstitial and relax it (NPT).
- Obtain offsets – i.e. displacement of atom from its original position.
- Sort the displacements in decreasing order.
- Identify two successive offsets having maximum space between them. The mid point between them is the “threshold offset”.

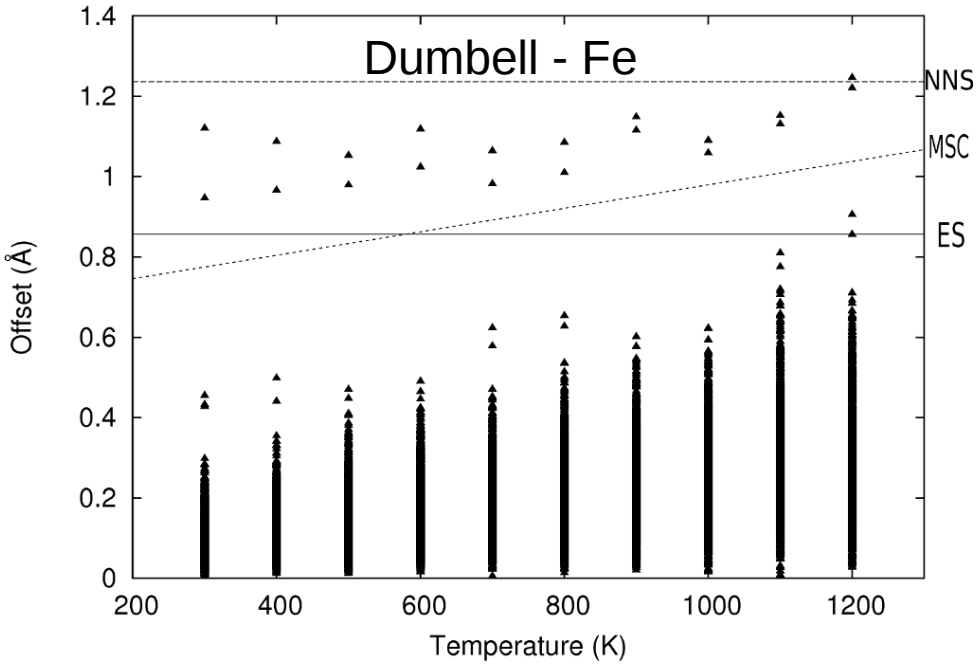


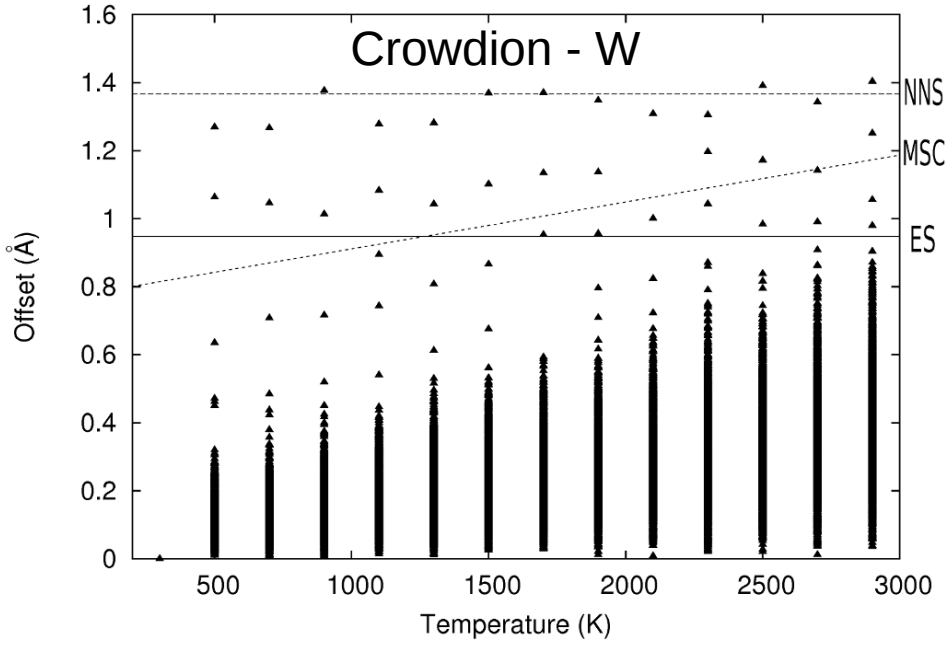
- Cut-off radius depends on T.
- Observe offsets of interstitial neighbors.
- Dumbbells and crowdions can be identified.
- How does the cut-off radius vary with temperature?

How does the cut-off radius vary with temperature?

- Single frame data points.
- For final analysis we average over several equilibrated frames.
- Did analysis for 5 bcc and 6 fcc elements.
- **Linear variation with T**







Linear variation of the cut-off radius with temperature

Ele.	Lattice	Slope (Å/K)	Y-intrcpt (Å)	a (Å)	r/a %
Cr	bcc	1.29×10^{-4}	0.87	2.88	30.06
Fe	bcc	2.92×10^{-4}	0.77	2.86	26.97
Mo	bcc	1.61×10^{-4}	0.8	3.15	25.40
Nb	bcc	2.47×10^{-4}	0.93	3.3	28.18
W	bcc	1.38×10^{-4}	0.82	3.16	25.79
Ag	fcc	5.96×10^{-4}	1.0	4.09	24.45
Au	fcc	4.29×10^{-4}	1.04	4.08	25.39
Cu	fcc	4.38×10^{-4}	0.89	3.615	24.54
Ni	fcc	3.67×10^{-4}	0.81	3.52	22.98
Pd	fcc	3.66×10^{-4}	0.95	3.89	24.42
Pt	fcc	3.10×10^{-4}	0.96	3.92	24.49

[Jnl. Nucl. Mater., **484** (2017) 258-269]

In general, bcc atoms show higher value for “Y-intercept/a”
The values are close to the “30% of a” suggested by Stoller et. al.

Identifying defects in-situ in collision cascade simulations Using LAMMPS

- **compute** cfrenk all frenkel/local cutoff_rad vac_rec_rad
  Outputs "positions", "id" and "type of defect" of candidate defects

- "cfrenk" outputs can be post-processed to
  i. Correct for dumbells and crowdions,
  ii. Obtain Number displaced / recombining / channeling, Maximum Displacement, PKA Displacement, etc,
  iii. Obtain defect distributions,
  iv. Evaluate in-cascade clustering,
  v. Study mechanisms of Frenkel pair formation / recombination

In situ calculation time is offset by the savings in I/O of all atomic positions for post-processing

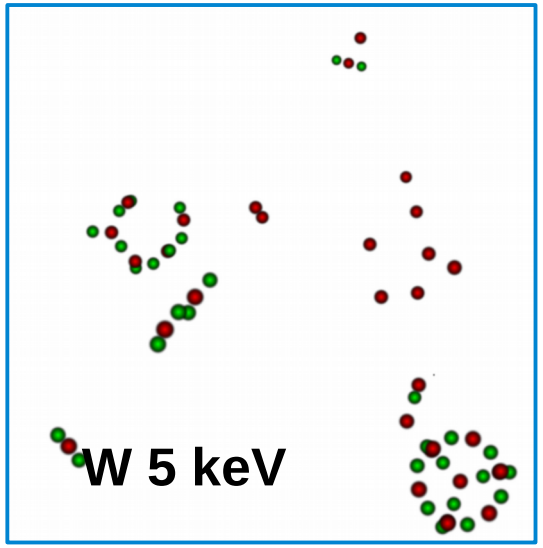
https://www.github.com/haptork/easylambda

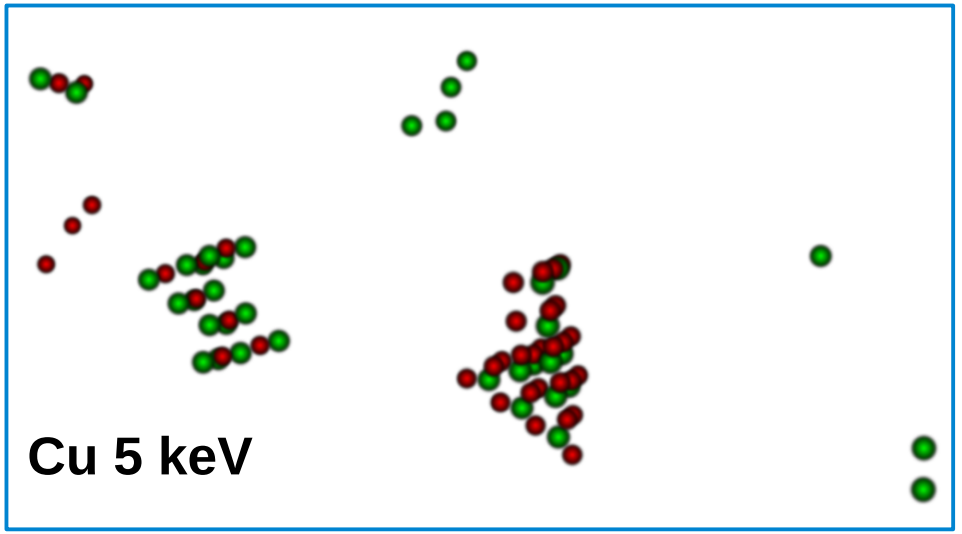
Clustering and correcting for dumbells and crowdions

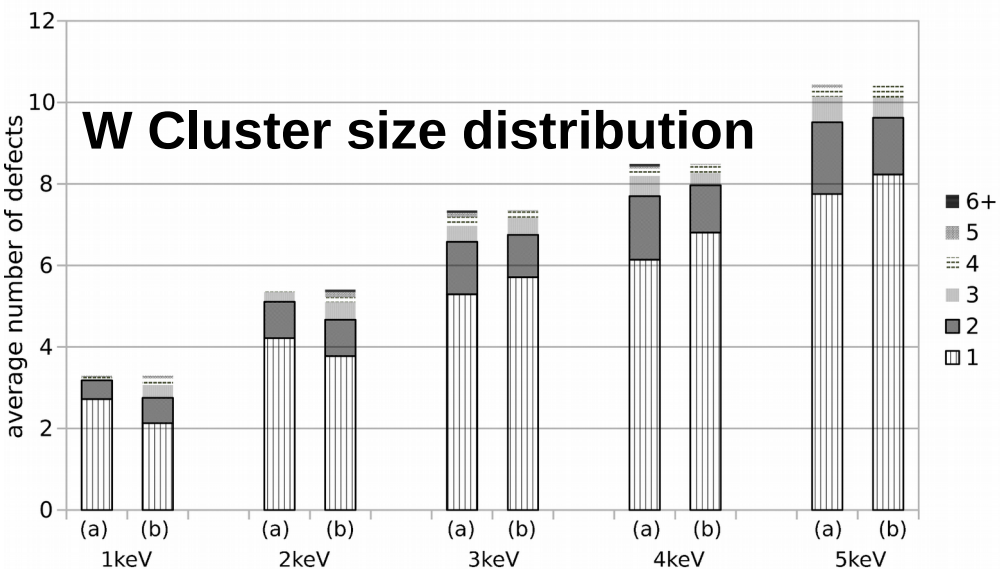
- **Use Union-find data structure to cluster defects closer than 1-NN**
  1) Initiate a Union-Find data-structure with every defect as the only member of its own group.
  2) If distance between any two defects is less than 1-NN union their groups.
  3) Tag each defect with the group it belongs to.

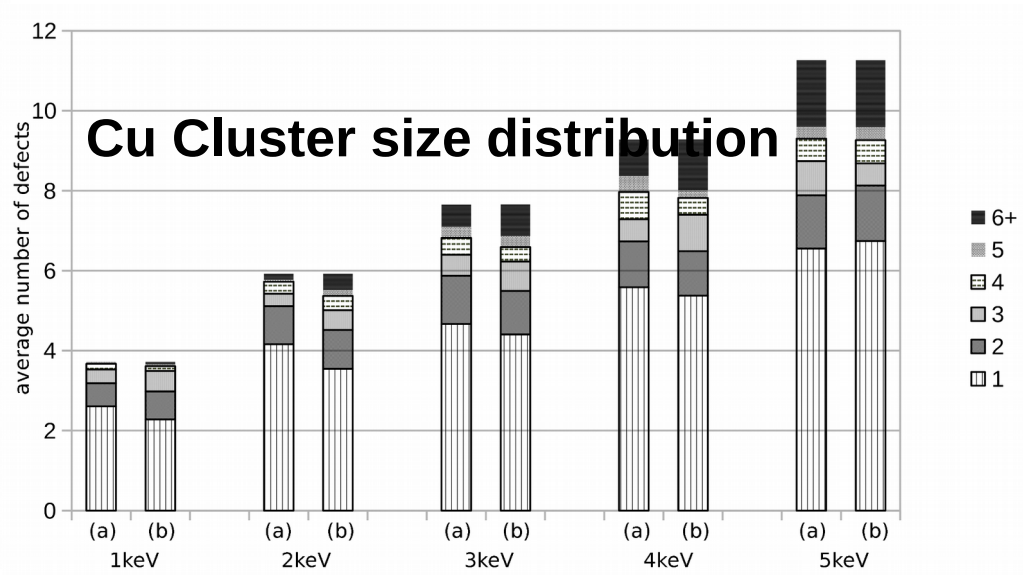
- **Correcting for Overcounting due to dumbells/crowdions**
  a) Label all vacancies as -1 and defects as 1
  b) Add all the labels within a group
  c) The resulting number is the number of defects in the cluster and the sign indicates if it is a cluster of vacancies (-ve) or interstitials (+ve).
  d) For every species that is not the same as the group type, find the closest pair of the opposite species.
  e) Mark both defects in the pair as deleted from the group

Crowdions, dumbells and single interstitials for W and Cu





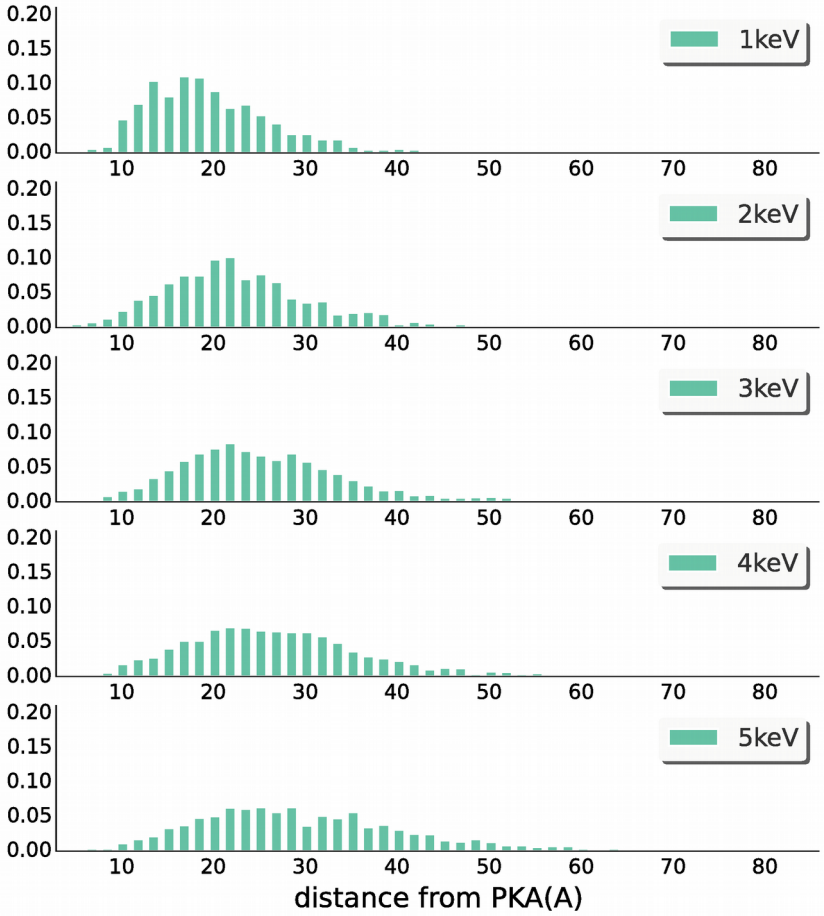




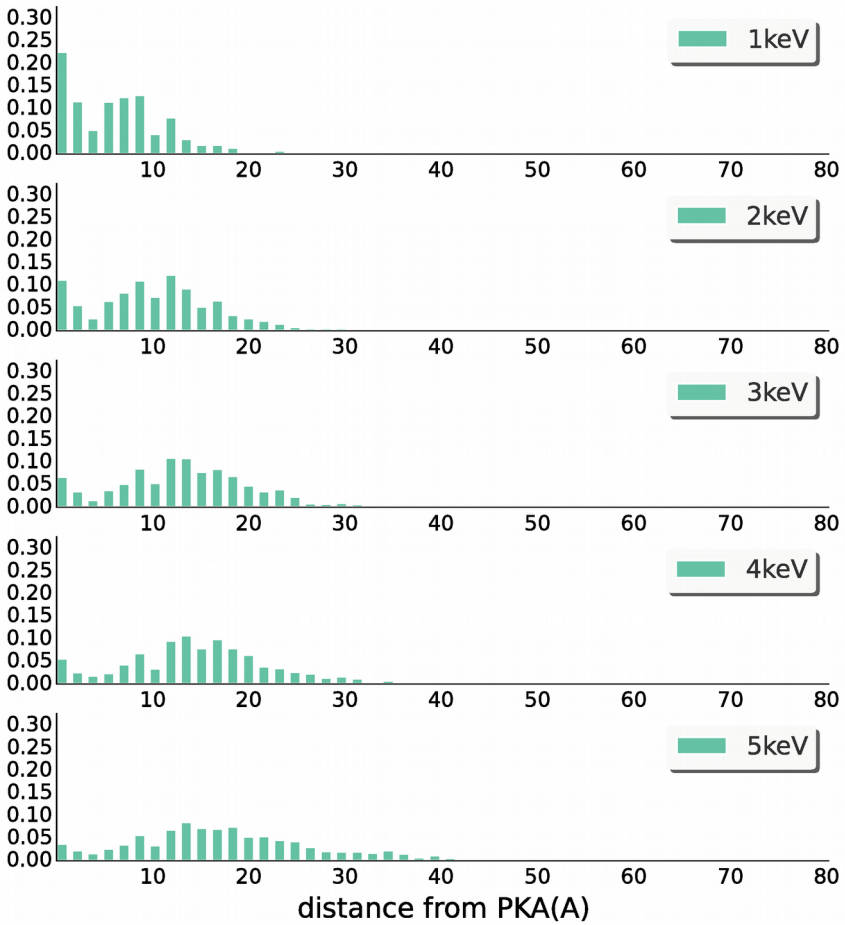
(a) for interstitials and (b) for vacancies

Cluster size distributions plotted after corrections

Defect Distributions for W



Interstitials

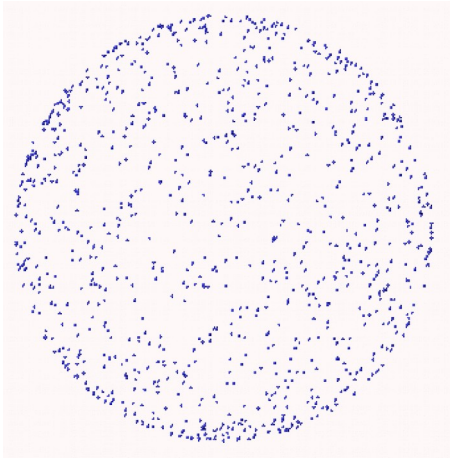


Vacancies

These results are averages over 200 random directions of the PKA

Molecular Dynamics Standards / Protocols

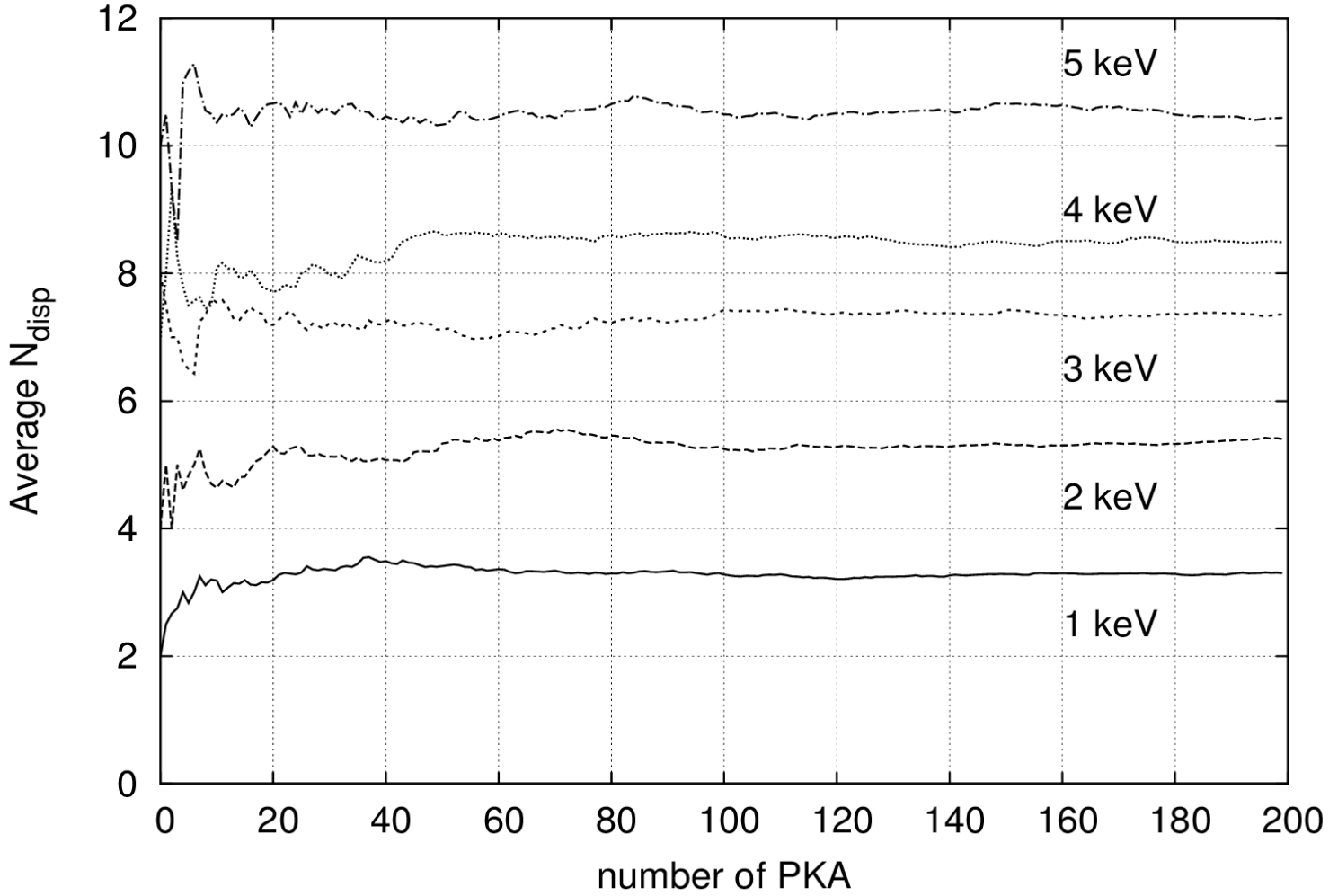
(Large-scale Atomic/Molecular Massively Parallel Simulator)



- Allows simultaneous launching of multiple MD runs for parameter scans – We scan random PKA directions.
- Ensembles used: PBCs along X-Y-Z, 10 ps NPT at 300 K, 10 ps NVE collision cascade.
- Variable time stepping.
- Boundaries fixed, with unit cells at the edges being temperature controlled. S**hock reflections from the fixed boundaries!?**
- Electronic losses not included. **Existing possibilities in LAMMPS (parameters for fix ttm for several elements are not available – would like to add a new fix to LAMMPS – like is being done in HCParCas?!)**

How many simulations are required for acceptable statistics?

Average number of defects as a function of number of directions explored ..

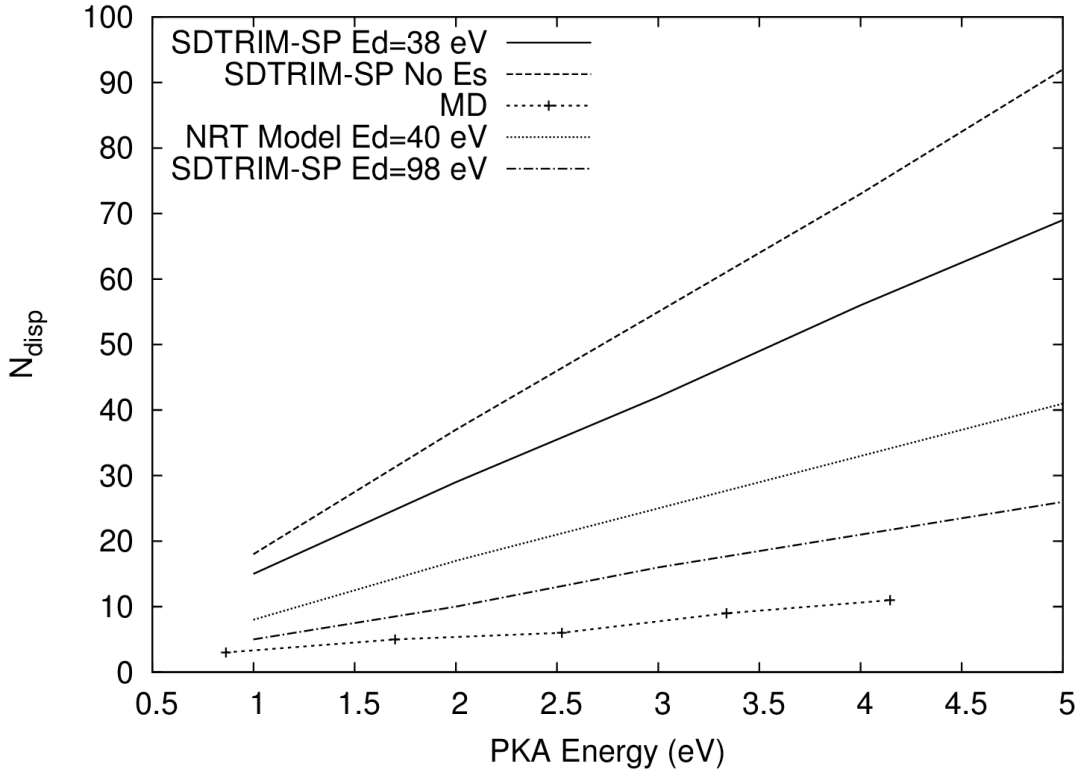


$$n_{avg,N} = \sum_{i=1}^{N} \frac{n_{d,i}}{N}$$

More than 60 random directions have to be sampled for n_{avg} to level off

Number of defects averaged over 200 directions - W

E_{PKA} (keV)	N_{disp}	$PKAD$ (Å)	$MaxD$ (Å)
1	3 ± 1	8.3 ± 3.1	9.51 ± 2.3
2	5 ± 2	11.7 ± 4.4	13.4 ± 3.5
3	7 ± 2	12.9 ± 6.6	16.3 ± 5.6
4	9 ± 2	15.7 ± 7.6	19.3 ± 7.0
5	10 ± 3	20.7 ± 13.9	25.1 ± 12.7



Maximum displacement shows that we have chosen a sufficiently large crystal size

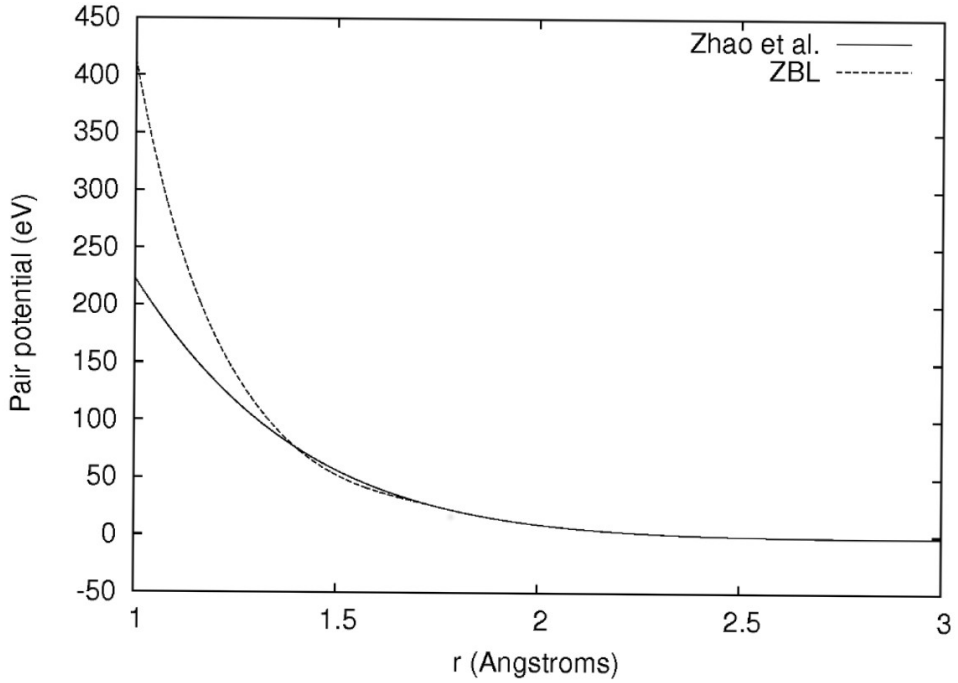
MD results in the plot are corrected for electronic stopping

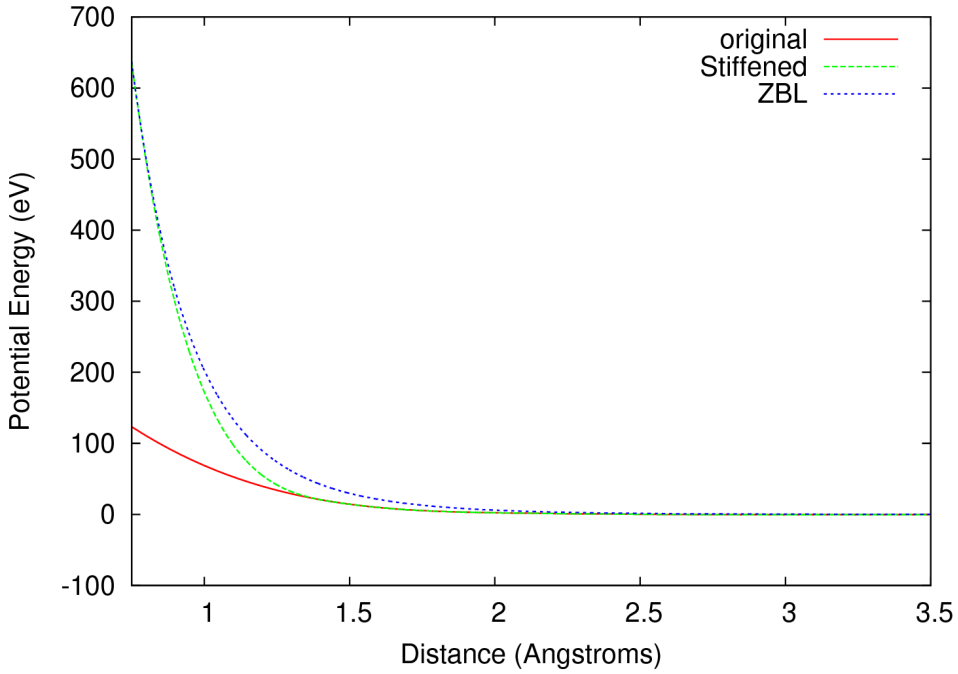
Vacancy–interstitial recombination must be introduced in SDTRIM-SP

Ed=98 eV from specialized MD simulations to obtain Ed (arXiv:1412.7452v1 – W. Setyawan Et al using W interatomic potential By Björkas et al.)

Choice of Ed crucial / important for MD – BCA-MC comparison

Stds.. / Protocols contd..: Potential stiffening



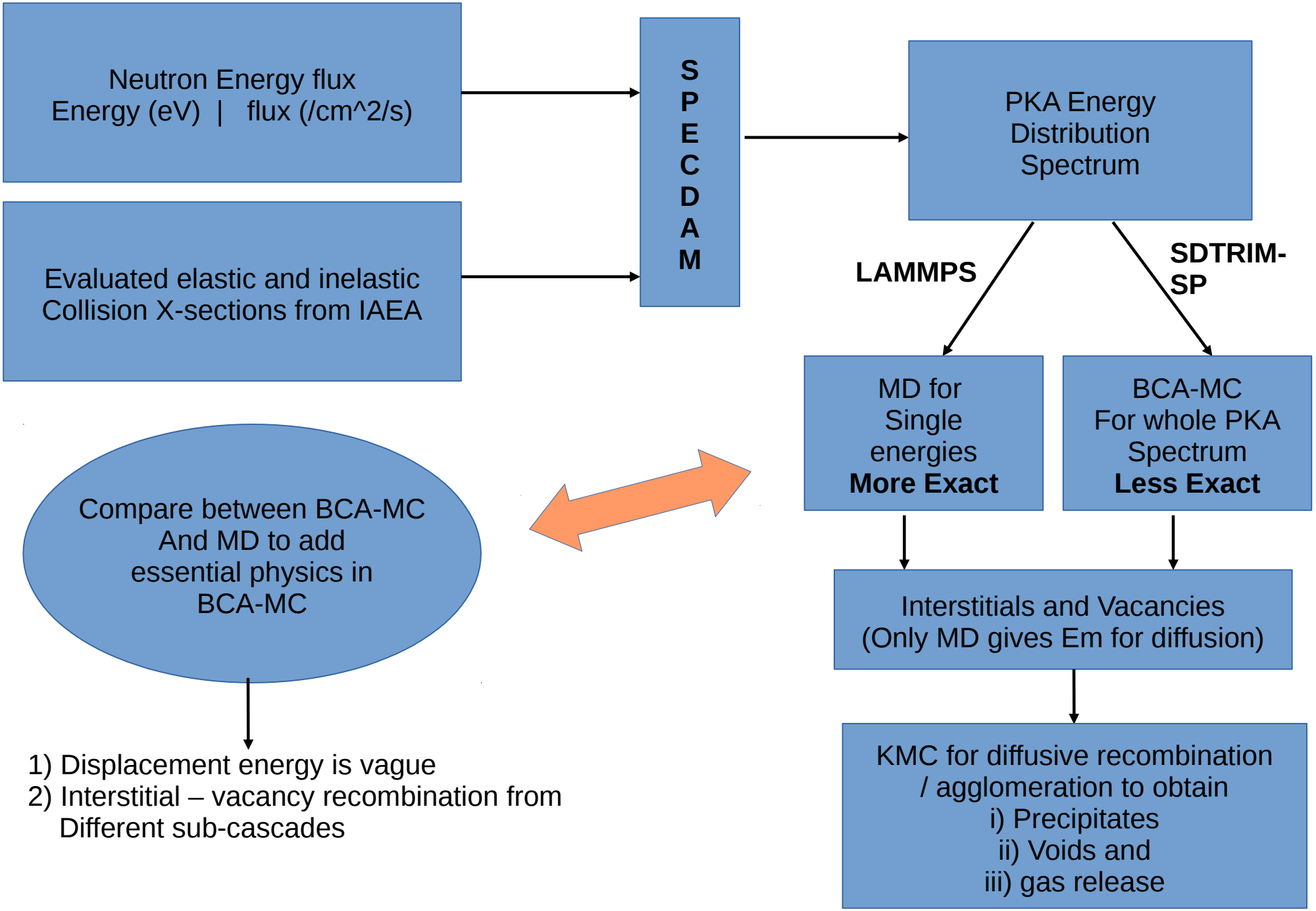


Typically bulk, equilibrium properties like vacancy formation energy, coeff of thermal conductivity, bulk modulus, lattice constant and elestic coefficients are matched before and after stiffening.

How to validate the stiffened, off-equilibrium pair potential?

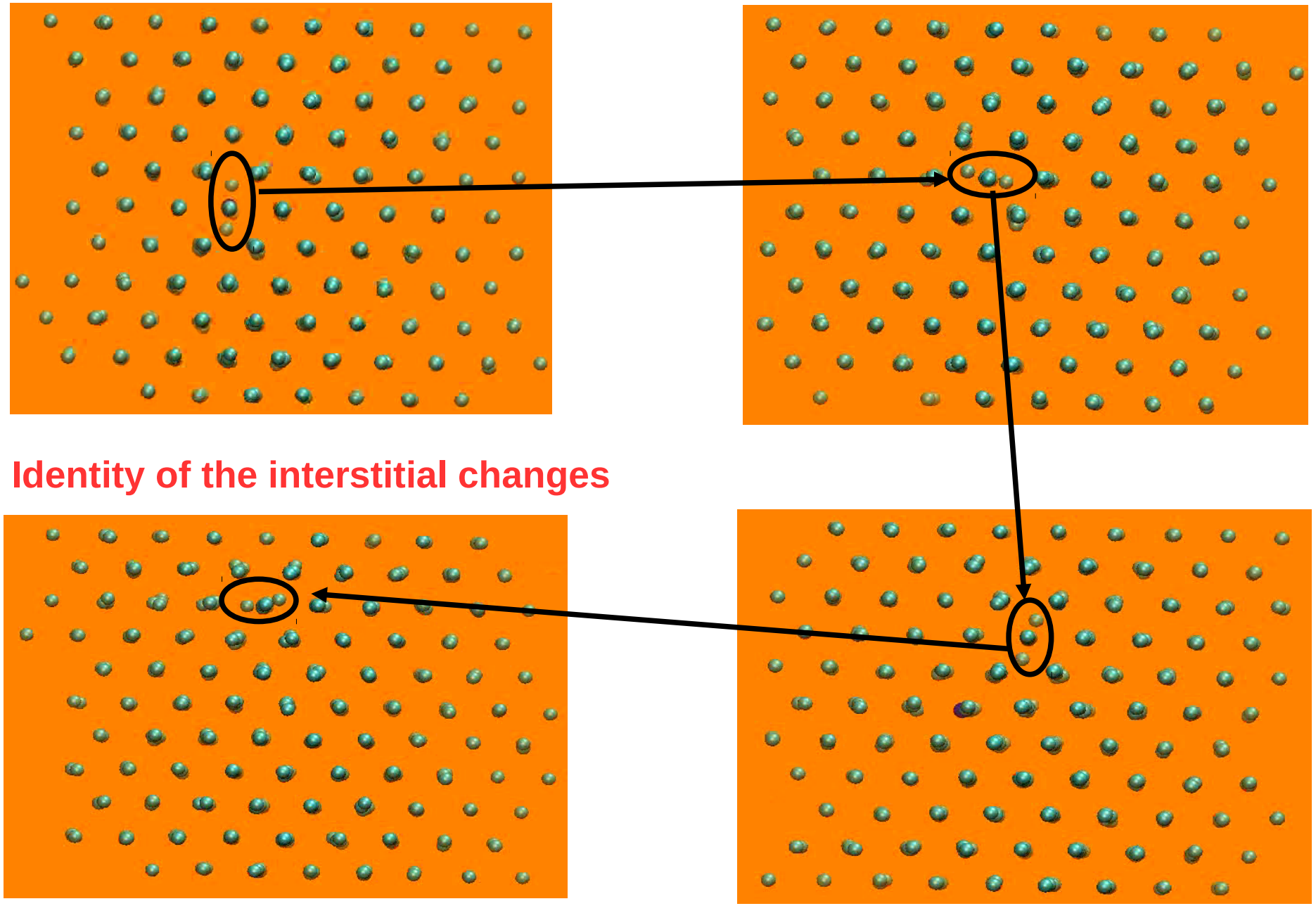
Ive been using cubic splines to connect from given pair potential to ZBL – too many iterations. Are polynomials better?

Extending scope: Inputs to higher scales from MD



1) Displacement energy is vague
2) Interstitial – vacancy recombination from
   Different sub-cascades

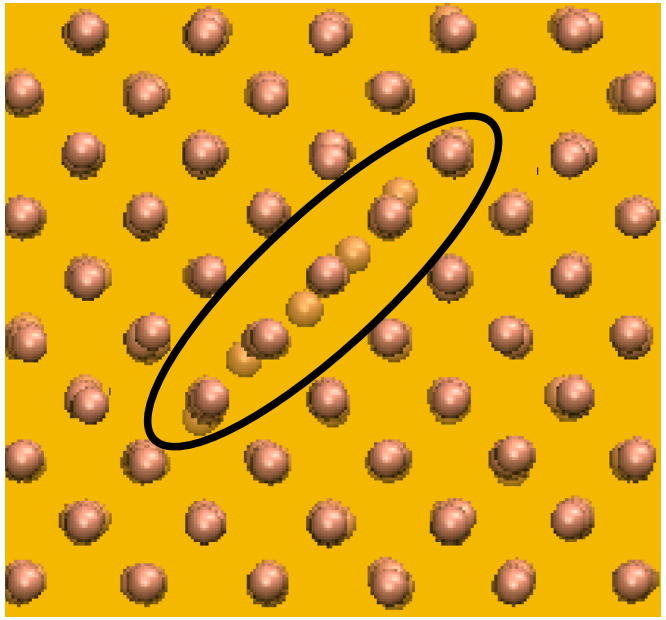
Extending scope: Interstitialcy Diffusion?



Identity of the interstitial changes

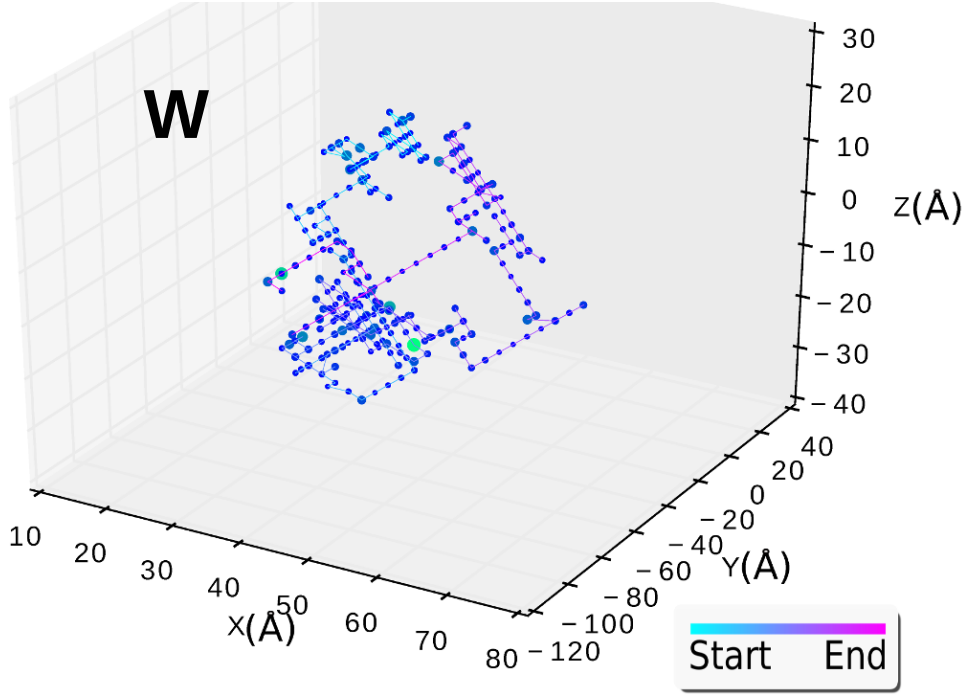
MD simulations with a single interstitial – first NPT and then NVE for 10 ns at 1100 K to 2900 K

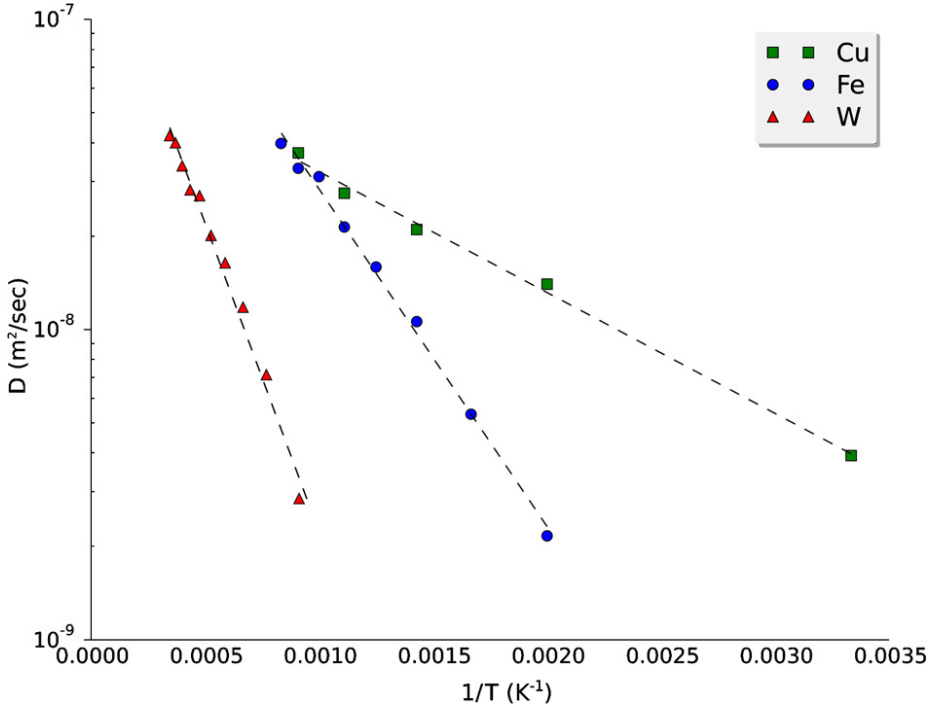
Extending scope: Inputs for higher scales?



<u>Obtaining details of the interstitial trajectory</u>:

- Candidate interstitials output in each frame are made **nodes of a graph-tree structure.**
- Various **cost functions are set up to identify the interstitial diffusion path.**
- **Jump correlation factors, migration energy and pre-factor for diffusion** obtained for 3 bcc and 4 fcc elements.
- Almost all jumps are **1NN**





Conclusions

✔ Max Space Clustering method to identify interstitials at different temperatures developed.

✔ The MSC method is coupled to a Union-Find data structure to identify defect clusters. Defect distributions corrected for dumbells and crowdions are obtained.

✔ Random directional statistics of defect data from MD simulations of collision cascades show that ~100 random directions are sufficient for the average number of defects to saturate.

✔ **Focus points:** Validation for potential stiffening? Electronic stopping? Effects of sub-cascade interactions at higher energies?

✔ Displacement energy, migration energy, jump correlation factors and other data are also interesting for scaling up studies of radiation damage. It is recommended that they be part of the proposed database?

Thank You