Towards a Standardised Methodology of Radiation Damage Defect Distributions for Microstructure Evolution Models

A. C. Gutiérrez-Camacho, M. J. Caturla

Dept. Física Aplicada, Universitat d'Alacant, Spain

9th Annual RaDIATE Collaboration Meeting, Granada, Spain, September 16-18, 2024

abel.gutierrez@ua.es

Outline

1. Motivation. Microstructure evolution of irradiated materials
2. Molecular Dynamics. The importance of the initial damage
3. Models to study microstructure evolution. Kinetic Monte Carlo & Cluster Dynamics
4. Our standardised methodology of radiation damage distribution
5. Conclusions

Motivation
Microstructure evolution of irradiated materials

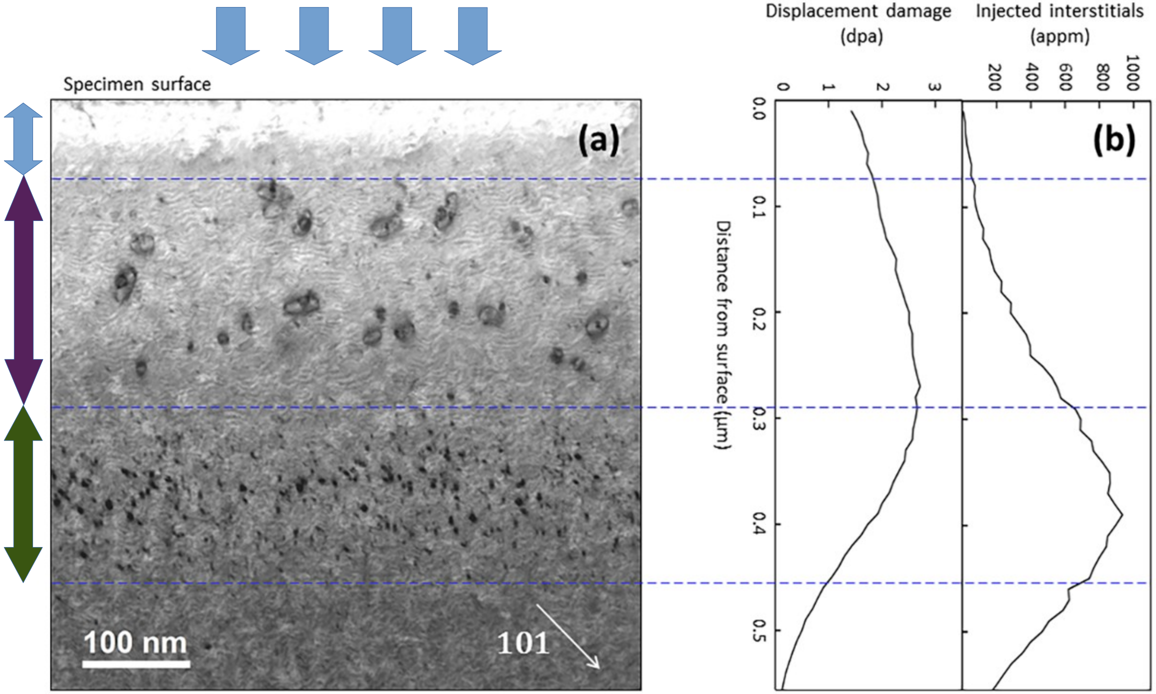
Experimental observations: 1 MeV Fe^{2+} in Fe9Cr at 300ºC

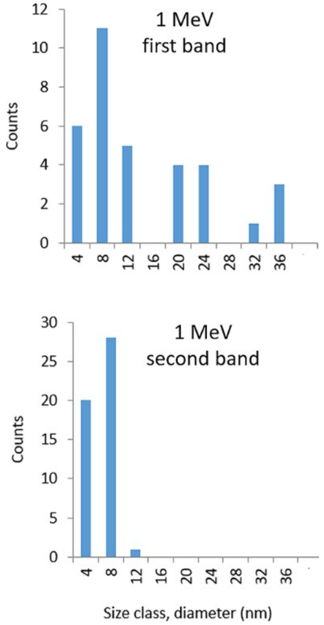
K. Vogel et al, Nuclear Materials and Energy 27 (2021) 101007

Denuded zone

Homogeneous distribution large clusters

Homogeneous distribution small clusters





Fluence 2.1×10^{19} ions/m^2

Defects form bands along the ion range of different sizes and densities

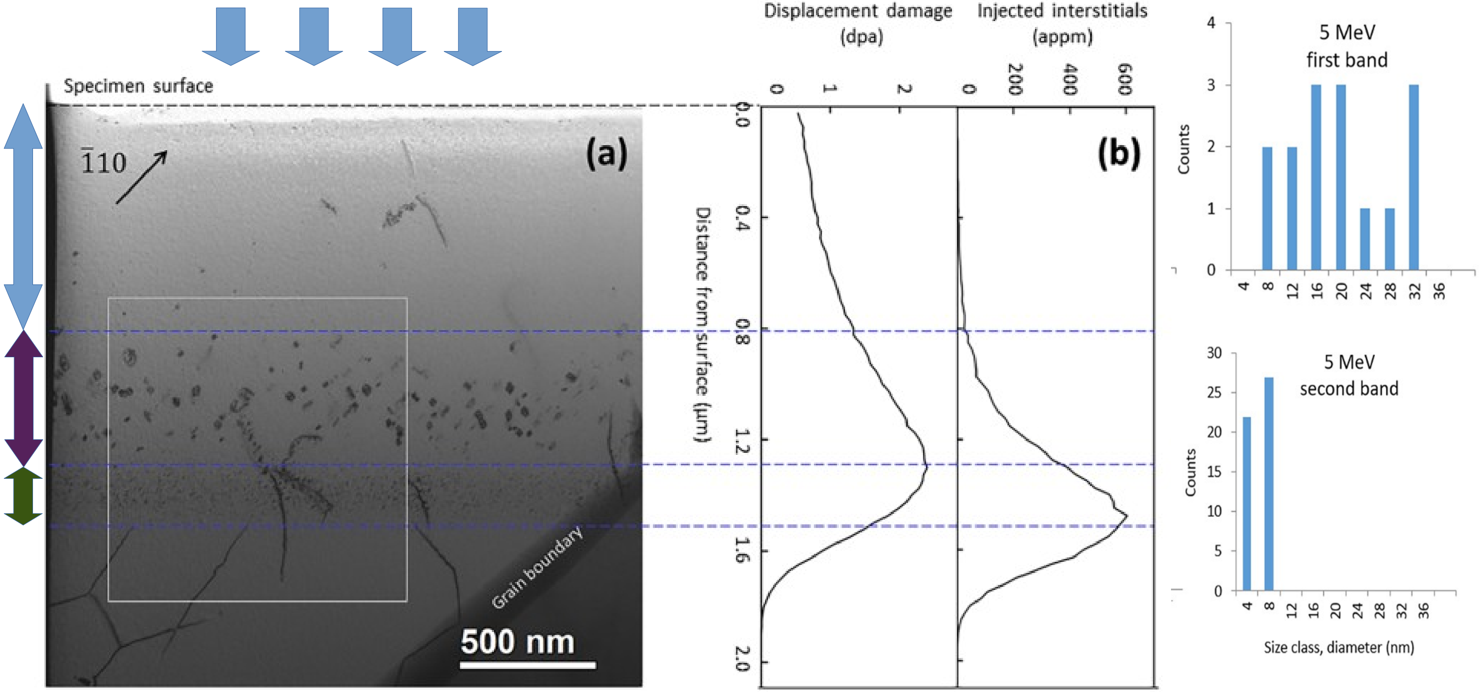
Experimental observations: 5 MeV Fe^{2+} in Fe9Cr at 300ºC

K. Vogel et al, Nuclear Materials and Energy 27 (2021) 101007

Denuded zone

Homogeneous distribution large clusters

Homogeneous distribution small clusters



Fluence 2.4×10^{19} ions/m^2

Defects form bands along the ion range of different sizes and densities

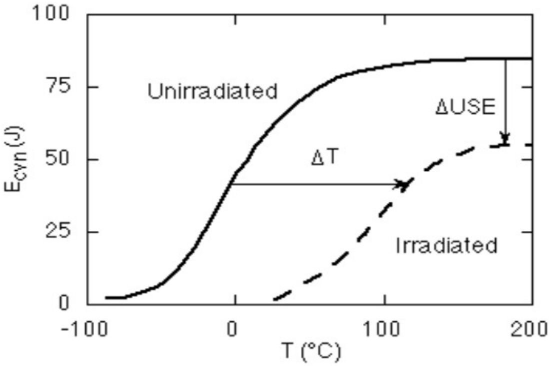
Exposure to radiation changes the mechanical behaviour of materials

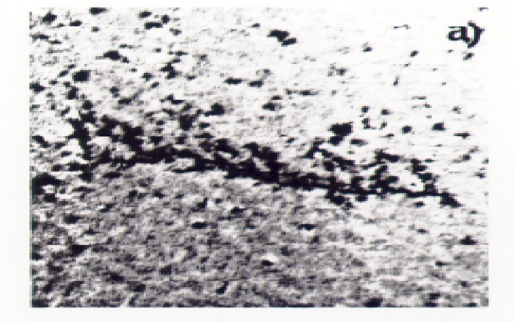
We need to understand microscopic processes to predict macroscopic properties

Macroscopic

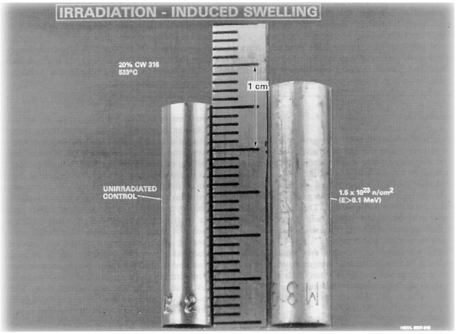
Microscopic

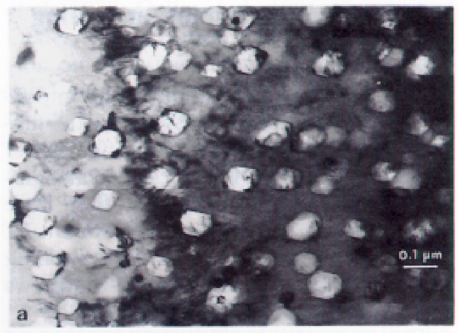
Embrittlement



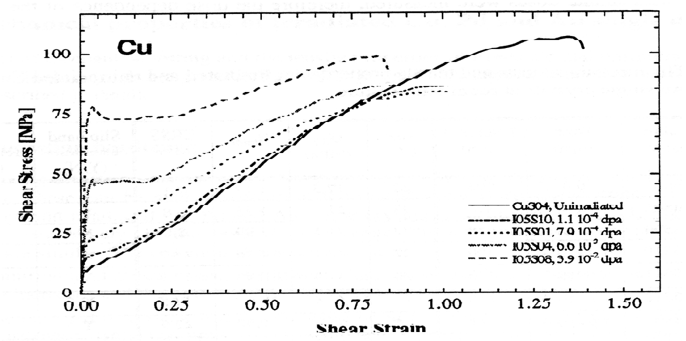


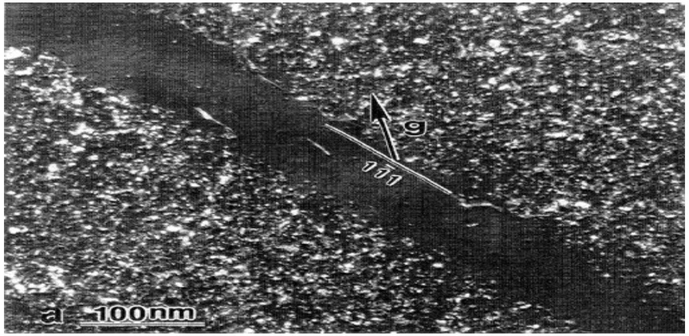
Swelling





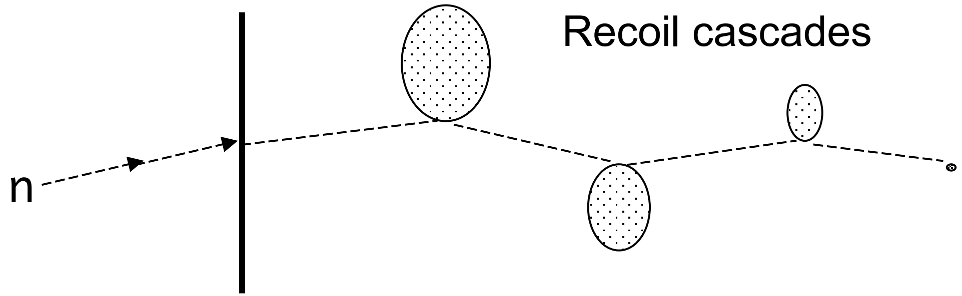
Plastic instability





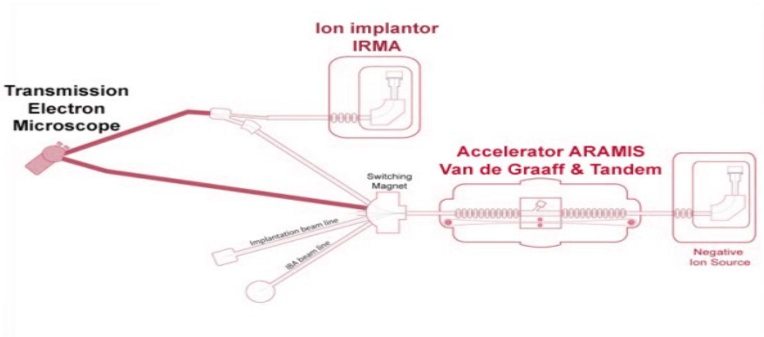
Models to extrapolate from ion irradiation to neutron damage

Need to understand neutron damage of ~MeV range:
Production of 10s keV recoils



Neutron experiments are complex, expensive and limited

Use of ion irradiation to understand defect production and its evolution



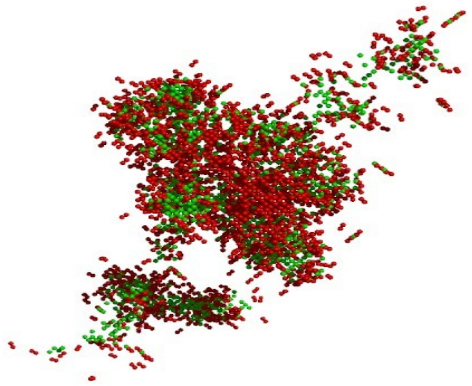
A dual beam and triple beam facility at CEA, France
https://jannus.in2p3.fr/

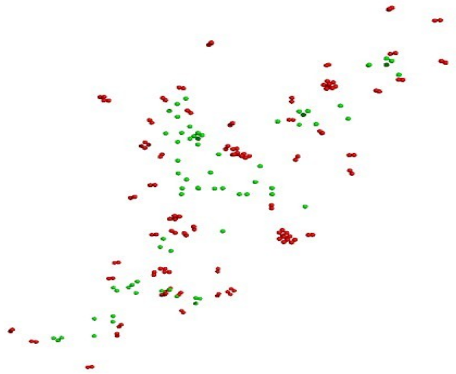
Model validation with ion irradiation
Extrapolation to neutrons

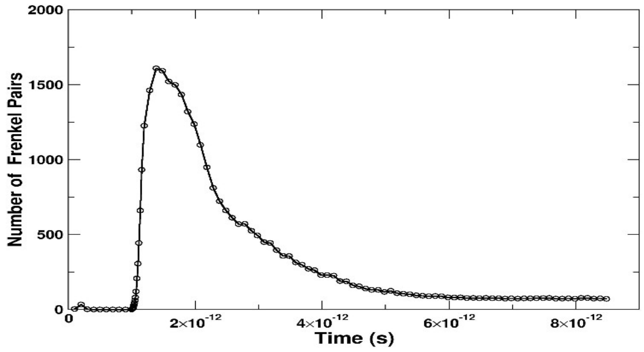
Molecular Dynamics
The importance of the initial damage

Primary damage: vacancies and SIAs

Fe in Fe, 30 keV



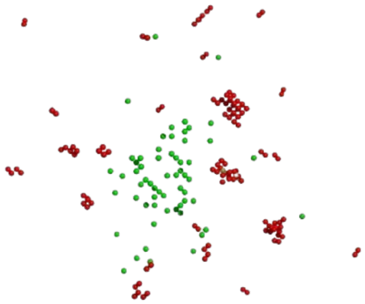




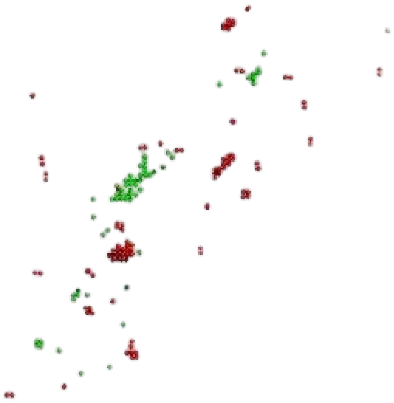
- Vacancies
- Self-interstitials

Time scale ps – ns, space scale ~10^3 nm^3, low energies

Initial defect distribution varies significantly between metals



bcc Fe
30 keV, 66 defects



fcc Cu
30 keV, 69 defects

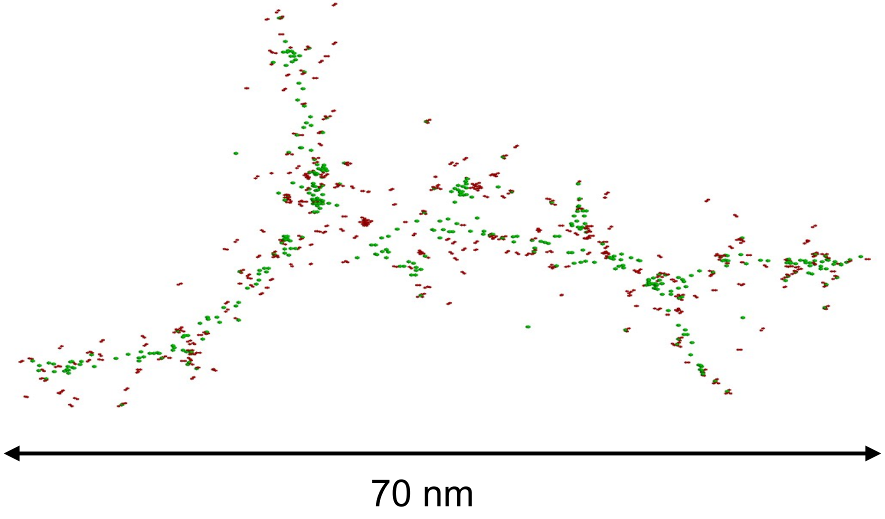
Vacancies
Self-interstitials

Similar number of defects, but more vacancy clustering in Cu than in Fe
This has important consequences in damage evolution

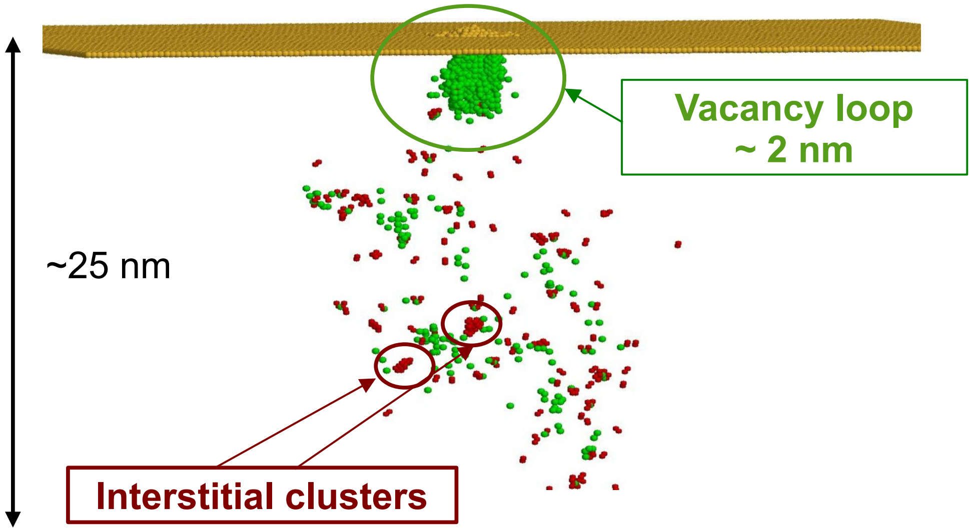
Bulk vs surface damage

100 keV Fe in Fe

Bulk



Surface



Vacancies

Self-interstitials

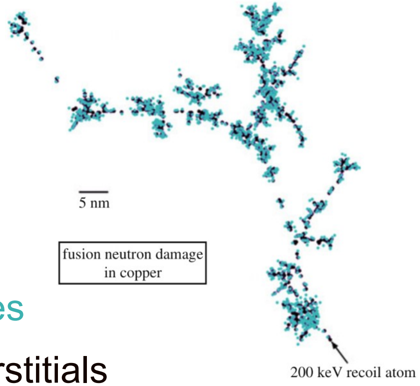
Aliaga, M. J. et al., Acta Materialia 101 (2015) 22-30

Irradiation of thin foils at low energies (50-150 keV) shows the formation of large (~1 nm radius) <100> vacancy loops.

BCA models

Binary Collision Approximation (BCA) models are a fast alternative to MD, but:

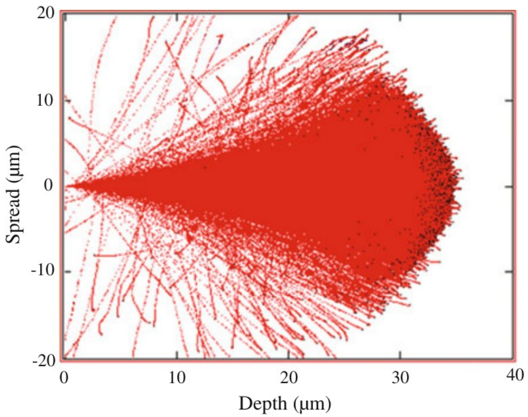
- No close range interactions
- No clustering
- No recombination
- Overestimation of the number of defects



Vacancies

Self-interstitials

(MARLOWE)



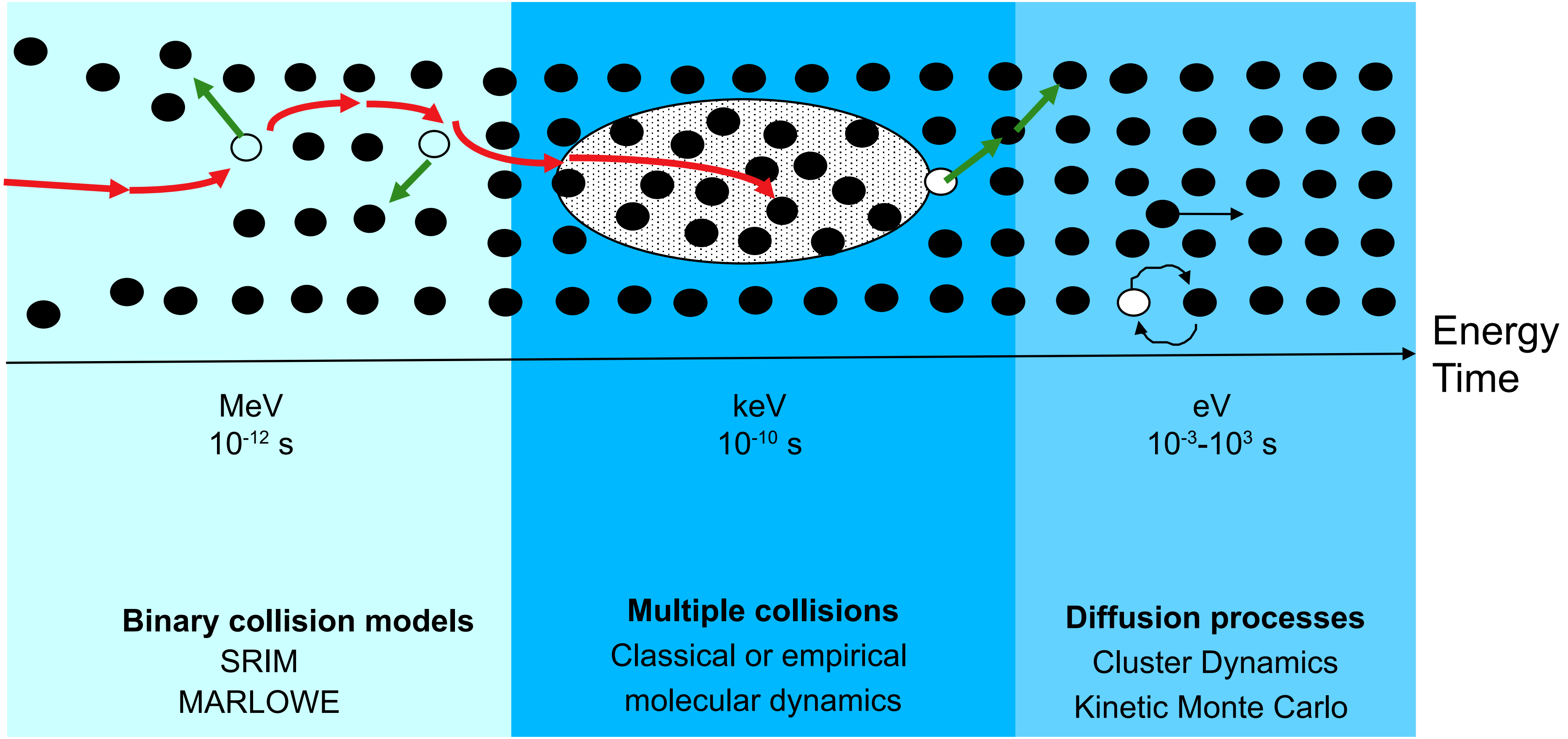
3 MeV proton in Ni (SRIM)

Was, G. S., Fundamentals of Radiation Materials Science (2017)

Useful for high energies, to get ion track, PKA distribution, ...

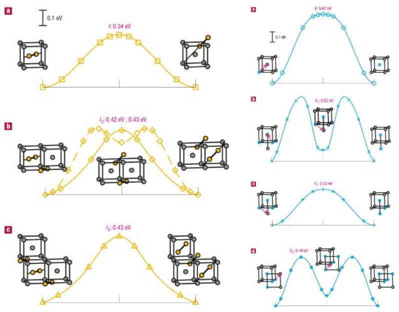
Models to study microstructure evolution
Kinetic Monte Carlo & Cluster Dynamics

Multiscale modeling is needed to understand radiation damage

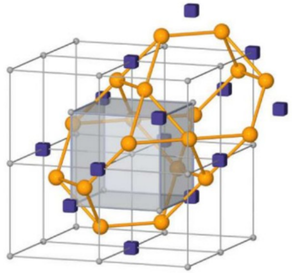


Linking simulation methods to expand time and length scales

DFT + MD → OKMC/CD → Experiment

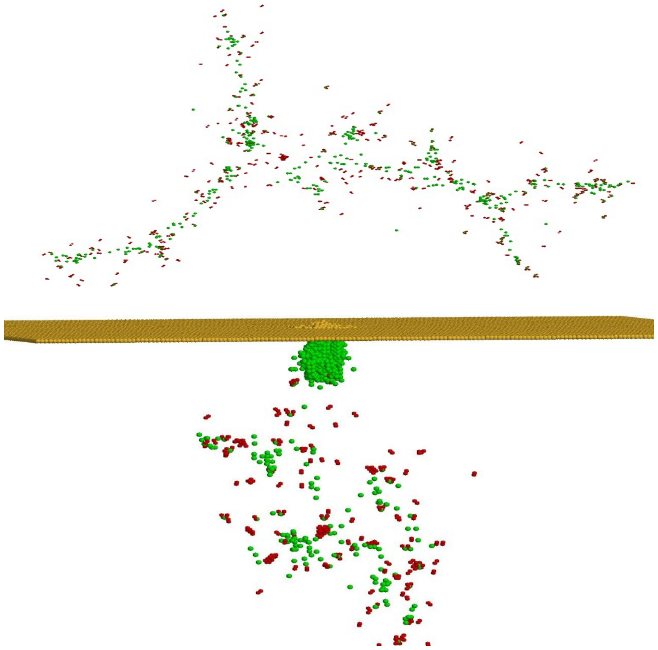


Fu, CC. et al., Nature Mater 4 (2005) 68-74

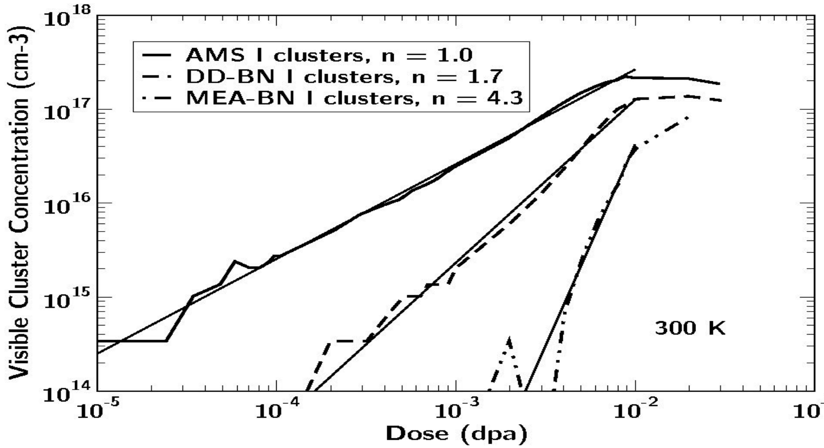


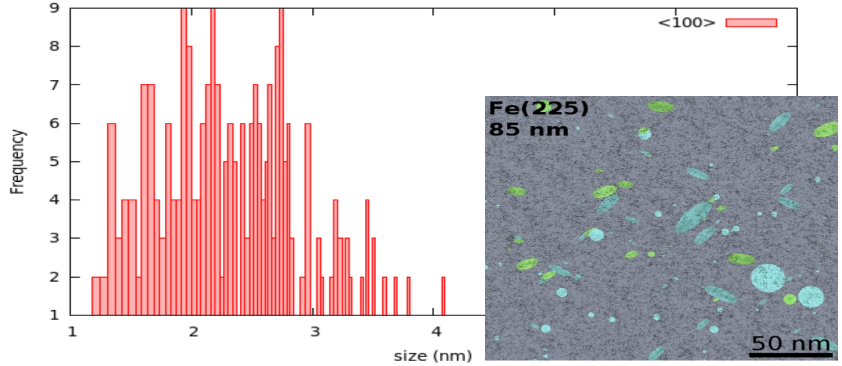
Marinica, M.C. et al., Phys. Rev. Lett. 108 (2012) 025501

100 keV Fe in bulk and surface



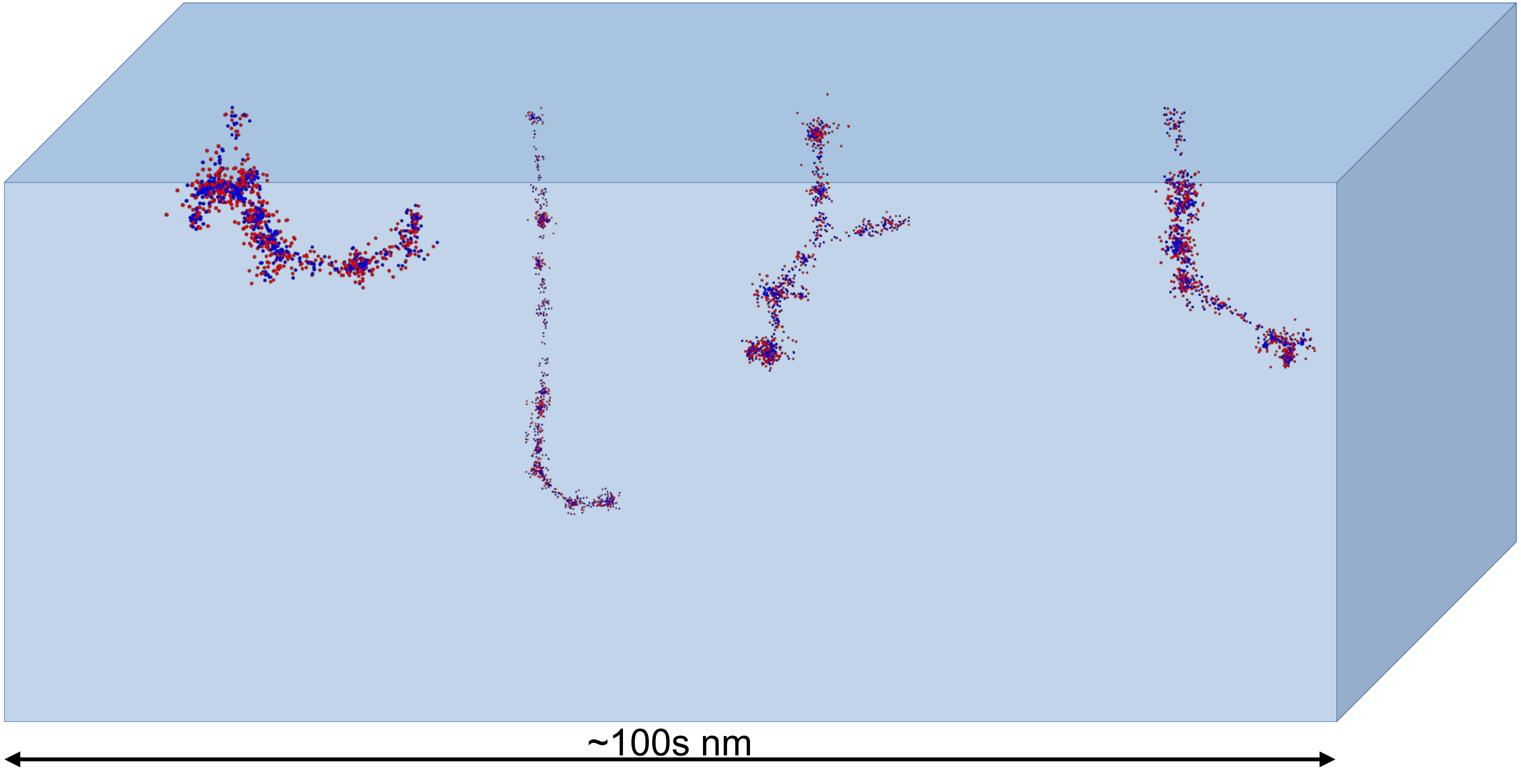
Aliaga, M. J. et al., Acta Materialia 101 (2015) 22-30





Simulation of continuous radiation

Time in between cascades according to experimental conditions

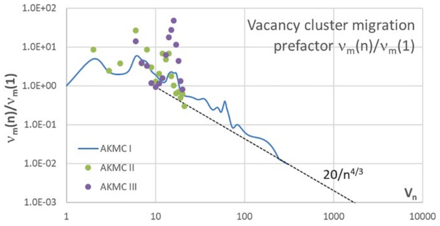


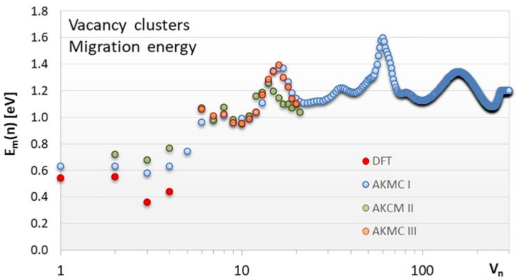
200 keV Fe in Fe cascades with the method presented afterwards

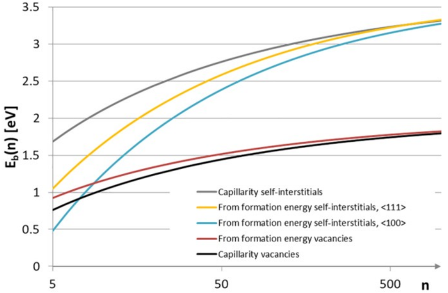
Input parameters

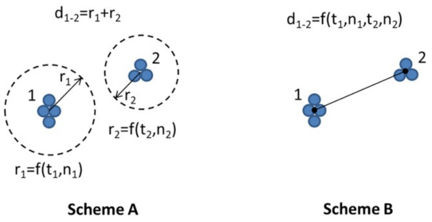
Diffusivity/ migration frequency | Migration energy | Binding energy | Capture radius

VAC

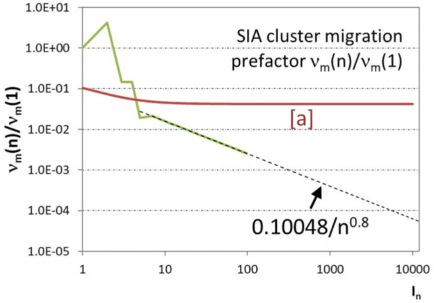


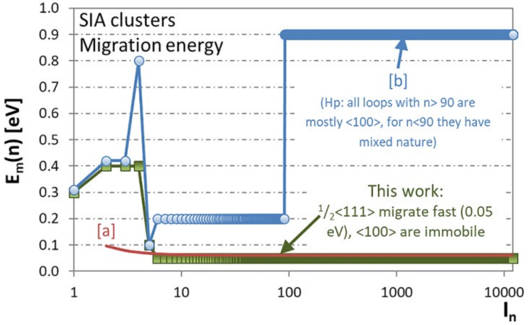


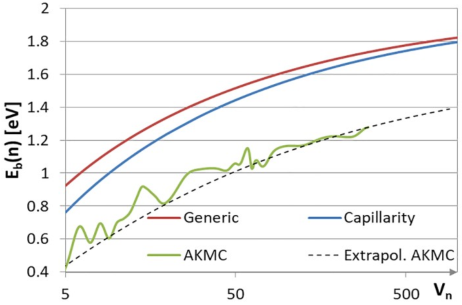


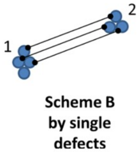


SIA









Malerba, L. et al., Nuclear Materials and Energy 29 (2021) 101069

Combinatorial explosion for impurities (C) and alloys (FeCr, ...)

Kinetic Monte Carlo algorithm

Input parameters:

- Defect distribution
- Defect jump (migration frequency and energy)
- Cluster dissociation (binding energy)
- New damage (dose rate)

Total rate
$R = sum(R_i N_i)$
N_i particles, R_i rate for event *i*

Pick a random number, *r*, between 0 and *R*

Select an event
$0 \leq r \leq R$

Update time
$\Delta t = - log(r)/R$

Perform event

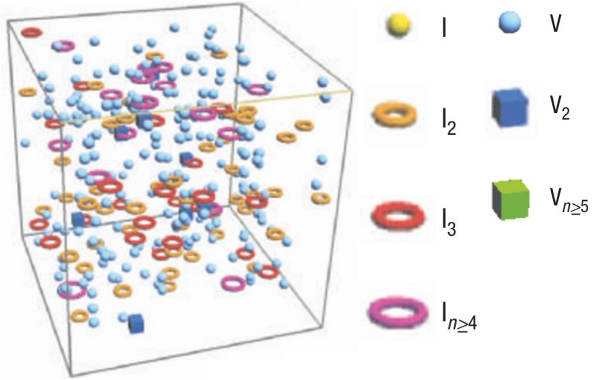
Final time/dose reached?

No

Update rates

Yes

End

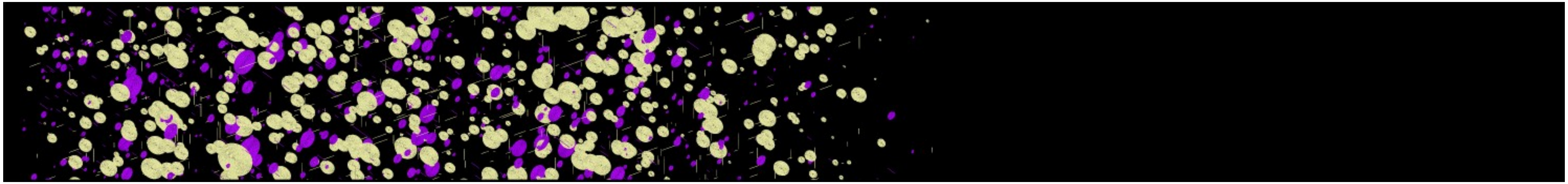


Fu, CC. et al., Nature Mater 4 (2005) 68-74

1 MeV Fe in Fe9Cr at 100ºC [C] = 6 ppm

Juan Pablo Balbuena (Universidad de Alcalá de Henares, Spain), OKMC simulation

Dose $1x10^{18}$ ions/m^2



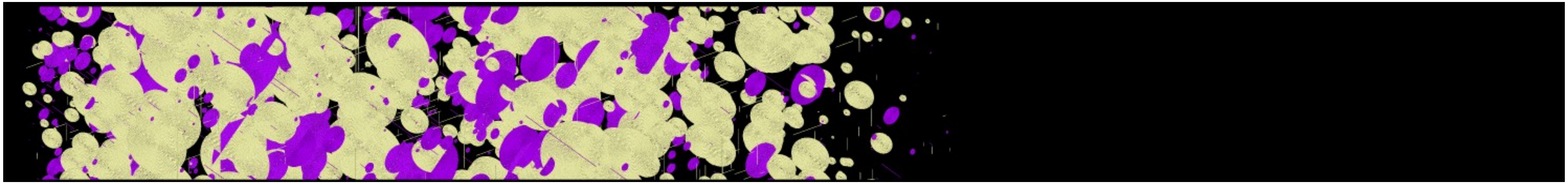
100 nm

900 nm

½<111>

<100>

Dose $5x10^{18}$ ions/m^2

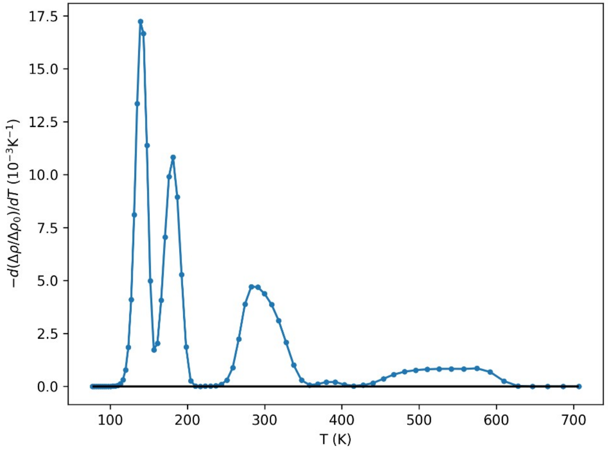


Cluster Dynamics algorithm

Input parameters:
- Defect distribution
- Defect jump (diffusivity, migration energy)
- Cluster dissociation (binding energy)
- New damage (dose rate)

→ Integrate system of equations until given time reached → End

$$
\begin{aligned}
d_t\mathcal{C}_i = \mathcal{I}_i & \\
& + \sum_{\substack{p+q=i \\ p,q\in\mathcal{A}}} \mathcal{T}_{p,q}\mathcal{C}_p\mathcal{C}_q \\
& + \mathcal{E}_{i+1}\mathcal{C}_{i+1} \\
& - \mathcal{C}_i \sum_{\substack{p\neq i \\ p+i\in\mathcal{A}}} \mathcal{T}_{p,i}\mathcal{C}_p - 2\mathcal{T}_{i,i}\mathcal{C}_i^2 \\
& - \mathcal{E}_i\mathcal{C}_i
\end{aligned}
$$



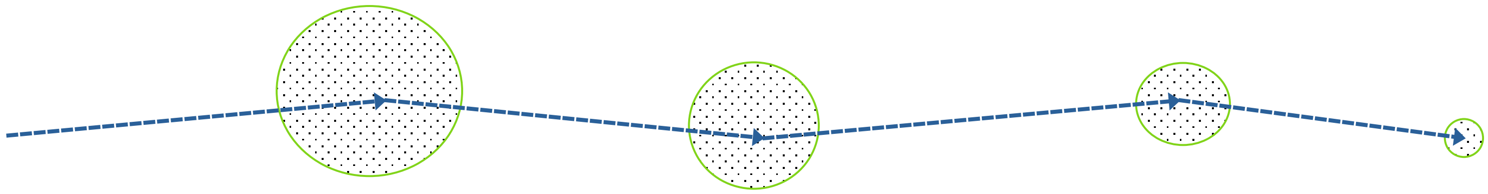
Isochronal annealing of Fe (my work)

abel.gutierrez@ua.es

Our standardised methodology of radiation damage distribution

A simple idea

BCA is a fast alternative to MD, but lacks of recombination and close range interactions
Combine both methods in a simple way



Trajectory and recoil energies from BCA

Debris from MD cascades

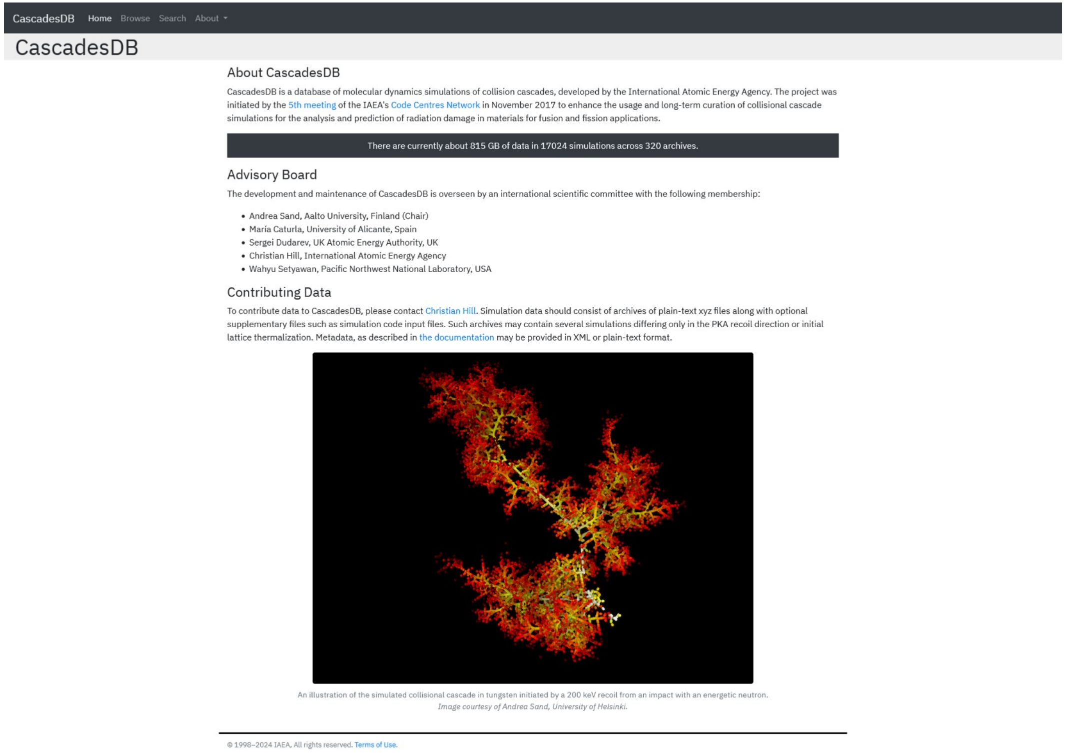
Many details need to be considered

CascadesDB

A freely accessible database of MD simulations.

Samples of W, Fe, Cu, Ni, Pd, Pt
Bulk and surface simulations.
Electronic stopping powers included.

https://cascadesdb.iaea.org/



CascadesDB

Usually, only the final configuration is given.

We created an algorithm to identify defects based on Bhardwaj, U. Computational Materials Science 172 (2020) 109264.

The only input parameter is the number of unit cells in each direction.

You might need to rescale positions if using samples with different lattice parameters.

Not too many energies and samples, but it is an starting point.

Iron samples

Energy (keV)	Number of samples
3	9
5	9
10	19
20	20 (25)
50	12 (13)
100	15
200	4 (5)

SRIM

A free program for the transport of ions in matter

The Stopping and Range of Ions in Matter (SRIM).

http://www.srim.org/

A file called COLLISON.txt contains the ion's trajectory and recoil energies. Use in full-calculation mode.

Energy decomposition

It is very unlikely that a given E_{PKA} will be exactly in our database

For each E_{PKA} the following cascade decomposition is proposed: $E_{PKA} = \sum_{i=1}^{N} n_i E_i^{MD} + \Delta E$

to project the energy into the "base" of available energies. The residual energy, *ΔE*, is converted into damage energy using the Lindhard formula and then it is introduced in the fer-arc-dpa equation.

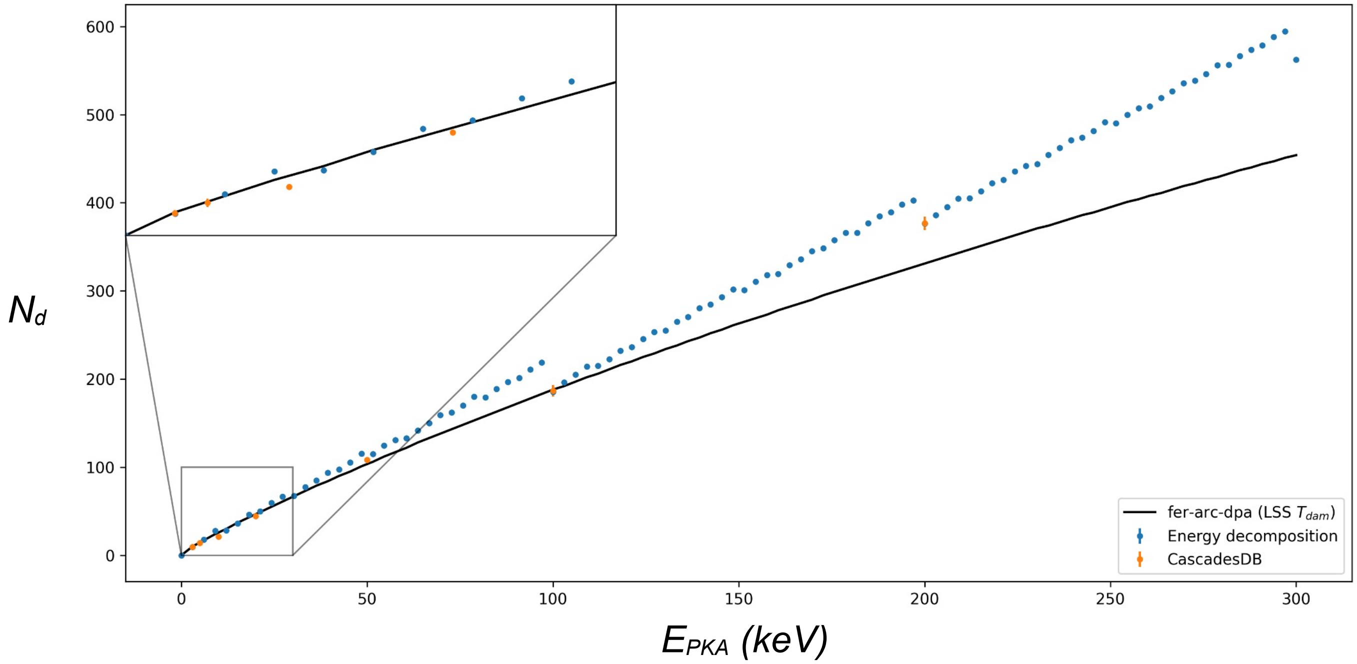
$$N_{d,\text{fer-arc-dpa}} = \begin{cases} 0, & T_{dam} < E_d^{min} \\ \frac{\kappa T_{dam}}{2E_d^{avr}}, & E_d^{min} \leq T_{dam} \leq 2E_d^{avr}/\kappa \\ \frac{\kappa T_{dam}}{2E_d^{avr}} \xi_{\text{arc-dpa}}\left(T_{dam}\right), & T_{dam} > 2E_d^{avr}/\kappa \end{cases}$$

It includes recombination and low energy corrections. ξ ~ 30% at high energies.

fer-arc-dpa: Yang, Q. and Olsson, P., Phys. Rev. Materials 5 (2021) 073602
arc-dpa: Nordlund, K. et al., Nature Communications 9 (2018) 1084
NRT-dpa: Norgett, M. J. et al., Nuclear Engineering and Design 33 (1975) 50-54
Lindhard: Lindhard, J. et al., Mat. Fys. Medd. Dan. Vid. Selsk. 33 (1963)

34 keV = 1x20 + 1x10 + 1x3 + 1x1 keV

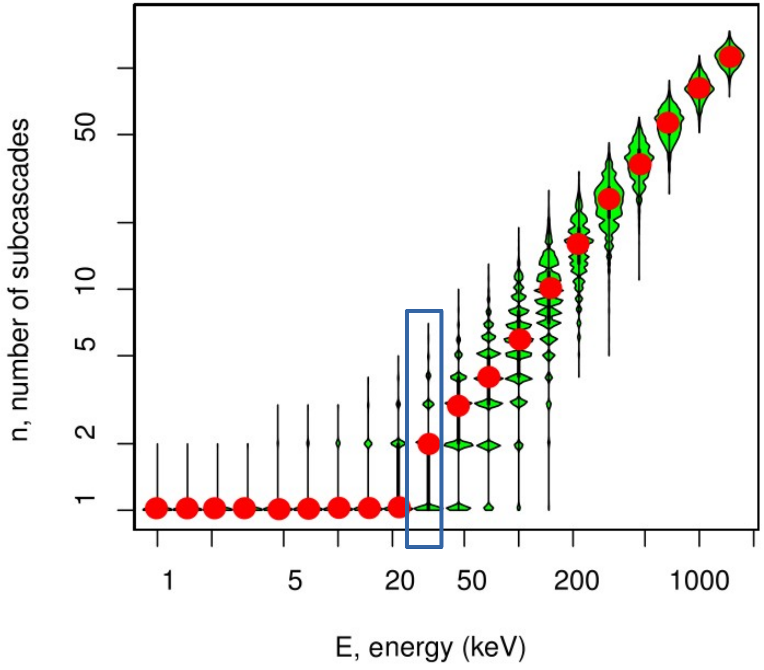
Energy decomposition in Fe



The energy decomposition is fairly valid until ~50 keV (we need a more complete database)

At high energies artificial clusters are introduced

Running SRIM iteratively



At 35 keV, it is expected that cascades split into subcascades in Fe.

We run SRIM iteratively until all PKA energies are less or equal to 35 keV. Use `pysrim` for that.

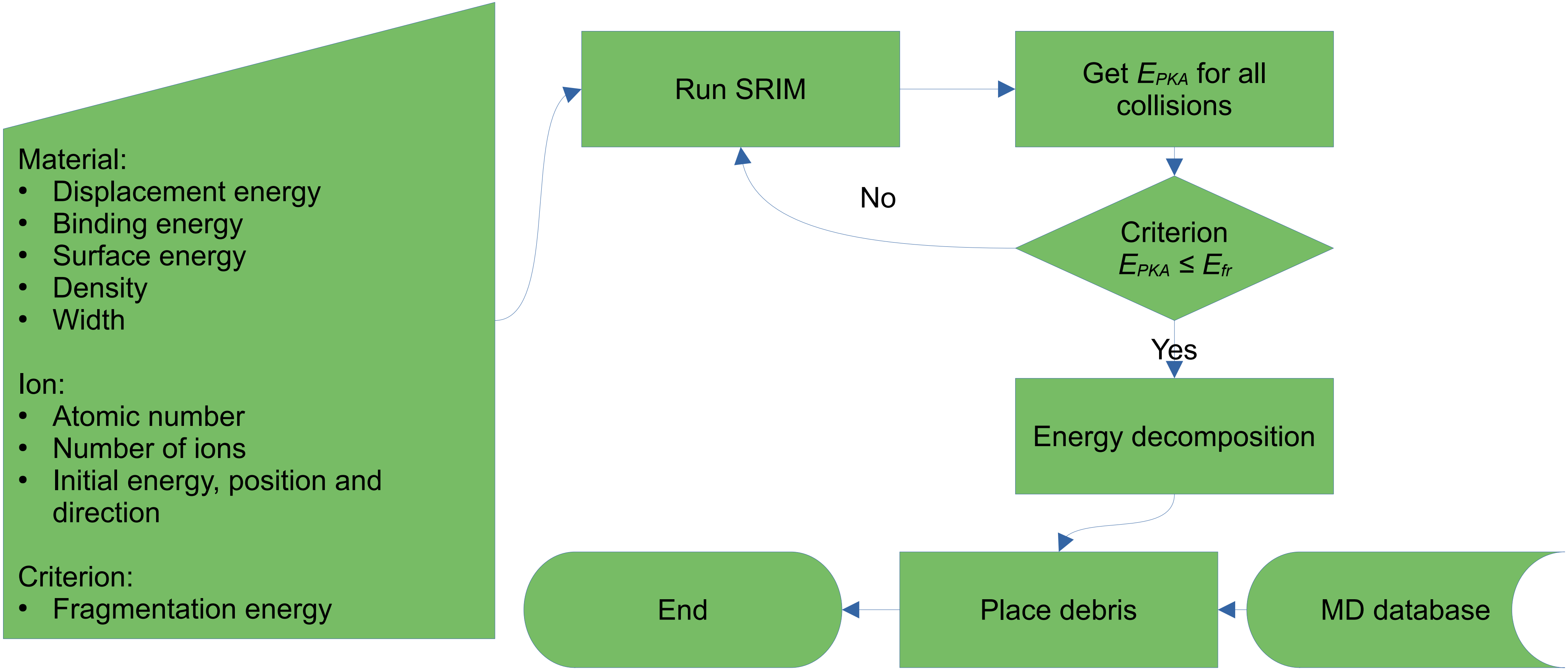
https://pypi.org/project/pysrim

De Backer, A. et al., J. Phys.: Condens. Matter 30 (2018) 405701

We need less energetic MD simulations
Less artificial clusters

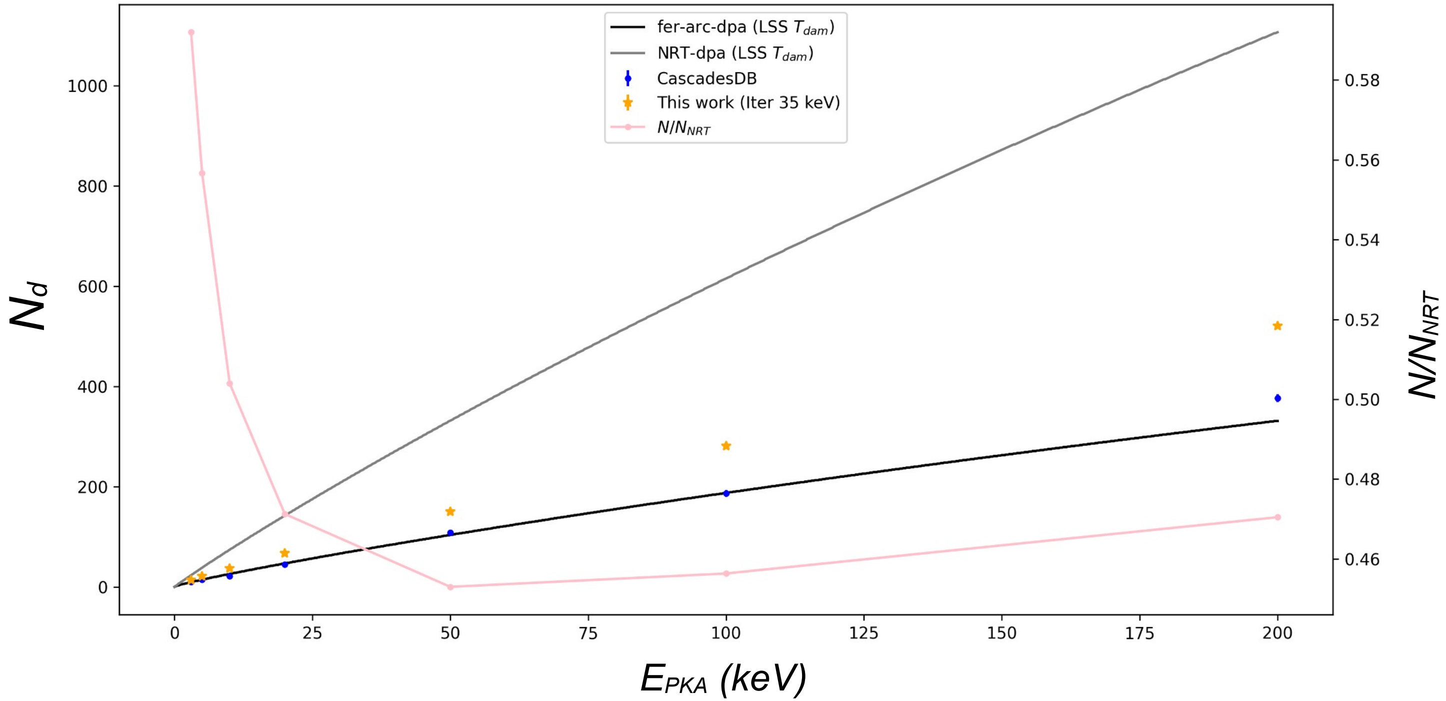
Running SRIM iteratively up to low energies reduces recombination, increasing damage

Algorithm



Material:
• Displacement energy
• Binding energy
• Surface energy
• Density
• Width
Ion:
• Atomic number
• Number of ions
• Initial energy, position and direction
Criterion:
• Fragmentation energy
Run SRIM
Get E_{PKA} for all collisions
No
Criterion $E_{PKA} \leq E_{fr}$
Yes
Energy decomposition
End
Place debris
MD database

fer-arc-dpa?



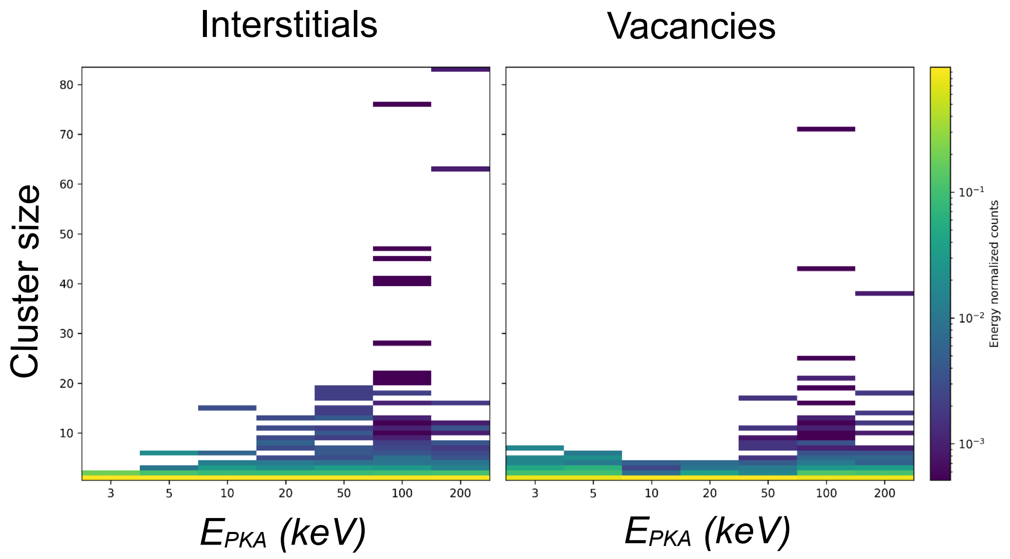
An improvement over NRT-dpa
We obtain cluster distributions, not only N_d

Still not fer-arc-dpa

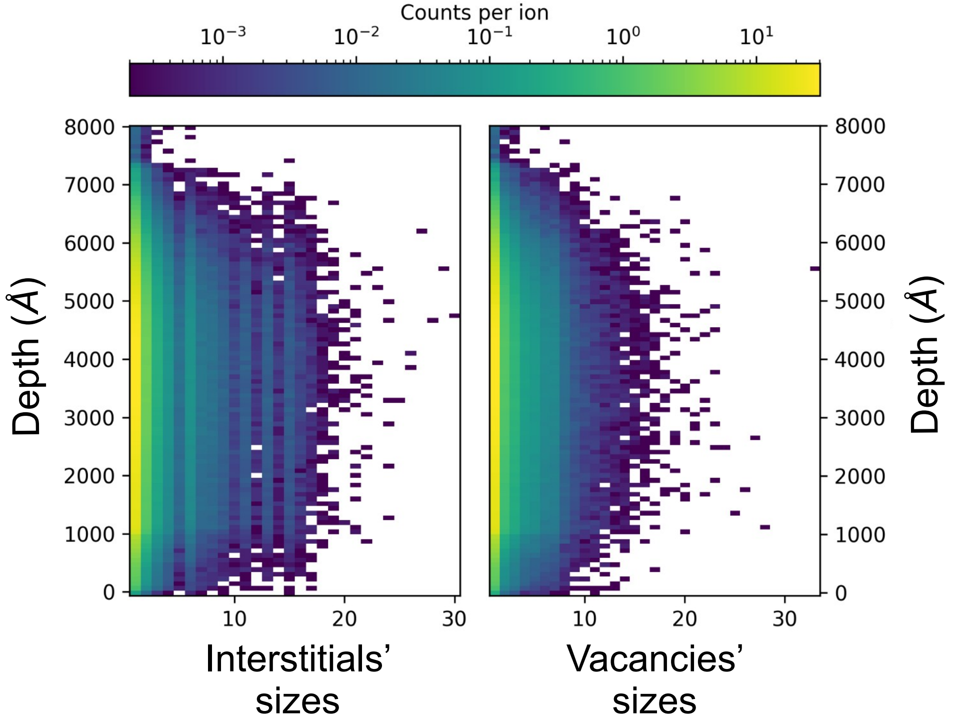
abel.gutierrez@ua.es

1 MeV Fe in Fe (bulk)

5000 ions at 1000 Å



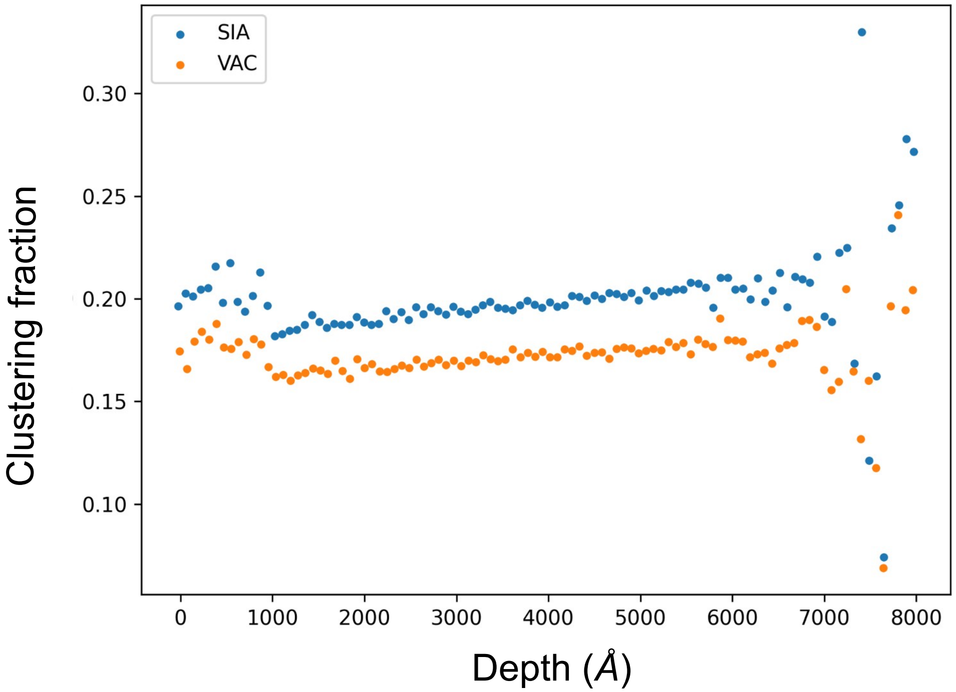
Database distribution



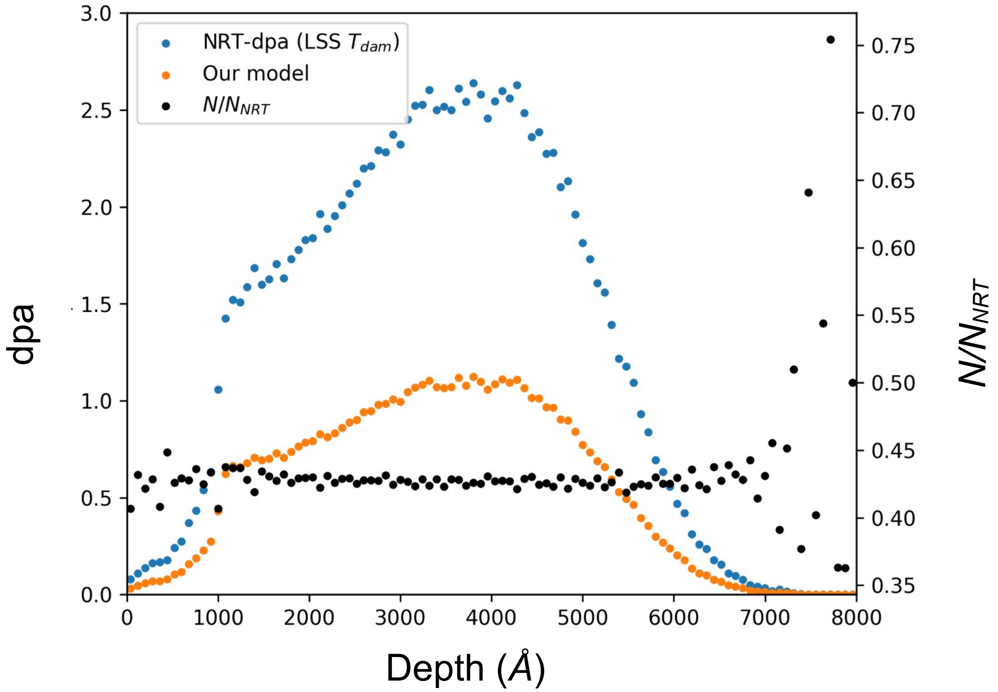
Cluster size – depth distribution

1 MeV Fe in Fe (bulk)

5000 ions at 1000 Å



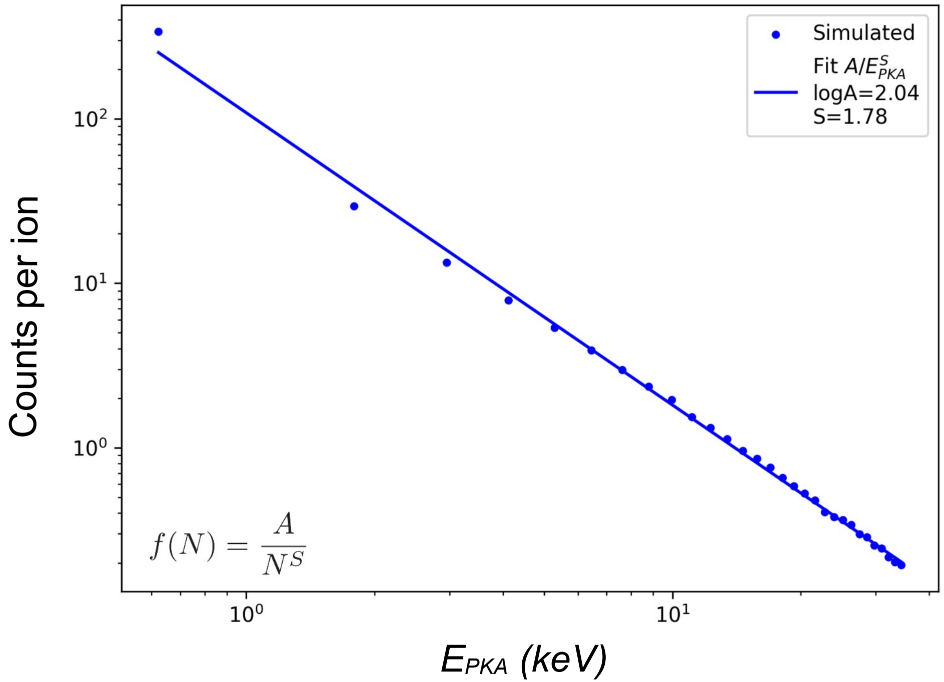
Clustering fraction – depth distribution



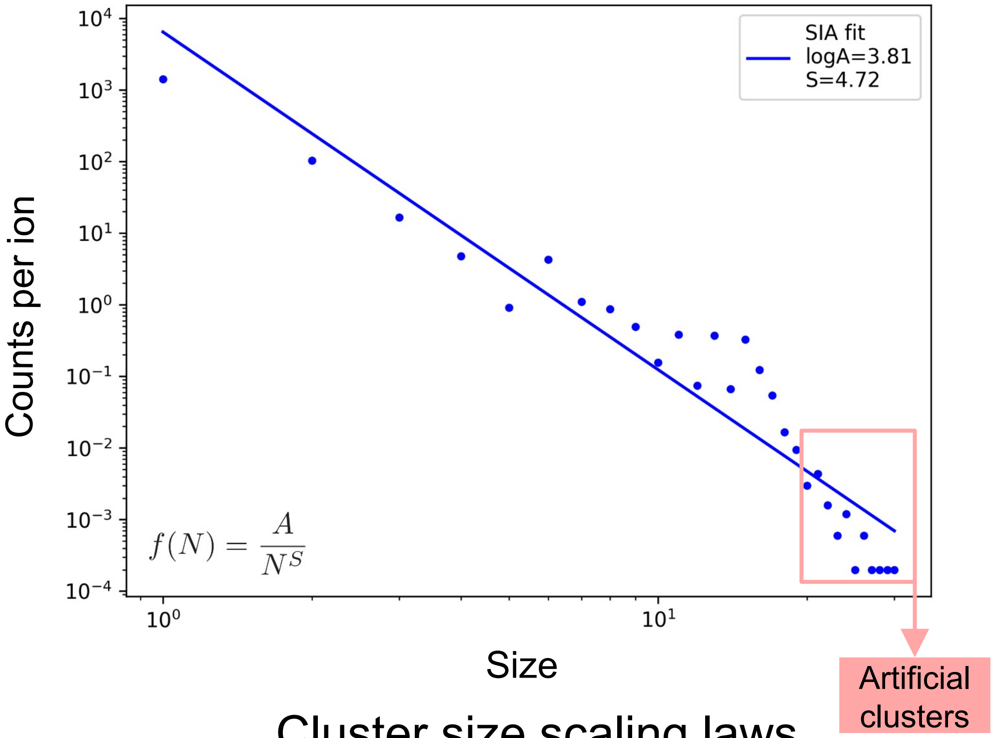
dpa – depth distribution

1 MeV Fe in Fe (bulk)

5000 ions at 1000 Å



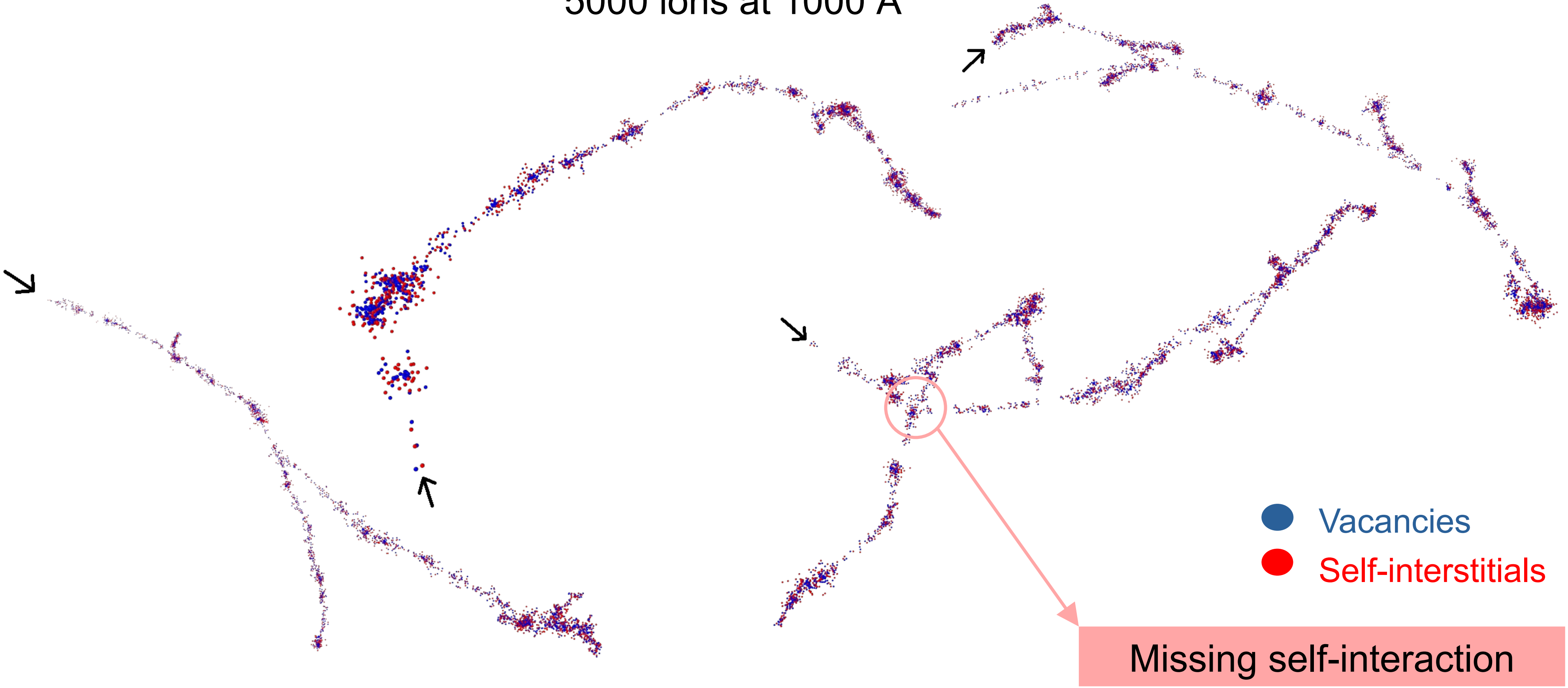
E_{PKA} scaling law



Cluster size scaling laws

1 MeV Fe in Fe (bulk)

5000 ions at 1000 Å



Conclusions

Conclusions and future work

Our program:

- Reconstructs the damage produced by highly energetic ions
- Damage between NRT- and fer-arc-dpa
- Provides cluster size distributions similar to the MD database
- Will be available for the community

In progress:

- Andrea Sand (Aalto University) is helping us: database improvements, close PKA merge, remove Lindhard dependency, use full database, tungsten, ...

This could be expanded to (not our focus):

- Neutron irradiation

Acknowledgments & funding

This work has been carried out within the framework of the EUROfusion Consortium, funded by the European Union via the Euratom Research and Training Programme (Grant Agreement No 101052200 – EUROfusion). Views and opinions expressed are however those of the author(s) only and do not necessarily reflect those of the European Union or the European Commission. Neither the European Union nor the European Commission can be held responsible for them.

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