The place of atomic scale modelling of materials in the overall process of modelling in nuclear

Robin Grimes

Imperial College London

General Comments – on the types of simulations reported here

- Computer simulations predict the behaviour of a system subject to the constraints of a mathematical model as implemented in a computer code (verification).
- The mathematical model consists of a set of governing equations (and assumptions) that describe aspects of the real system.
- However, a computer model is necessarily a simplification of the real system.
- The result of a simulation can be an identification or understanding of the physics/chemistry (the processes) that underpin the behaviour of the system – if the right questions/model has been chosen (validation).
- Simulation helps us explore possibilities, experiment helps simulators determine if they are exploring in a sensible manner.

Performance Development of HPC

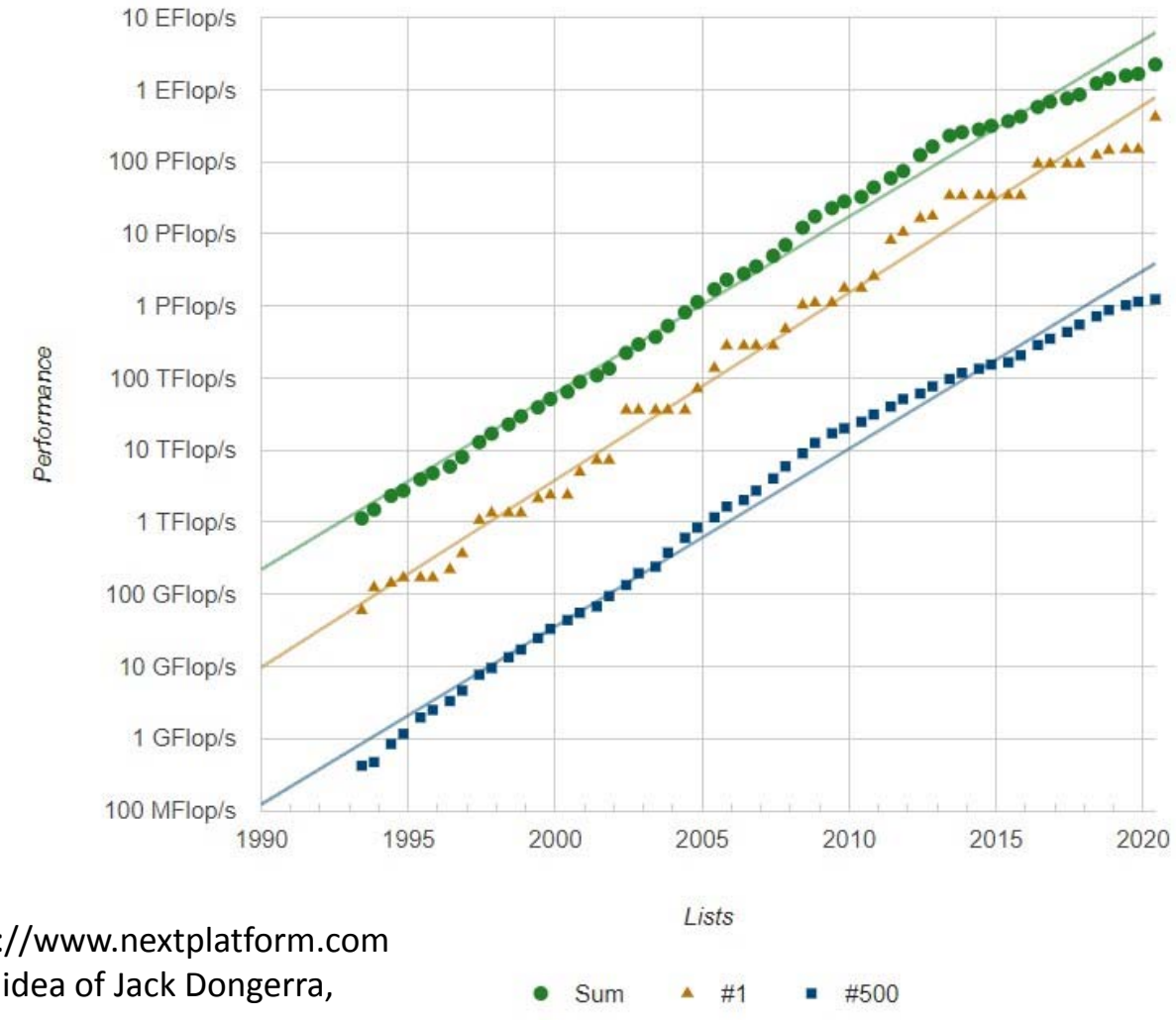


Image from https://www.nextplatform.com
Based on original idea of Jack Dongerra,
U of Tennessee

There are many different tasks for simulation:

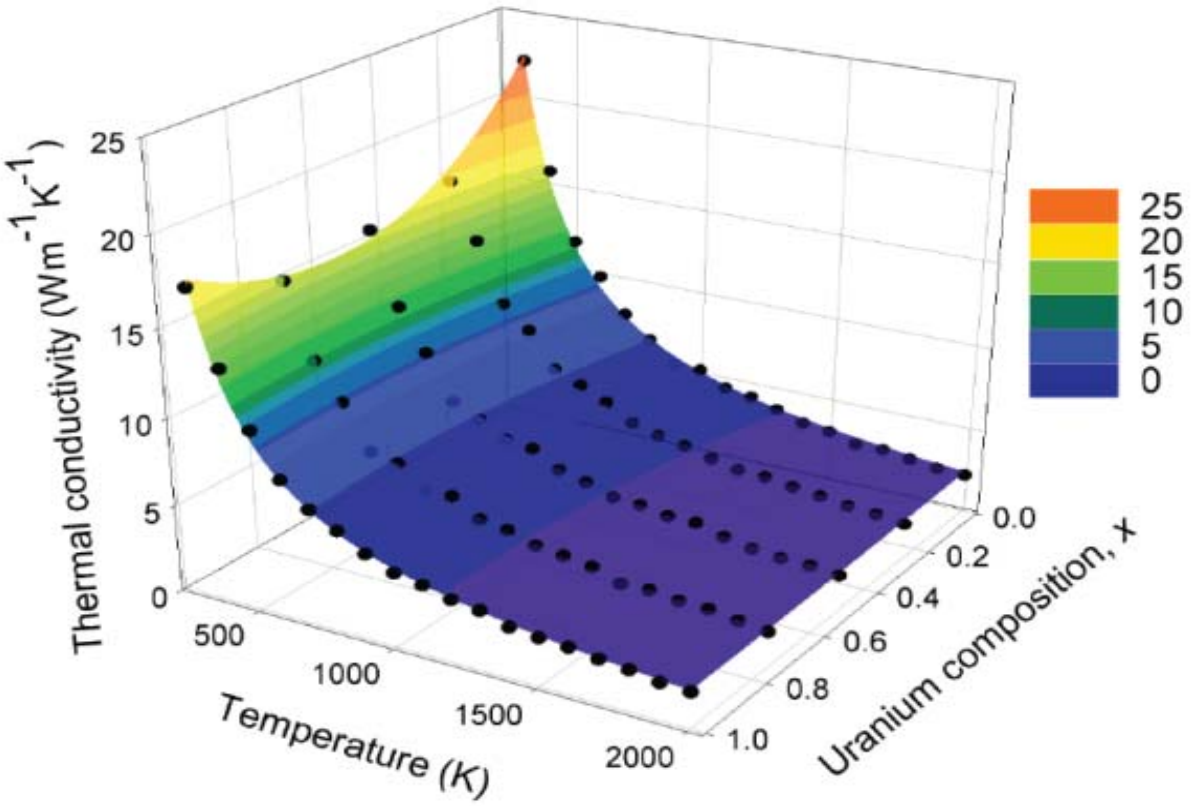
- To provide property values for existing models and further analyze experimental values.
- To ‘check’ or ‘test’ existing assumptions.
- To improve existing models by ‘developing’ the physical models.
- To develop totally new models, using the simulations to ‘discover’ or ‘identify’ the physics/chemistry behind the process.

There are many different tasks for simulation:

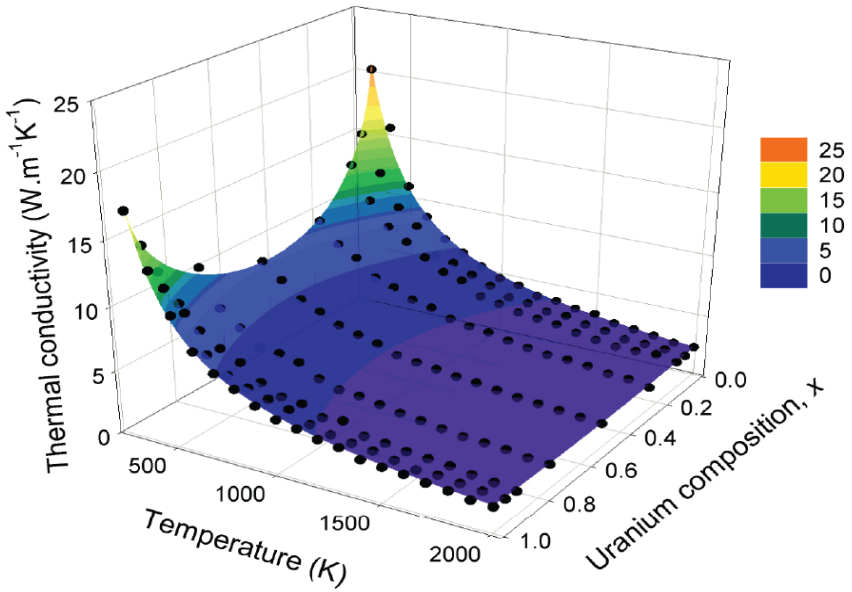
- To provide property values for existing models and further analyze experimental values.
- To ‘check’ or ‘test’ existing assumptions.
- To improve existing models by ‘developing’ the physical models.
- To develop totally new models, using the simulations to ‘discover’ or ‘identify’ the physics/chemistry behind the process.

Compositional dependence of properties

Thermal conductivity as a function of composition & temperature for $(U_xPu_{1-x})O_2$. MD data are shown with black spheres whilst the result of fitting a constitutive equation to this data is indicated by the surface.



Cooper M. W. D., Middleburg S. C., & Grimes R. W. "Modelling the thermal conductivity of $(U_xTh_{1-x})O_2$ and $(U_xPu_{1-x})O_2$" *J. Nucl. Mater.* **466**, 29 (2015).



Prediction of thermal conductivity, k, for $U_xPu_{(1-x)}O_2$

The usual equation used is:

$$k = \frac{1}{a + bT + cx}$$

Where a, b and c are parameters. But this only works for one composition.

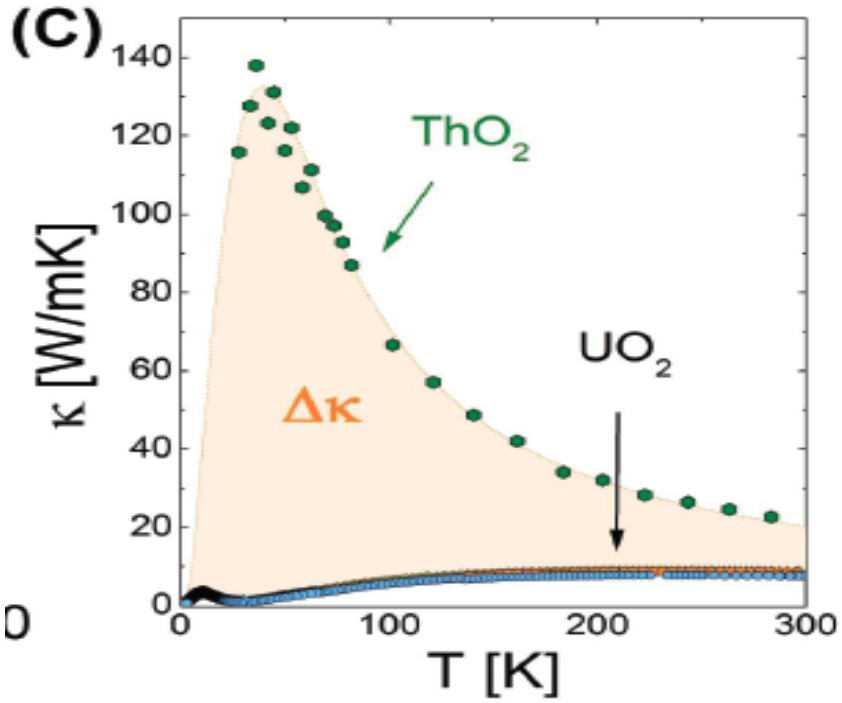
We want an equation that will work for any composition – and that can be extended to work with any number of components in the system.
The modelling gave us the data set to develop the model below:

$$k = \frac{1}{xW_{AC} + (1-x)W_{BC} + x(1-x)C_{AB}}$$

$$W_{AC} = a_{AC} + b_{AC}T$$
$$W_{BC} = a_{BC} + b_{BC}T$$

Experimental data can be used now to tweek the model parameters so we can be sure it reproduces real experimental data and is then really predictive – going well beyond existing experimental data.

UO_2 thermal conductivity behaviour is different compared to chemically and crystallographically similar compounds



K. Gofryk et al. *Nature Comm.* (2014)

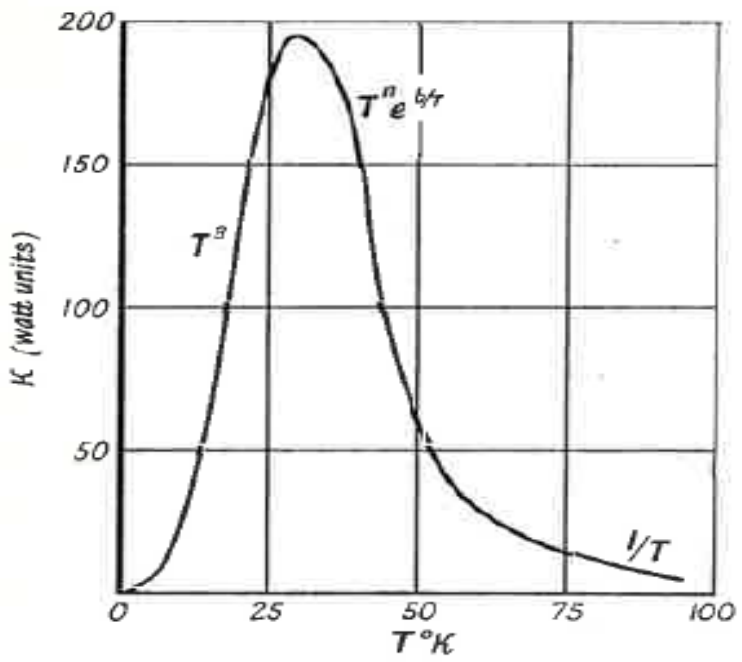


FIG. 83. Thermal conductivity of Al_2O_3 (Berman, 1958).

Phonon Scattering by Magnetic Spins

Below 30 K, UO_2 exhibits AFM ordering and above is paramagnetic. ThO_2 is diamagnetic.

In UO_2 (and not ThO_2) scattering by spin excitations on the uranium ions are responsible for the unusual shape of the thermal conductivity curve.

Scattering between phonons and spins on uranium ions occurs by phonons exciting energy levels of the magnetic ions, see e.g. Van Vleck, *Phys. Rev.* (1940).

The standard Callaway model (*Phys. Rev.* (1959))

$$\kappa = \frac{k_B}{2\pi v}\left(\frac{k_B T}{\hbar}\right)^3 \int_0^{\Theta_D/T} \frac{\tau_p x^4 e^x}{(e^x-1)^2}\,dx$$

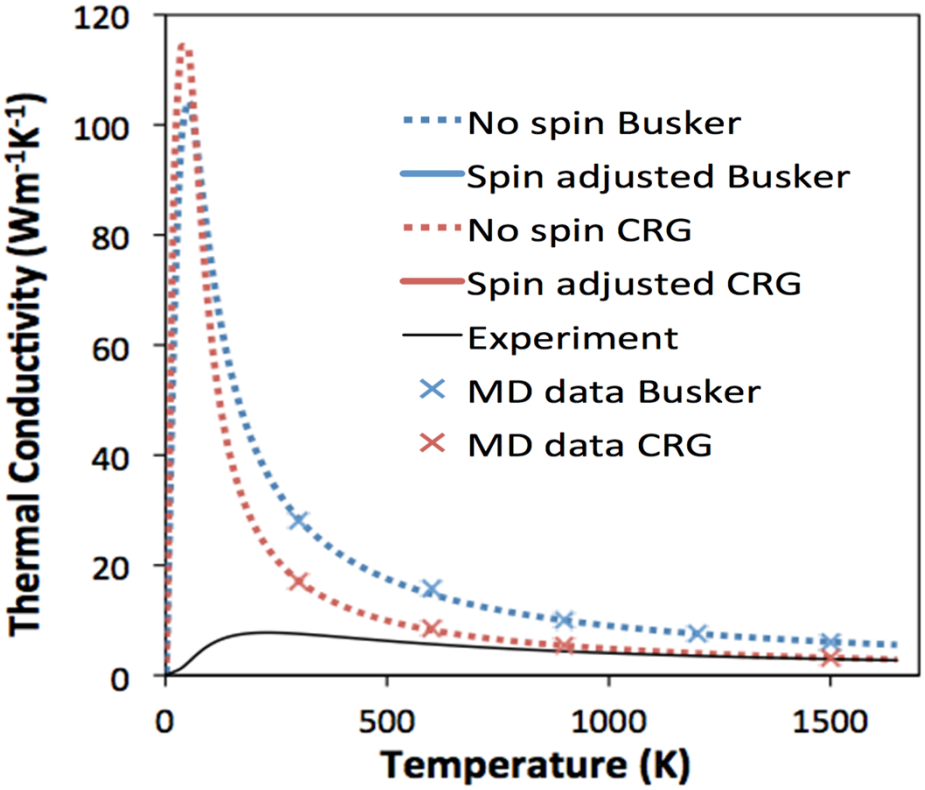
can be extended to include a spin scattering term:

$$\tau_p^{-1} = \tau_D^{-1} + \tau_B^{-1} + \tau_U^{-1} + \tau_S^{-1}$$

$$\tau_S^{-1} = \sum_i \frac{C_i \omega^4}{\left(\omega^2 - \omega_{S,i}^2\right)}$$

Neelmani & Verma, Phys. Rev. B (1972).

Results without spin scattering term



$$\kappa = \frac{k_B}{2\pi v}\left(\frac{k_B T}{\hbar}\right)\int_0^{\Theta_D/T}\frac{\tau_p x^4 e^x}{(e^x-1)^2}dx$$

$$x = \hbar\omega/k_B T$$

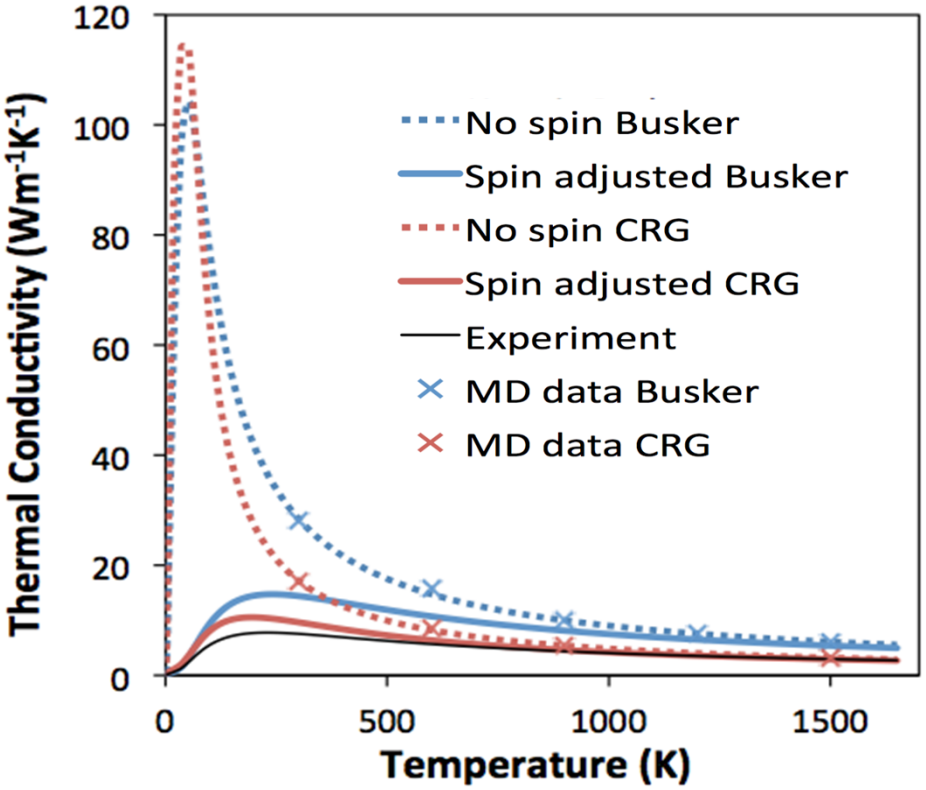
$$\tau_D^{-1} = Dx^4T^4 = D\left(\frac{\hbar\omega}{k_B}\right)^4$$

$$\tau_B^{-1} = B$$

$$\tau_U^{-1} = UT^3x^2e^{-\Theta_D/bT} = UT\left(\frac{\hbar\omega}{k_B}\right)^2 e^{-\Theta_D/bT}$$

$$\tau_S^{-1} = \sum_i \frac{C_i\omega^4}{(\omega^2-\omega_{S,i})}F_i(T)$$

Including spin scattering term



$$\kappa = \frac{k_B}{2\pi v}\left(\frac{k_B T}{\hbar}\right)\int_0^{\Theta_D/T}\frac{\tau_p x^4 e^x}{(e^x-1)^2}dx$$

$$x = \hbar\omega/k_B T$$

$$\tau_D^{-1} = Dx^4T^4 = D\left(\frac{\hbar\omega}{k_B}\right)^4$$

$$\tau_B^{-1} = B$$

$$\tau_U^{-1} = UT^3x^2e^{-\Theta_D/bT} = UT\left(\frac{\hbar\omega}{k_B}\right)^2 e^{-\Theta_D/bT}$$

$$\tau_S^{-1} = \sum_i \frac{C_i\omega^4}{(\omega^2-\omega_{S,i})}F_i(T)$$

Liu et al. Phys Rev Applied **6**, 044015 (2016)

There are many different tasks for simulation:

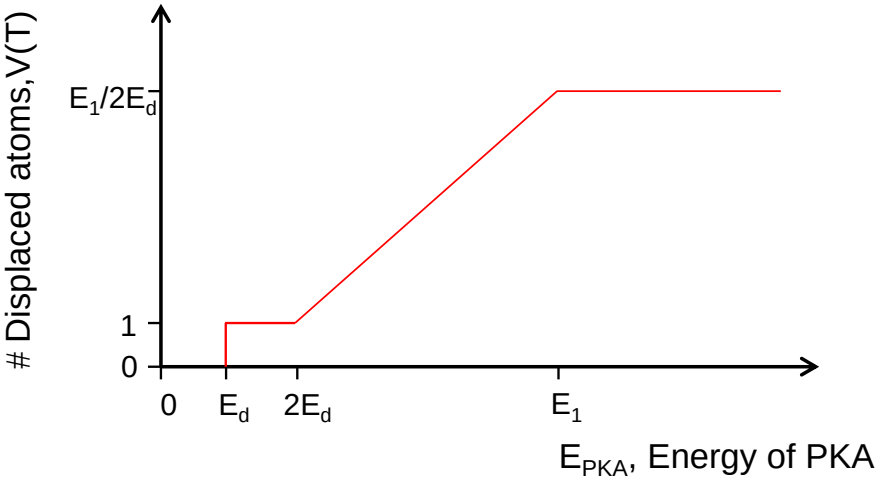
- To provide property values for existing models and further analyze experimental values.
- To ‘check’ or ‘test’ existing assumptions.
- To improve existing models by ‘developing’ the physical models.
- To develop totally new models, using the simulations to ‘discover’ or ‘identify’ the physics/chemistry behind the process.

The Kinchin-Pease Model

The displacement cross-section: $\sigma_d(E_i) = \int_{E_{\min}}^{E_{\max}} \sigma_d(E_i, E_{PKA})\, \nu(E_{PKA})\, dE_{PKA}$

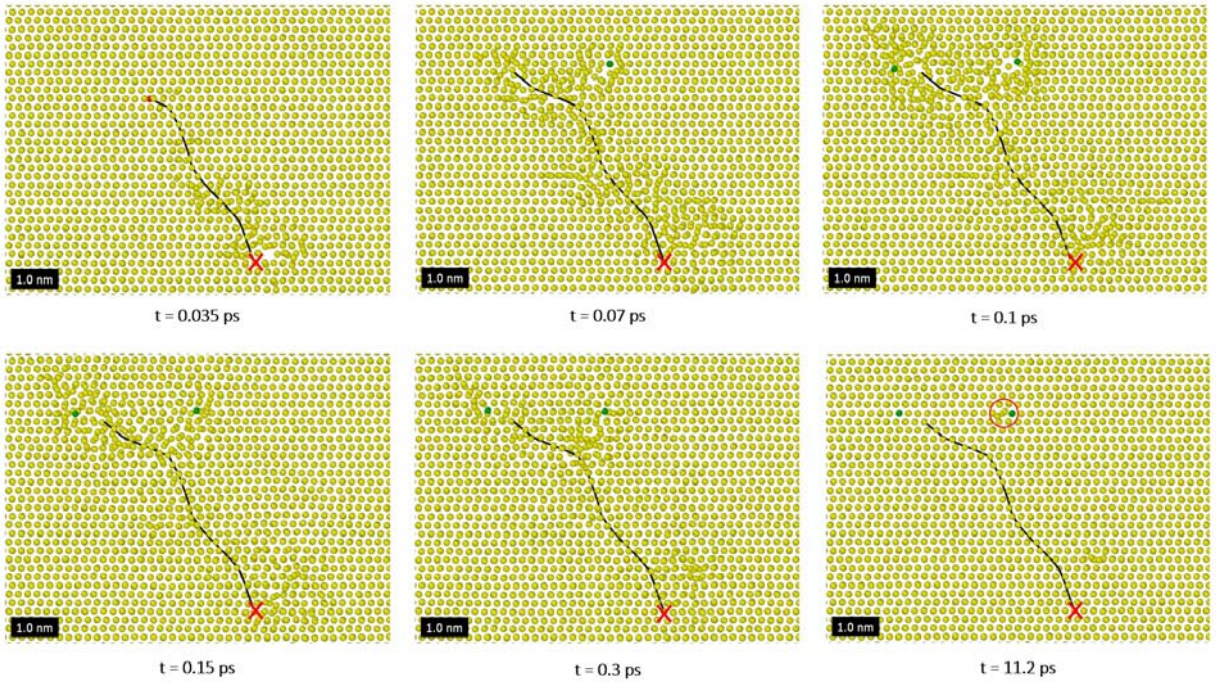
The number of displaced atoms, $\upsilon(E_{PKA})$, per PKA can be estimated through the K-P model, where:

0	$0 < E_{PKA} < E_d$
1	$E_d < E_{PKA} <$
$2E_d$	
$E/2E_d$	$2E_d < E_{PKA} < E_1$
$E_1/2E_d$	$E_1 < E_{PKA} < \infty$



E_d is the critical energy for displacement

Cascade Progression in Be



5-atom thick slice through the simulation cell

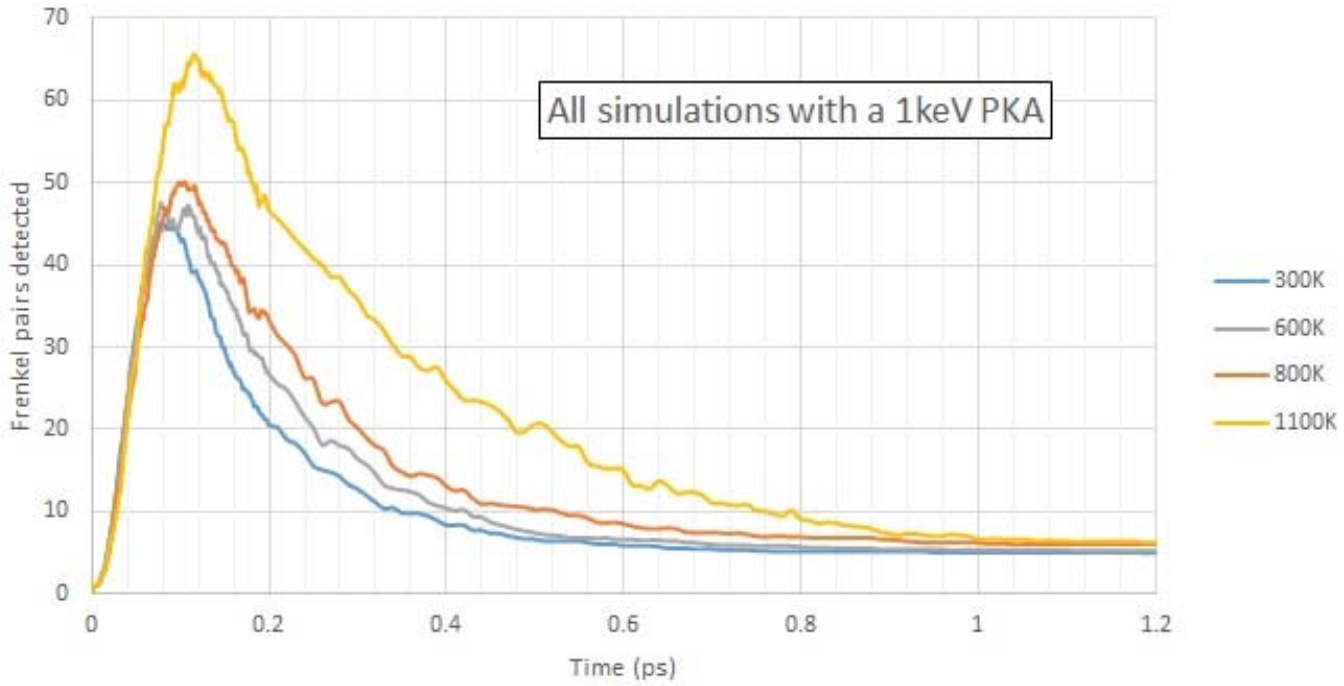
X starting point

Black line is the trajectory

Red circle a residual defect >6 Å displaced

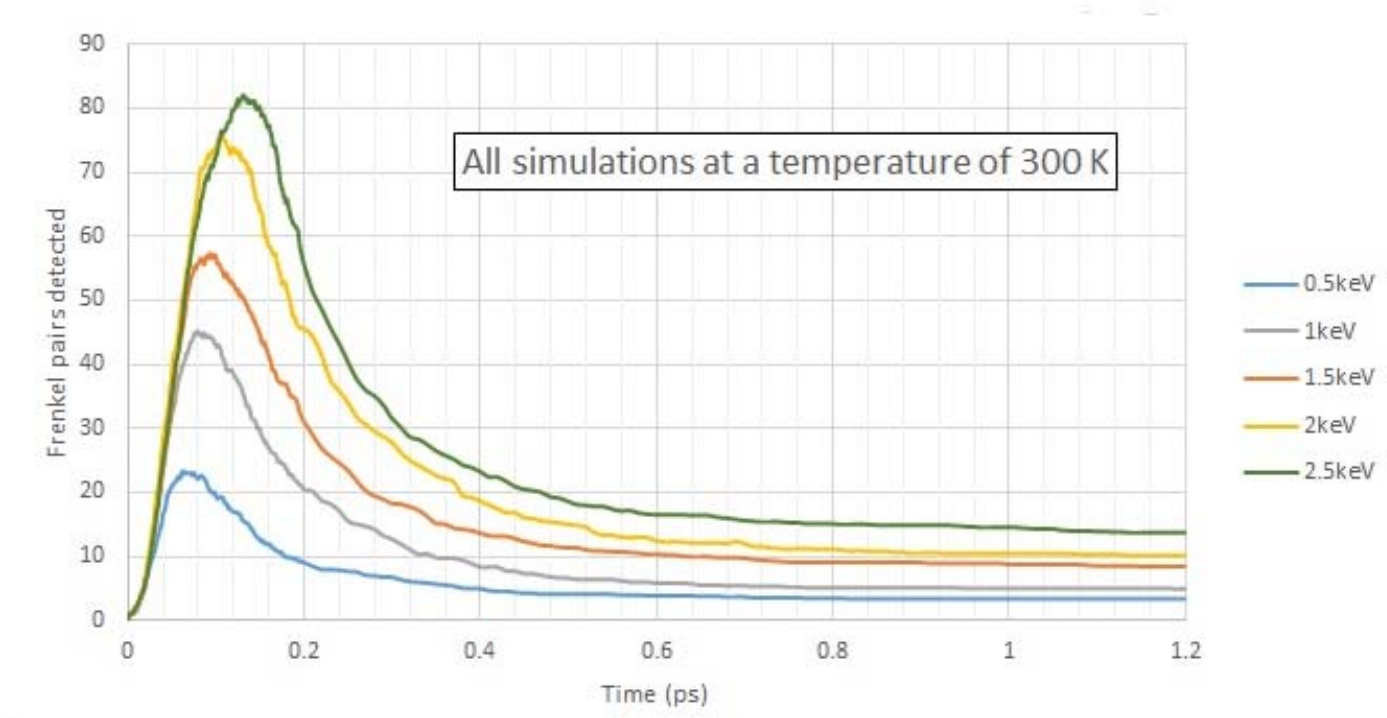
- Distorted region formed along PKA trajectory = initial damage
- Most damage recovers to leave a few residual defects

Frenkel pairs as f^n of simulation time



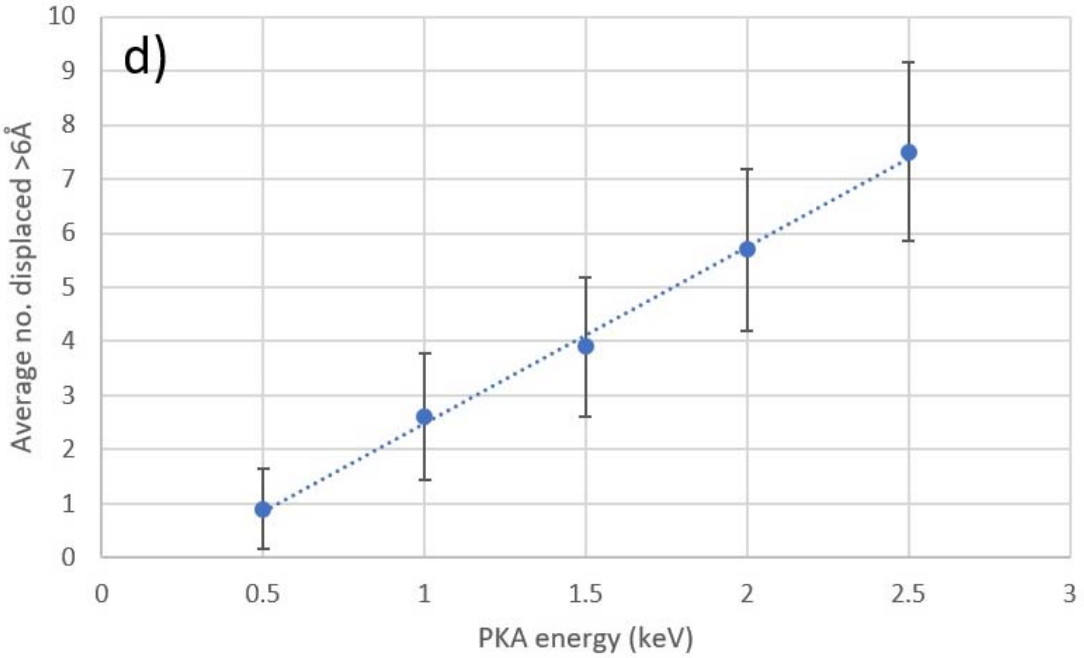
- >90% of damage recovers by 1 ps then steady to 11.2 ps (ie end of sim)
- Residual damage not a strong function of temperature
- Residual damage >> equilibrium [Frenkel] (@1100 K $x10^{-7}$)

Frenkel pairs as f^n of E_{PKA}



- Maximum damage and time to maximum damage increase with E_{PKA}
- Residual damage is a strong function of temperature (K-P model)

Atoms displaced (υ) throughout the simulation

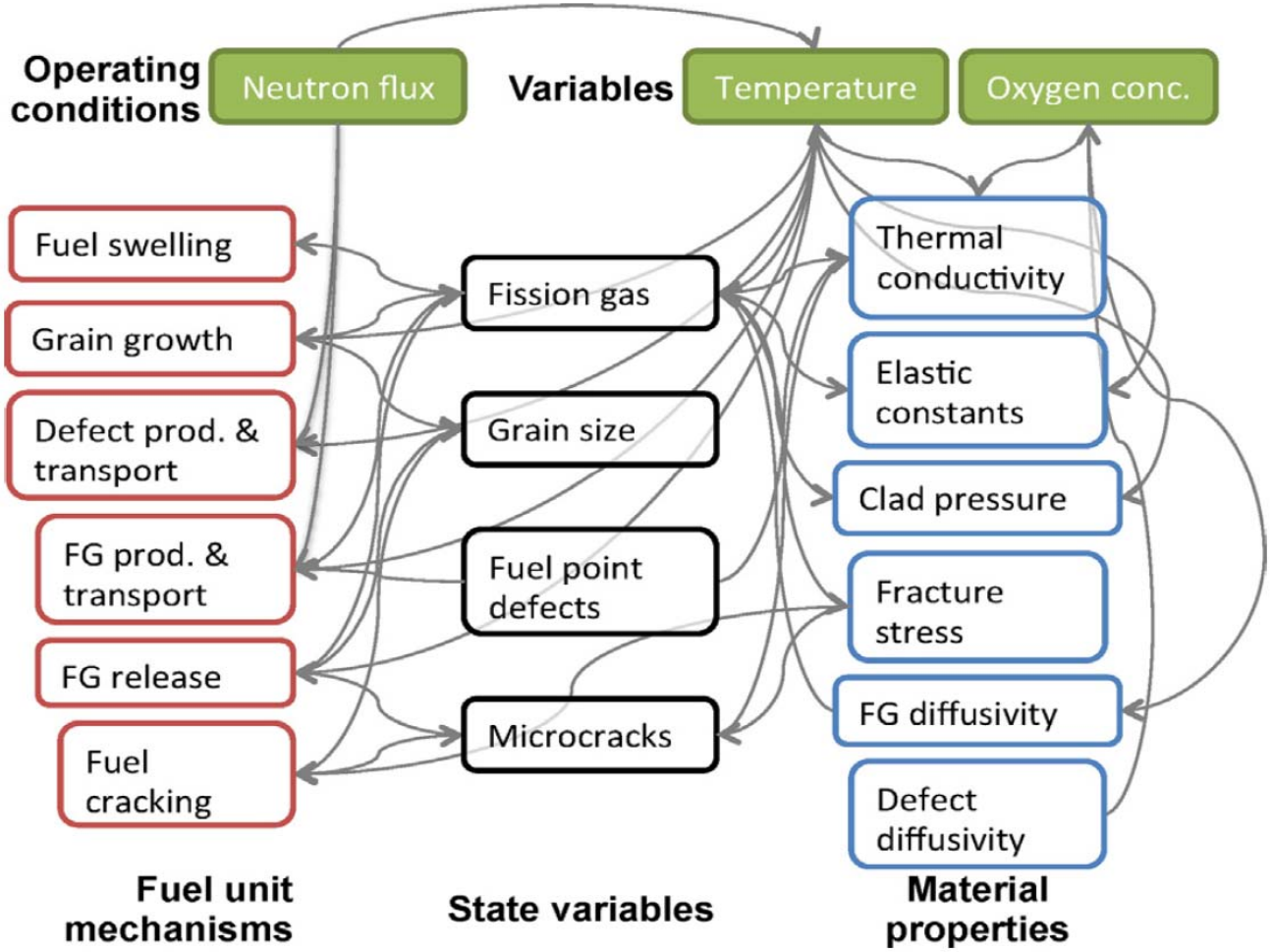


- # atoms displaced beyond 6 Å increases linearly with E_{PKA}
- …for this light metal

There are many different tasks for simulation:

- To provide property values for existing models and further analyze experimental values.
- To ‘check’ or ‘test’ existing assumptions.
- To improve existing models by ‘developing’ the physical models.
- To develop totally new models, using the simulations to ‘discover’ or ‘identify’ the physics/chemistry behind the process.

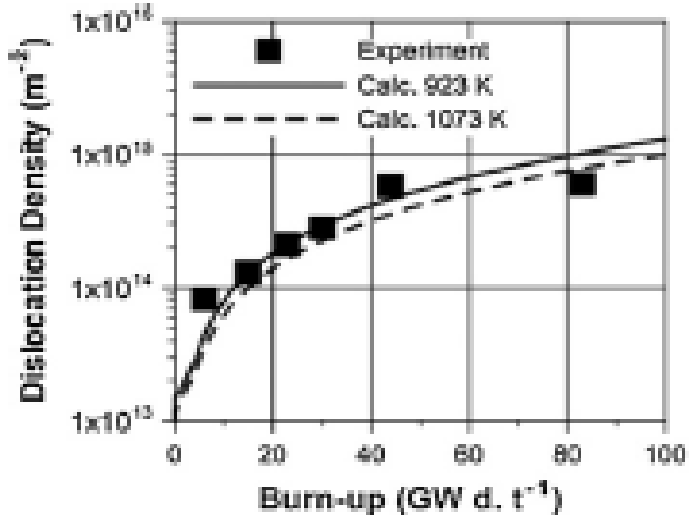
State variable approach for fission gas



Tonks et al. “Mechanistic materials modeling for nuclear fuel performance” Annals of Nuclear Energy 105 (2017) 11–24

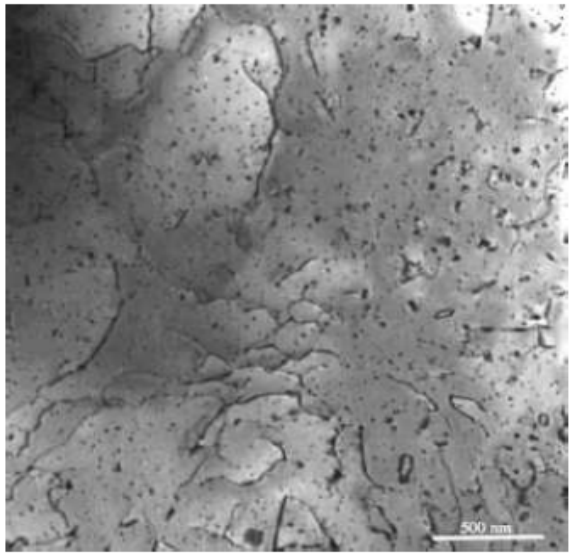
Dislocations

- One of the consequences of irradiation is the formation of dislocations.
- Dislocations can have a major impact on mechanical properties and also thermal conductivity.

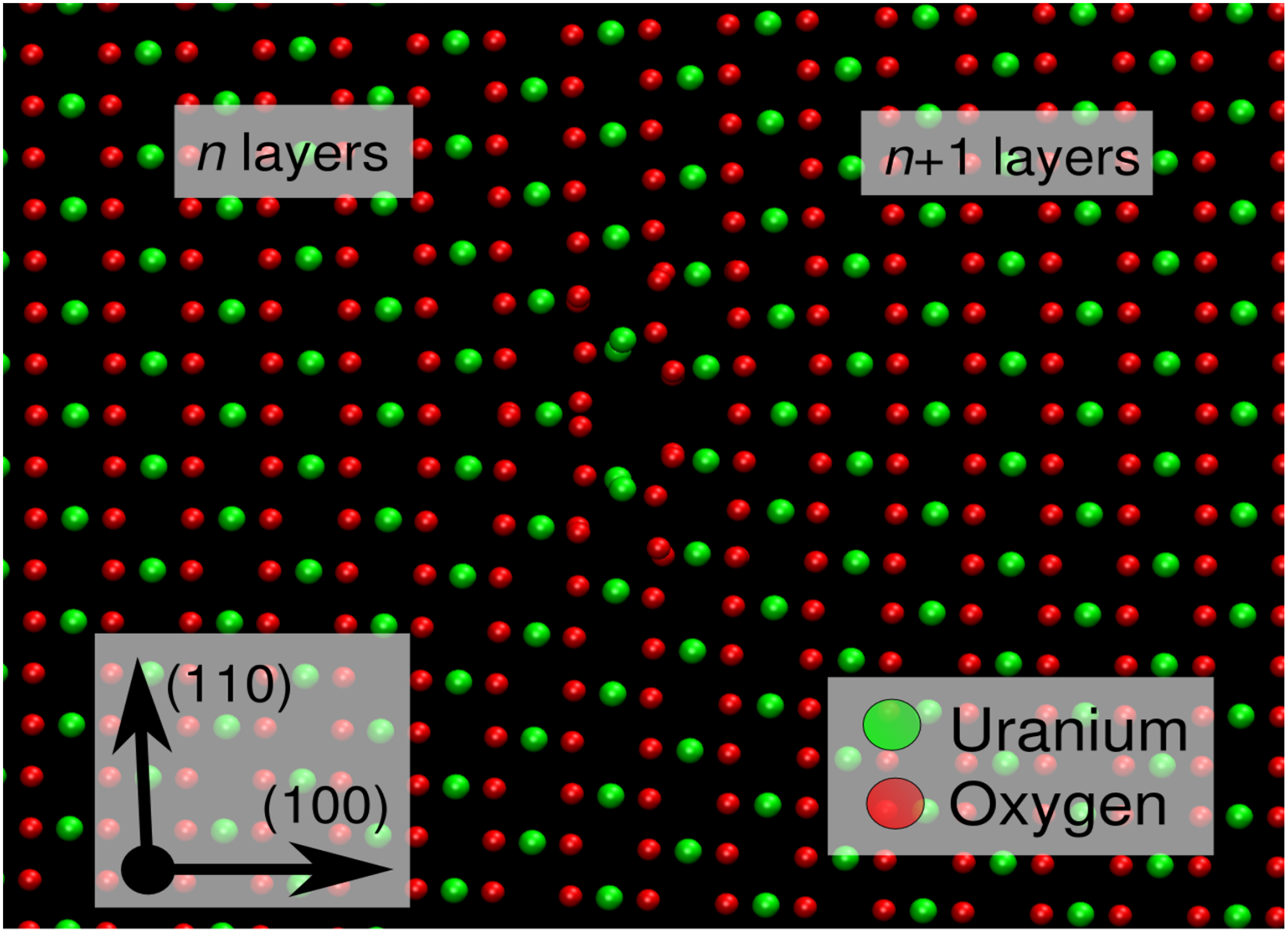


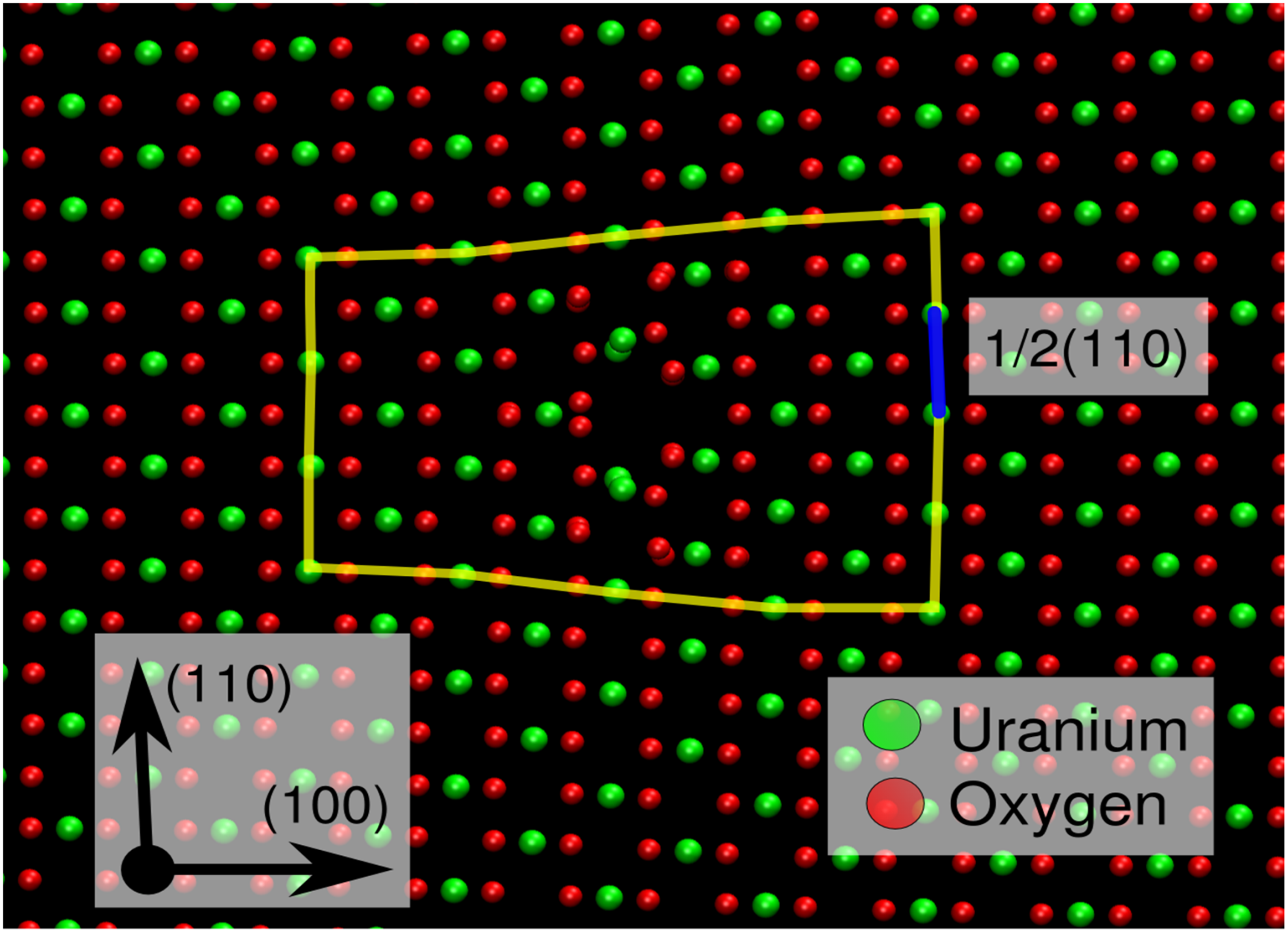
Graph showing the increase in dislocation density as a function of burn-up.

Micrograph showing dislocations in nuclear fuel (EDF energy).



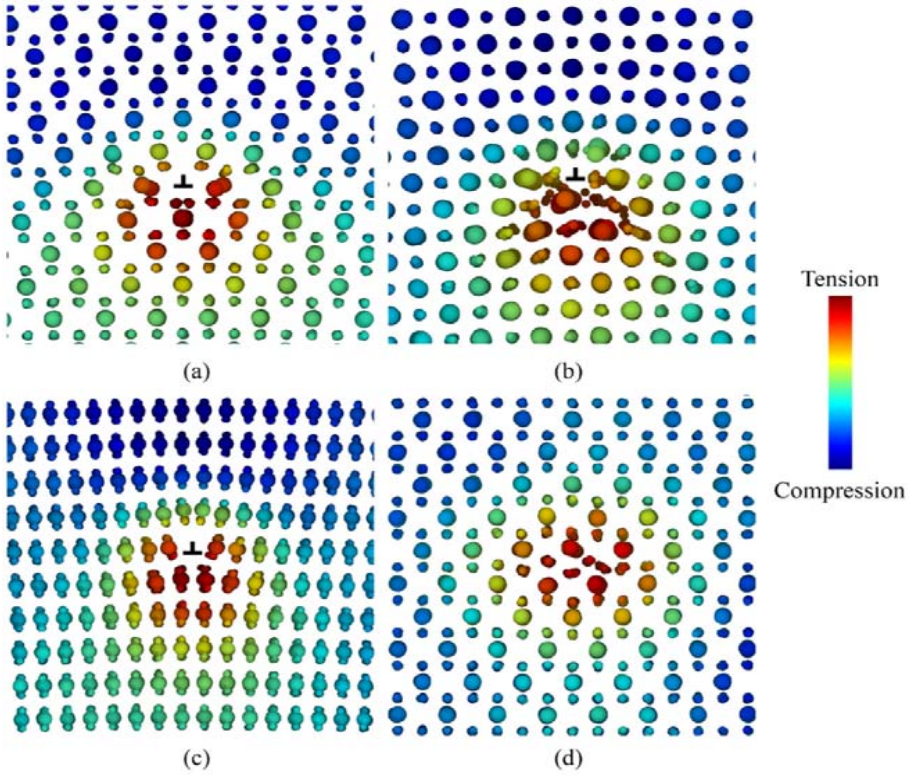
- Dislocations also provide pathways for enhanced migration i.e. pipe diffusion.


n layers
n+1 layers
(110)
(100)
Uranium
Oxygen


1/2(110)
(110)
(100)
Uranium
Oxygen

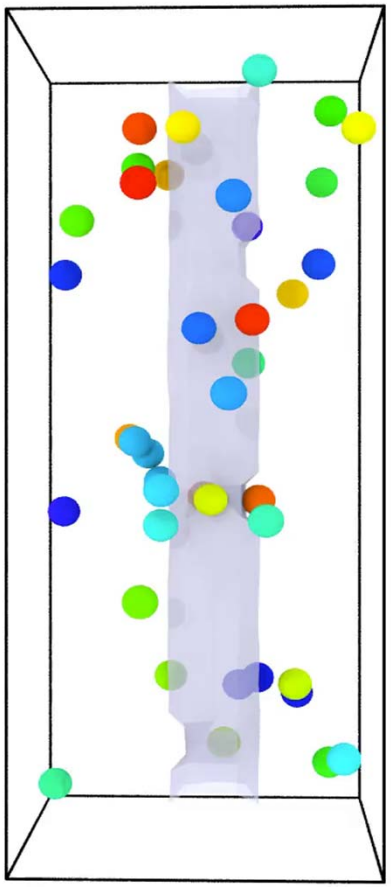
MD Dislocation Modelling in UO_2

- Focus on the interaction of Xe, Kr & He with dislocations as gas
- Dislocation core structures at 300K: a) {100}<110>, b) {110}<110>, c) {111}<110>, d) <110> screw.
- He transport greatly enhanced in all dislocation core regions.



Galvin et al., "Pipe and Grain Boundary Diffusion of He in UO_2" *J. Phys.: Condens. Matter,* **28,** 405002 (2016).

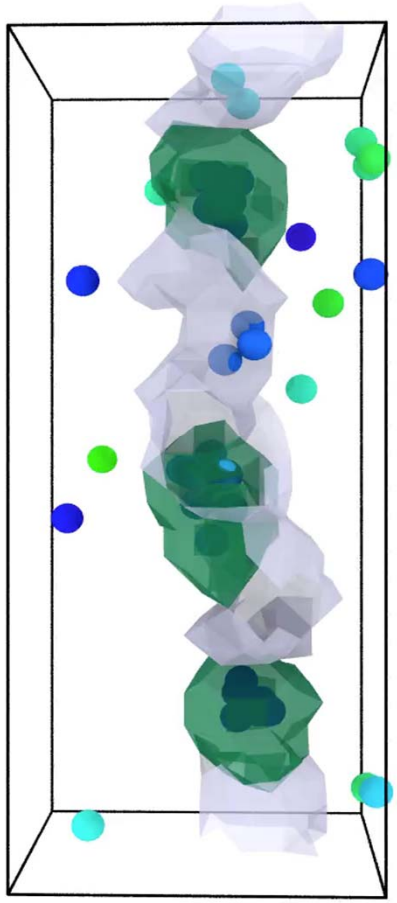
Pipe diffusion of Xe



Initial configuration

Color: cluster analysis
(37 clusters detected)

Pipe diffusion of Xe



Final configuration

Color: cluster analysis
(19 clusters detected)

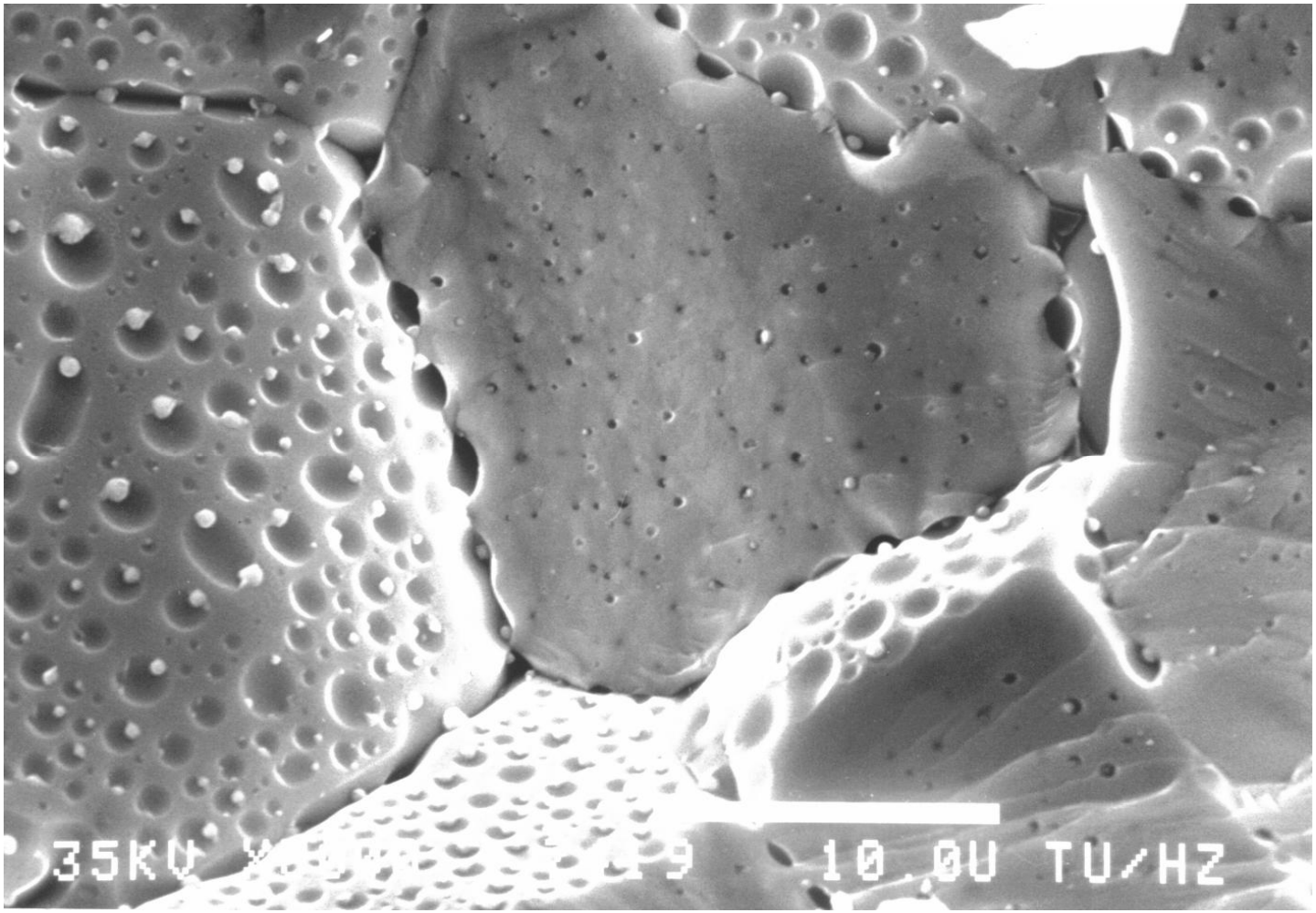
- **Pipe diffusion facilitates clustering trapping Xe**
- **Dislocations might enable longer-distance diffusion between bubbles (coarsening)**

Murphy S. T., Fossati P. & Grimes R. W. "Xe diffusion and bubble nucleation around edge dislocations in UO_2" *J. Nucl. Mater.* **466**, 634 (2015).

There are many different tasks for simulation:

- To provide property values for existing models and further analyze experimental values.
- To ‘check’ or ‘test’ existing assumptions.
- To improve existing models by ‘developing’ the physical models.
- To develop totally new models, using the simulations to ‘discover’ or ‘identify’ the physics/chemistry behind the process.

Intergranular and Transgranular Fracture

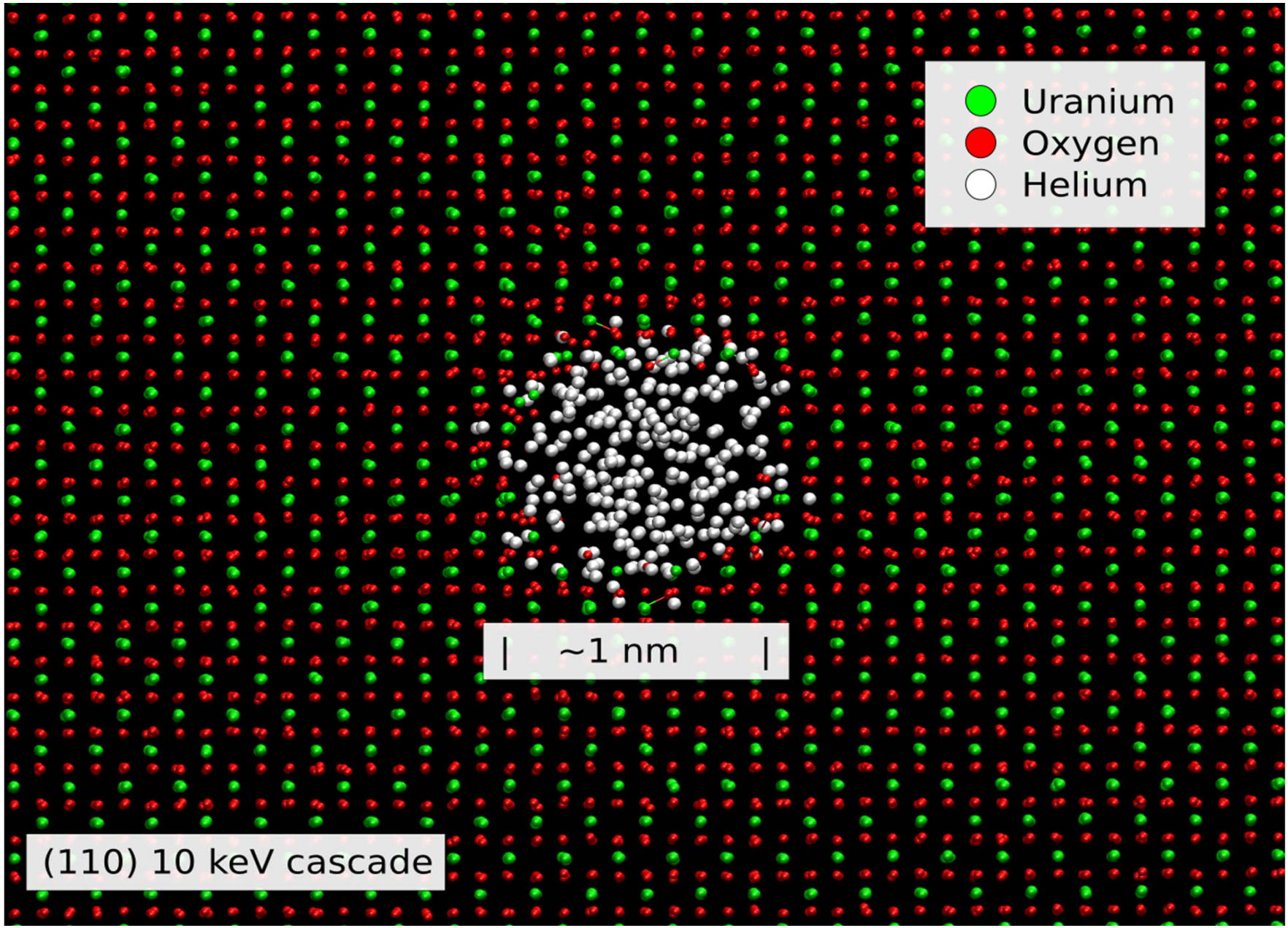


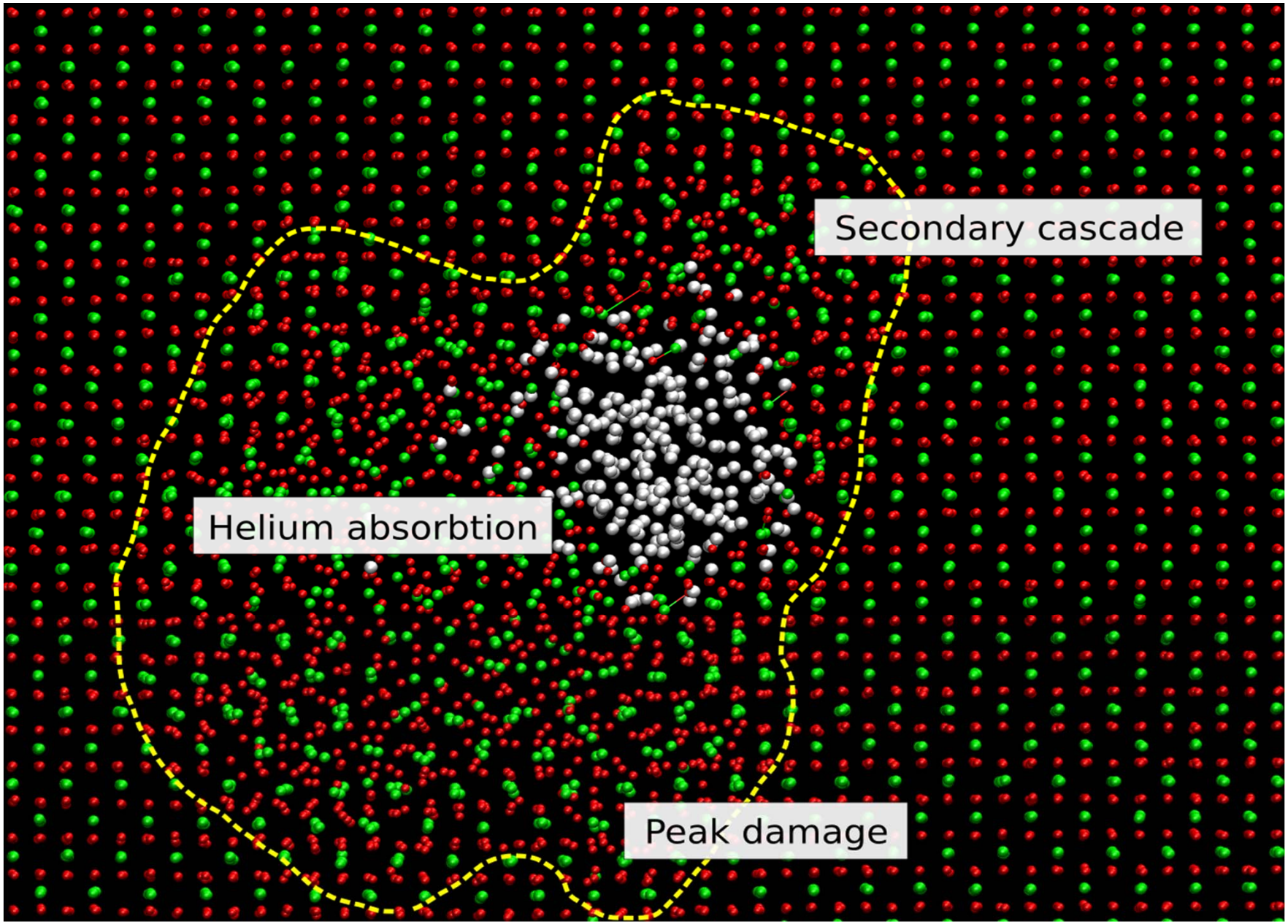
Molecular dynamics of radiation enhanced helium re-solution

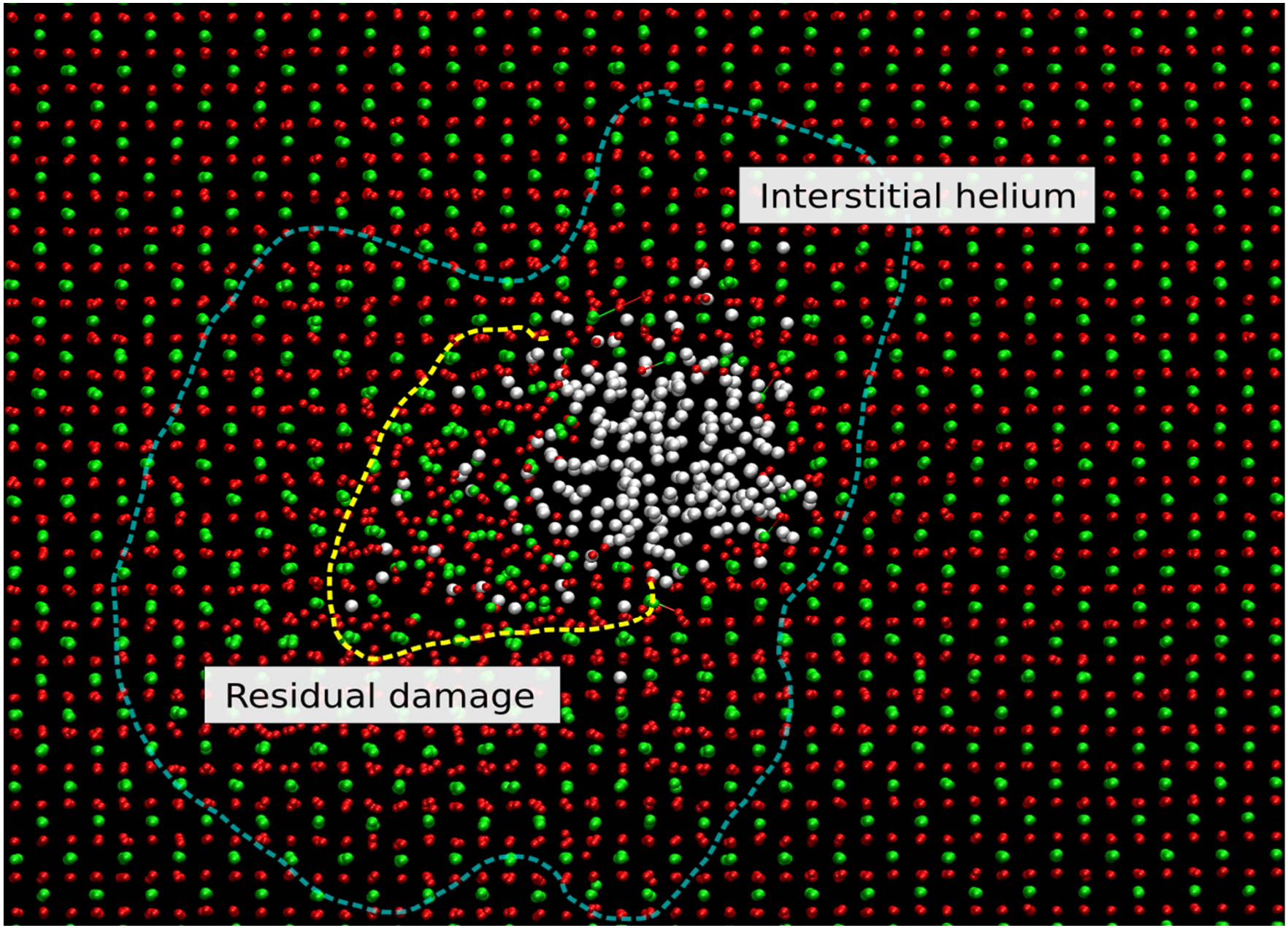
Helium in bubbles can return to the crystal lattice via radiation-enhanced re-solution rather than thermal resolution

...*But how does this actually work in practice?*

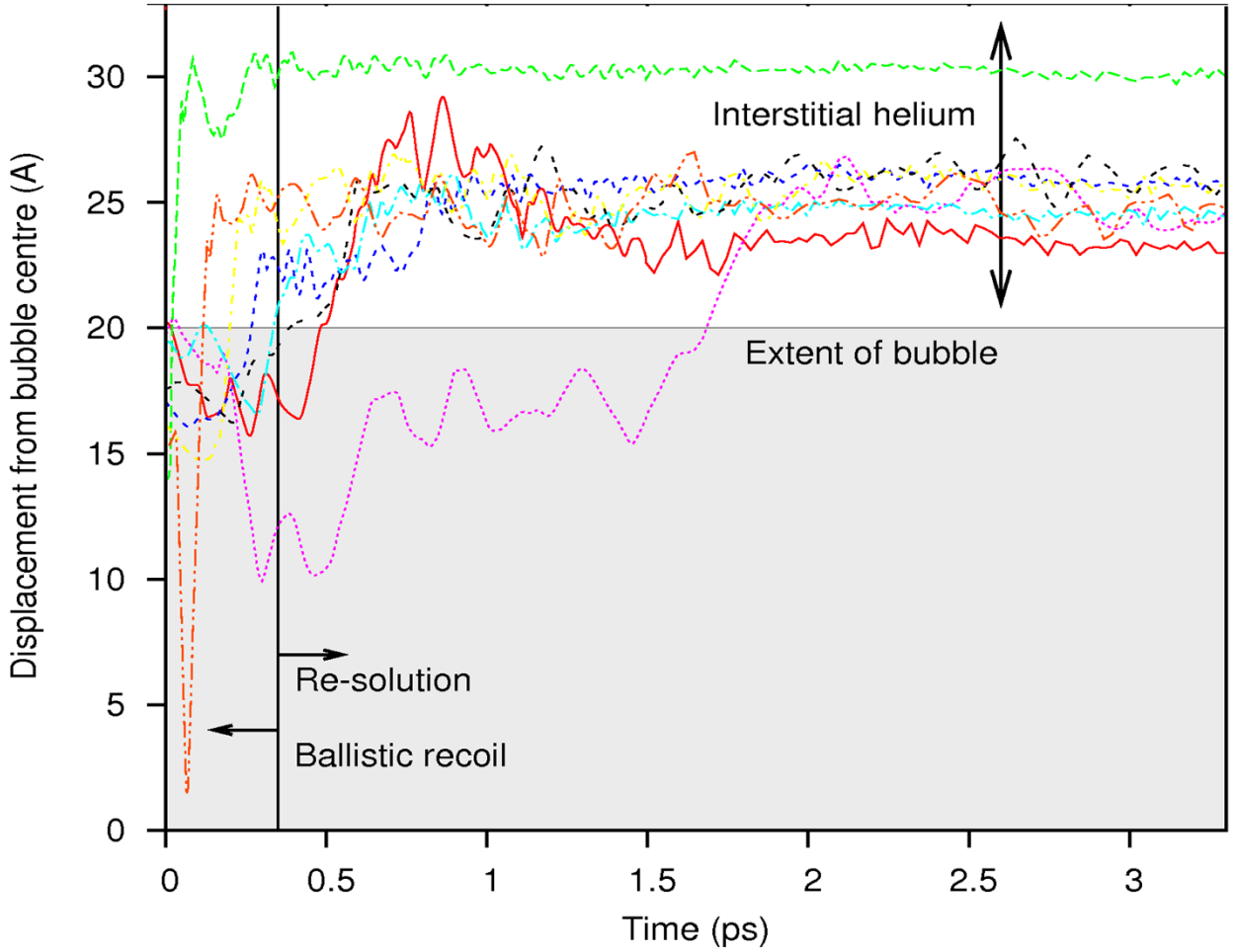
It is thought that high-energy fission fragments 'knock out' helium atoms from bubbles leading to resolution.


Uranium
Oxygen
Helium
| ~1 nm |
(110) 10 keV cascade


Secondary cascade
Helium absorbtion
Peak damage

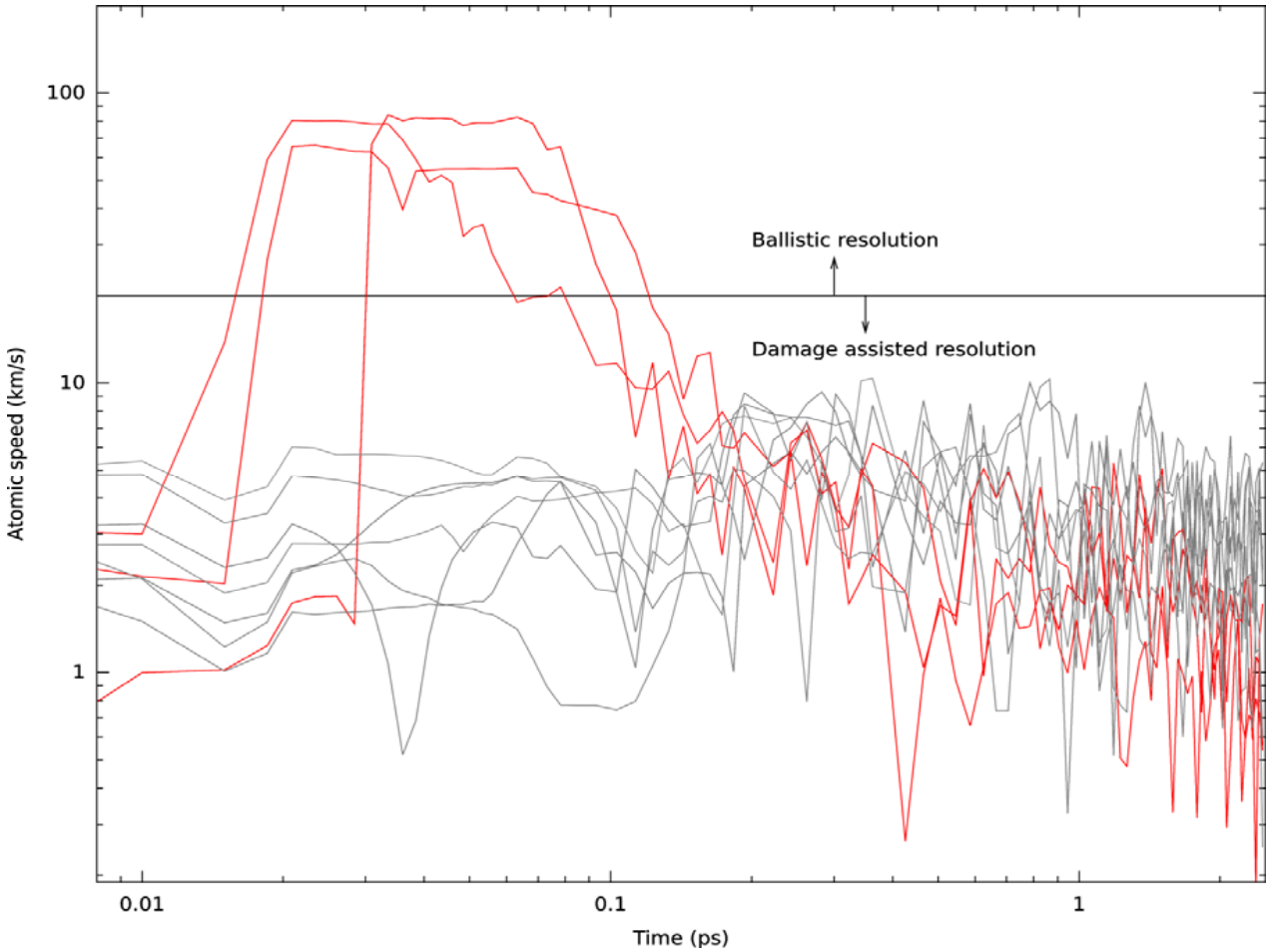

Interstitial helium
Residual damage

Resolution in 4nm bubble



<111> 10 keV cascade damage

Resolution in 4nm bubble



<111> 10 keV cascade damage

Why is this exciting?

- Physics behind this mode of radiation enhanced resolution is fundamentally different to what has been proposed previously.
- May explain some 'anomalous' terms in bubble migration models.
- More accurate and confident modelling leads to less conservatism in fuel performance codes.

Summary

- High performance computing has allowed us to make predictions of materials performance that we would not have thought possible even a decade ago.
- Success has required collaboration between computer facilities people and simulators.
- Length scales and time scales have been extended, although connecting these remains a challenge.
- Simulators can always use more computational power. Production runs take from between 1 hr and 2 days: they always did and always will!
- The link to experimental work remains key. For nuclear materials, a regulator will not accept a simulation on its own and never unless the validation step is clear.