Modelling irradiation damage in metals with Ehrenfest dynamics

Imperial College London

C. P. Race

Department of Physics, Imperial College London, London, SW7 2AZ, UK

100 years of living science

Simple abstract

Any metallic material used in nuclear fusion reactors will be bombarded by high-energy sub-atomic particles and will be damaged at the atomic scale. The amount and type of damage will affect which materials can safely be used and for how long. In support of experimental tests, we require a theoretical understanding of the damage process. Our project aims to improve that understanding by modelling damage events with new levels of sophistication.

Full abstract

A high energy neutron entering a metal soon collides with one or more ions of the material. These ions exchange energy with the other ions and with the electrons along their path and are brought to rest in displacement spikes: localised regions of concentrated damage. Over tens of picoseconds these displacement spikes cool and a final defect concentration is established. These defect populations determine the microstructural evolution of the metal. The exchange of energy between ions and electrons is thus critically important in determining the useful (and safe) life of materials under neutron bombardment.

The transfer of energy between ions in a metal is well modelled by classical molecular dynamics simulations. Energy transfer between ions and electrons is less well understood; theoretical work has been confined to highly idealised situations and simulations have at best treated the electrons implicitly as a viscous drag on moving ions. Literature estimates of energy loss to the electrons vary by several orders of magnitude.

Our project takes the modelling of collision cascades to the next level of sophistication, going beyond the Born-Oppenheimer approximation. By applying Ehrenfest dynamics in a time-dependent tight-binding simulation we (i) treat the electrons explicitly and quantum mechanically and (ii) determine their evolution under the influence of a Hamiltonian that explicitly includes the positions of classical ions. By examining the effect of the electrons on the motion of the ions we will determine their influence on the defect population. We will obtain quantitative estimates of the energy transfer from ions to electrons.

Simulation method

Our simulation method exploits a simple tight-binding model with a single, non-overlapping s-orbital on each atom. The model is parameterised to reproduce properties of copper and provides the simplest possible Hamiltonian with explicit treatment of the ionic configuration. We evolve a set of classical ions (positions $\{\mathbf{R}_n\}$ and momenta $\{\mathbf{p}_n\}$) according to the Ehrenfest equations within the Ehrenfest approximation, under forces derived from the tight-binding Hamiltonian $\hat{H}(t)$

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle\hat{\mathbf{R}}_n\rangle = \frac{\langle\hat{\mathbf{p}}_n\rangle}{m}, \qquad \frac{\mathrm{d}}{\mathrm{d}t}\langle\hat{\mathbf{p}}_n\rangle = -\boldsymbol{\nabla}_{\langle\hat{\mathbf{R}}_n\rangle}(\mathrm{Tr}\hat{H}\hat{\rho} - E_{\mathrm{rep}})$$

The electrons of the metal are treated quantum mechanically and evolved according to the quantum Liouville equation for the density matrix $\hat{\rho}(t)$

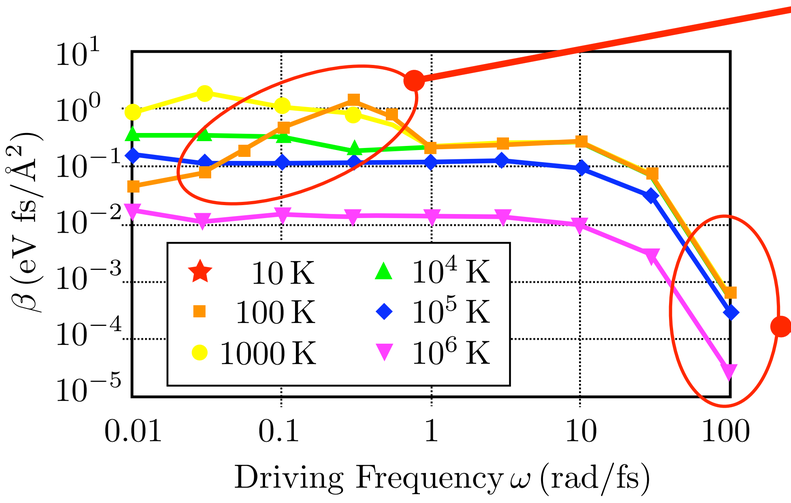
$$\dot{\hat{\rho}}(t) = -\frac{i}{\hbar}[\hat{H}(t), \hat{\rho}(t)]$$

Single oscillating ion

Results

To test the suitability of our system we have simulated a single oscillating ion in a 1120 atom block of our tight binding model. We have measured the energy transfer from the ion to the electrons in terms of an effective viscous damping coefficient β. The response has been measured as a function of oscillator frequency and electronic temperature.

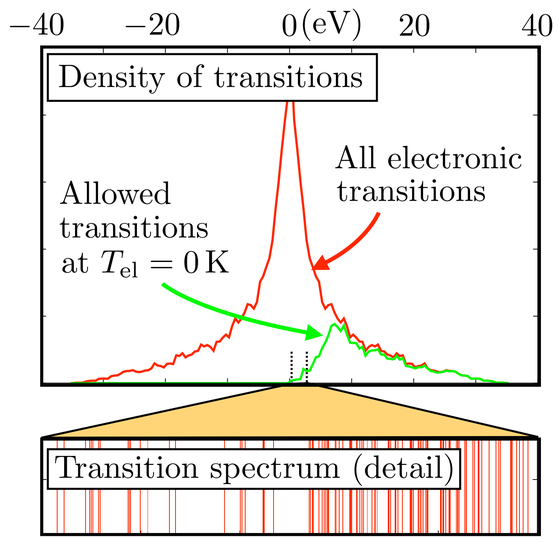
The behaviour of the damping coefficient highlights some important issues with our model.



Finite size effects

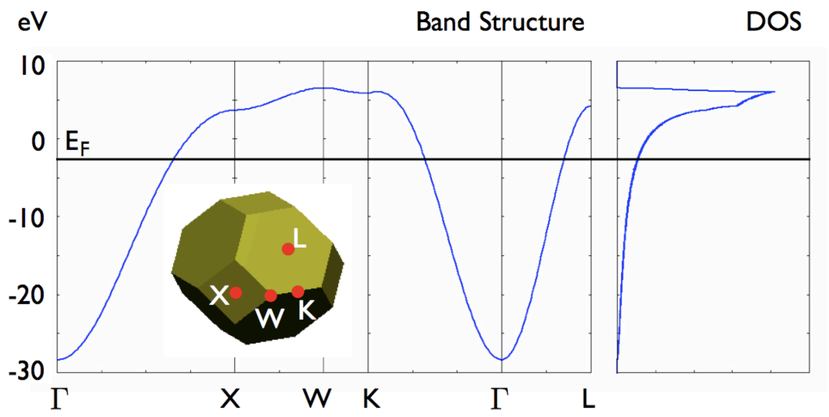
At low frequency and low temperature we see badly behaved variation in the damping coefficient due to the effect of the finite system size. To obtain good behaviour we need the the oscillator to stimulate a significant number of electronic transitions.

Calculations show that at low temperature the transition spectrum for the electronic system has sparse regions at low frequency.



Finite bandwidth effects

At high frequencies the damping falls off to zero. Our simple tight-binding model has a finite band width and cannot so cannot accommodate electronic transitions above a certain maximum energy.



Conclusions

Our single oscillator results show that the nature of the finite s-orbital tight binding model will place limits on the regimes of applicability of our simulations. However, we see that a simulation cell of over 1000 atoms should be large enough to give good results. In a dynamic simulation the ionic motion will include many characteristic frequencies and the spectrum of transitions will vary continually, reducing the impact of finite size effects. Also, our s-band is wide enough to cope with frequency components characteristic of the ionic velocities in displacement cascades of up to 20 keV.

Expected benefits from the course:

I hope to gain an overview and experience of a range of materials modelling methods. This will provide a context for my own work and help me to understand how my project compares with different approaches to similar problems. The output of my simulations of radiation damage will be defect distributions that constitute the initial conditions for materials simulations on longer time and larger length scales. Understanding the techniques used for such simulations will provide important context for my work.