The Transfer of Energy Between Electrons and Ions in Radiation Damage

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Abstract

We use time-dependent tight-binding simulations to investigate displacement cascades caused by irradiation of copper with high-energy neutrons. The neutron collides with an atom of the metal, the primary knock-on atom, and subsequent ion-ion collisions cause a cascade. We have run over 200 large scale simulations to observe the effect of different initial momenta.

Governing Equations

We assume that the Ehrenfest approximation is valid for modelling the combined system of electrons and ions. Thus the ions are modelled using classical mechanics and the electrons are treated using full quantum mechanics [1]. The electron system is represented by a quantum mechanical density matrix, $\hat{\rho}(t)$, and the ion system is given by a point $(\mathbf{R}(t), \mathbf{P}(t))$ in a $6N$ dimensional phase space, where N is the number of ions.

The system is evolved by the following $N+1$ coupled equations

$$i\hbar\dot{\hat{\rho}}(t) = \left[\hat{H}_e(\mathbf{R}(t)), \hat{\rho}(t)\right]$$

$$m_i\ddot{\mathbf{R}}_i(t) = -\nabla_{\mathbf{R}_i} H_I(\mathbf{R}(t)) - Tr\left\{\hat{\rho}(t)\left(\nabla_{\mathbf{R}_i}\hat{H}_e(\mathbf{R}(t))\right)\right\}$$

Here $\mathbf{R}_i(t)$ denotes the position of the ith ion; $\hat{H}_e(\mathbf{R}(t))$ is the electronic Hamiltonian parameterised by the ionic positions; $-\nabla_{\mathbf{R}_i}\hat{H}_e(\mathbf{R}(t))$ is the operator of the electronic force acting on the ith ion and $-\nabla_{\mathbf{R}_i}H_I(\mathbf{R}(t))$ is the ionic force acting on the ith ion.

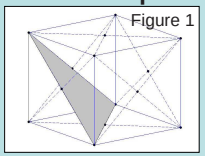
We further assume that it is valid to model the electronic system with a tight-binding framework [2]. The model chosen is described in [3]. Additionally we consider only a single-electron density matrix and assume electron-electron interactions are negligible.

Simulation Set-Up

Simulation Cell:

The simulation cell consists of 9x8x7 replicas of the face-centred cubic unit cell in fig 1 and hence 2,016 atoms.

Figure 1



Initial Condition for the Electrons:

The initial ionic lattice of the ions specifies an initial Hamiltonian. Let E_i denote the ith eigenvalue and $|\psi_i\rangle$ the corresponding eigenvector. We then set the initial single-electron density matrix, with a given temperature $T_e = 500\text{K}$, using

$$\hat{\rho}(0) = \sum_i \frac{1}{\exp\left(\frac{E_i - \mu}{k_B T_e}\right) + 1} |\psi_i\rangle\langle\psi_i|$$

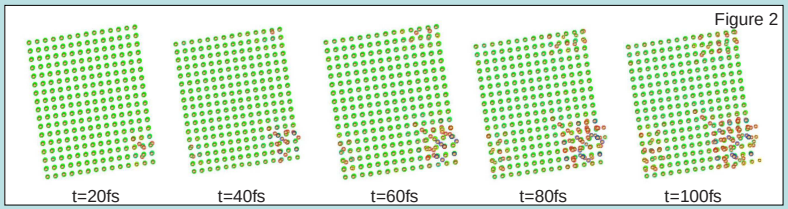
where the chemical potential μ is set to ensure the band filling specified in [3].

Initial Condition for the Ions:

A range of initial kinetic energies and directions were chosen for the primary knock-on atom. Kinetic energies of 100eV to 1keV were considered. Reflective and rotational symmetry means we need only consider 1/48 of the full sphere of possible angles.

Ionic Evolution

Figure 2



Since the ions are modelled classically as point particles we can observe their evolution in the same manner as molecular dynamics simulations. Figure 2 shows the initial stages of a cascade. Each of the plots is looking in the x-direction. The primary knock on atom has an initial energy of 100eV and initial direction (0.51,0.61,0.61).

Energy Transferred to the Electrons

The energy transferred to the electrons is not calculated correctly in a typical Born-Oppenheimer simulation. In such simulations the state of the electronic system is assumed always to be the ground state for a given set of ionic positions, $\hat{\rho}_{\text{BO}}(\mathbf{R}(t))$. Therefore if the ions returned to the same location the electronic system would return to the same state too. In violent events such as radiation damage this behaviour is unphysical.

In order to determine the error in Born-Oppenheimer simulations we consider the following energy difference

$$\Delta E(t) = \text{Tr}\{\hat{\rho}(t)H_e(\mathbf{R}(t))\} - \text{Tr}\{\hat{\rho}_{\text{BO}}(\mathbf{R}(t))H_e(\mathbf{R}(t))\}.$$

We refer to ΔE, which is always positive in our simulations, as *the energy transferred to the electrons*. The effect of the primary knock-on atoms initial direction and kinetic energy on ΔE is explored in the following results.

1. Figure 3 shows how the energy transfer to the electron system over time depends on the initial direction. The initial kinetic energy is 100eV. Each point on the triangle represents a different initial direction that can be determined in the following fashion:

i. the triangle corresponds to that portion of the (111) plane that intersects with the unit cell. (Shaded portion of fig 1.)

ii. the vector from the origin to the point on the triangle is an approximate value of the initial direction.

The middle of each edge corresponds to an initial direction toward the first nearest neighbour. Each corner corresponds to an initial direction toward the second nearest neighbour. The colour scale is normalised to 1 for the largest energy transfer after 50fs over any angle and 0 for no energy transferred.

2. Figure 4 shows the effect of initial direction in more detail at a time of 30fs. The colour scale is normalised to 1 for the largest energy transfer in this time and 0 for the smallest. The histogram only corresponds to a irreducible 1/6 of the triangle. Energy transferred may double depending on the direction considered.

3. Figure 5 shows the effect of different initial kinetic energies for an initial direction of (0.51,0.61,0.61).

Figure 4

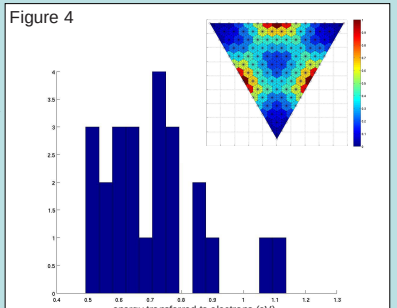
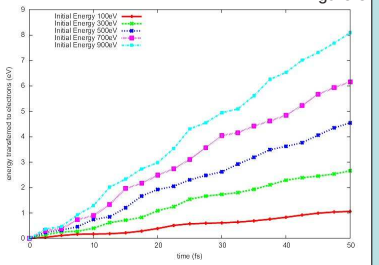


Figure 5



Bibliography

[1] J. B. Delos, W. R. Thorson and S. K. Knudson, Phys. Rev. A, Vol. 6, p709-720 (1972).

[2] T. N. Todorov. J. Phys: Condens. Matter, Vol 13, p10125-10148 (2001).

[3] A. Sutton et al, Phil. Mag. A, Vol 81, p1833-1848 (2001).

Figure 3