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Electronic effects in radiation damage simulations

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ABSTRACT

A methodology for including electronic effects in classical radiation damage simulations is presented. The method is used to calculate the number of residual defects for low energy (10 keV) cascades in Fe, as a function of the electron–phonon coupling strength. It was found that strong electron–phonon coupling reduced the number of residual defects by rapidly removing energy from the cascade and reducing the thermal spike. Intermediate coupling increased the number of defects by quenching the thermal spike and reducing defect recombination. Thermostatting the cascade with the local, time dependent electronic temperature, rather than the ambient temperature, reduced the number of residual defects by enhancing defect recombination. Swift heavy ion irradiation in tungsten was modeled using the same methodology. In this case we found that the number of residual defects created by a given electronic stopping power was strongly dependent on the temperature variation of the electronic heat capacity. In contrast to cascade simulations, the interstitials were located closer to the core of the ion track than the vacancies.

1. Introduction

Structural modification of materials by radiation is relevant to a broad range of technologically relevant processes, including the design of nuclear power plants and the nanostructuring of materials. Models of these radiation effects could play a major role in the understanding and control of such processes, but accurate predictive models are currently restricted to a limited number of applications. For example, low energy particle irradiation can be modeled successfully using cascade simulations in materials for which there are reliable interatomic potentials.

Models of radiation events which result in significant electronic excitation, such as laser irradiation, swift heavy ion irradiation and fusion neutrons, offer additional challenges to those presented by low energy particle irradiation. The inelastic energy loss due to electronic stopping has been included in cascade simulations by introducing a friction term in the equations of motion [1,2], but the effect of the deposited energy on subsequent lattice evolution was neglected. Laser and swift heavy ion irradiation deposit energy primarily in the electronic system and the lattice and electrons remain out of equilibrium for a timescale of the order of a few picoseconds. Femtosecond laser experiments are often interpreted using the two temperature model [3], which describes the temporal and spatial evolution of the electronic and lattice temperatures by coupled thermal transport equations [4]. Two temperature models do not, however, give information about structural modifications, therefore several groups have coupled such models to molecular dynamics simulations to give what is known as 2TM-MD hybrid models. The idea behind such models is that the electronic temperature evolution is calculated from numerical solutions of diffusion equations and energy is exchanged between the electronic model and the atomistic model of the lattice. Models of this kind have been used to simulate low energy cascades and laser ablation [5–7].

We have recently developed a 2TM-MD model that can be used simulate a range of radiation environments, by imposing different initial conditions [8]. Energy lost by the atomic system, by inelastic electronic scattering and electron–phonon coupling, is deposited in the electronic system, where it diffuses and re-deposits energy to the lattice. Thus the electronic system acts as a means for energy transport and storage. Low energy cascades are simulated by standard procedures but the energy lost to the electrons is gained by the electronic system [9]. The effect of swift heavy ion radiation is modeled by initializing the simulation with a column of "hot" electrons [10]. Intermediate radiation energies can be simulated

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by a combination of a cascade and a column of hot electrons. Thus the synergy between displacement damage and electronic effects can be captured [11].

The model improves traditional cascade simulations in metals, in that it includes a method for removing excess energy from the simulation cell which depends on the thermal properties. The number of stable defects created by a radiation event depends critically on the rate of cooling of the thermal spike; therefore a realistic representation of the cooling process is vital for accurate damage prediction. Whilst classical molecular dynamics simulations give a good description lattice thermal conductivity, at least above the Debye temperature, metallic thermal conductivity is dominated by electronic transport and is poorly described by MD simulations. The methodology we have developed introduces a mechanism whereby energy transferred to the electrons, via inelastic scattering or electron–phonon coupling, can be transported from the simulation cell by electronic thermal conductivity.

2. Methodology

The methodology is explained in detail in earlier papers [8,10]. The basic concept is that the trajectories of the atoms are calculated by standard molecular dynamics (MD), using the DL_POLY code [12], and the spatial and temporal evolution of the electronic temperature is calculated using a finite difference solution of the heat diffusion equation. At each time step energy is exchanged between the atomic and electronic systems in such a way that energy is conserved locally, and the amount of energy exchange depends on the local temperature difference between the electronic and atomic systems. A cubic grid is superimposed on the atomistic MD simulation cell and the electronic temperature is defined on this grid. The cells are referred to as coarse-grained electronic temperature cells and each contains a few hundred atoms. The electronic temperature grid extends well beyond the atomistic simulation cell and the outer cells are kept at a constant temperature, to represent the temperature of the "rest of the system".

Following Caro and Victoria [13], we employ a Langevin thermostat to implement energy exchange between the electronic and atomic subsystems. We consider two mechanisms for energy exchange with the electrons, electron–phonon coupling and electronic stopping. Electronic stopping is included as a friction term, which is implemented for atoms above some critical velocity (v_c). Electron–phonon coupling is also included as a friction term but, as it is a two-way energy exchange, there is an additional term representing energy input to the lattice from the electrons, and this takes the form of a random force with characteristics determined by the fluctuation dissipation theorem. Therefore the atomic equations of motion are given by:

$$m\frac{\partial \mathbf{v}_i}{\partial t} = \mathbf{F}_i(t) - (\gamma_p + \gamma_s)\mathbf{v}_i + \widetilde{\mathbf{F}}(t). \tag{1}$$

Here $\mathbf{v}_i$ and m are the velocity and mass of atom i and $\underline{\mathbf{F}}_i(t)$ is the force acting on atom i due to the interaction with the surrounding atoms at time t. The friction terms, with coefficients γ_p and γ_s, represent energy loss to the electronic system by electron–phonon coupling and electronic stopping, respectively. The stochastic force $\widetilde{\mathbf{F}}(t)$ returns energy to the atoms, from the electronic system, by electron–phonon coupling. The mean value of the force is zero and the magnitude of the fluctuations is related to the local electronic temperature (T_e) by the fluctuation dissipation theorem

$$\left\langle \widetilde{\mathbf{F}}(t') \cdot \widetilde{\mathbf{F}}(t) \right\rangle = 2k_B T_e \gamma_p \delta(t' - t). \tag{2}$$

The spatial and temporal evolution of the electronic temperature is calculated by a finite difference solution of the heat diffusion equation:

$$C_e\frac{\partial T_e}{\partial t} = \nabla(\kappa_e \nabla T_e) - g_p T_e + g_p T_a + g_s T'_a. \tag{3}$$

Here C_e and κ_e are the electronic specific heat and electronic thermal conductivity respectively, g_p and g_s are coupling constants for energy exchange between the atomic and electronic systems. T_a is related to the average kinetic energy of the atoms in a coarse-grained cell and T'_a is related to the average kinetic energy of the subset of atoms with energy greater than v_c. Here energy is lost to the lattice by term 2 on the right hand side of (3) and gained from the lattice by terms 3 and 4. Energy conservation is imposed by relating the parameters (g_p, g_s, T_a and T'_a) to the parameters of the MD simulation (m, $\underline{v}_i$ and γ_p and γ_s) as outlined in [8].

The simulation setup depends on the type of radiation to be modeled. For low energy cascades we impose periodic boundary conditions on the atomistic simulation cell and constant temperature boundary conditions on the boundary of the extended electronic temperature cell. The cascade is initiated in the centre of the atomistic cell. Energy lost by the atomistic system, due the friction term in the equation of motion, is input as a source term in the electronic heat diffusion equation. The temperature associated with stochastic force in (2) refers to the local electronic temperature, which varies in time and space. For this reason we refer to it as an inhomogeneous Langevin thermostat.

Swift heavy ion irradiation can be modeled using the same methodology. Here we assume that the ion has passed through the simulation cell leaving a track of excited electrons in its wake. We assume that after a few femtoseconds the highly excited electrons will have experienced sufficient collisions to establish an approximate Fermi Dirac distribution, associated with a local electronic temperature. We calculate the excited electronic temperature (T_x) in the track from the initial electronic temperature (T_0), the electronic stopping power (S, the energy loss per unit distance), the cross-section area of the track (A) and the temperature dependant electronic heat capacity (C_e). If we assume that the energy is initially deposited in the central column of coarse-grained cells (cross-section area, A), the electronic temperature (T_x) is calculated using the following equation:

$$S/A = \int_{T_0}^{T_x} C_e(T'_e)\,dT'_e. \tag{4}$$

Constant temperature boundary conditions are imposed in the electronic simulation cell in the two directions perpendicular to the track and periodic boundary conditions are used in the direction parallel to the track. The electronic energy diffuses and deposits energy in the lattice, resulting in heating and, in some cases, local melting.

The model requires a number of material parameters, such as the electronic heat capacity, the electronic thermal conductivity, the electron–phonon coupling constant and electronic stopping parameter. Each parameter will depend on the electronic temperature as discussed for a range of metals by Lin et al. [14,15]. For results presented in this paper we have used the experimental room temperature thermal conductivity for both Fe ($80\ \mathrm{Wm^{-1}\,K^{-1}}$) and W ($177\ \mathrm{Wm^{-1}\,K^{-1}}$). We use a form of the specific heat capacity for Fe that varies linearly with temperature at low temperature and saturates at $3k_b$/atom (k_b is Boltzmann's constant) at high temperature. We compare the results for two heat capacity models for W as discussed below. A range of electron–phonon coupling constants is used for the Fe simulations and a typical experimental value is used for W.

3. Results and discussion

We present the results from two types of simulations in this paper, which represent extensions of work presented previously. The