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Including electronic effects in damage cascade simulations

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A B S T R A C T

A method for including the effects of electronic losses and electron–phonon coupling in radiation damage simulations has been developed and implemented for 10 keV cascades in Fe. The MD simulations are coupled to a continuum model for the electronic energy and energy lost by the atoms, due to electronic friction and electron–phonon coupling, is gained by electronic system. Electronic energy transport is described by the heat diffusion equation and energy is returned to the lattice via a stochastic force. Thus the temperature of the atomic system is controlled by a Langevin thermostat at the local electronic temperature, which varies with time and space. The results of simulations with this inhomogeneous thermostat are compared with those of homogeneous (constant temperature) thermostat simulations for a range of electron–phonon coupling strengths. The residual defect concentration was found to have a non-monotonic variation with coupling strength.

1. Introduction

Classical cascade simulations have a long and successful history in the area of modeling radiation damage; however such simulations generally include electronic effects only through the interatomic potentials. The role of electron–phonon coupling in transporting energy from a cascade was noted two decades ago [1] and implemented in simulations via a friction term [2] and a Langevin thermostat [3]. Fast atoms moving in solids lose energy due to inelastic collisions with electrons and this electronic stopping has been included in cascade simulations via a friction term [4]. For metals with strong electron–lattice interactions and low thermal conductivity the energy lost to the electronic system will be localized near the cascade and redistributed to the lattice, which may result in additional damage or defect annealing. Previous models have neglected this redistribution of the electronic energy to the lattice.

We have recently developed a methodology that includes the electronic energy transport and redistribution to the lattice. The molecular dynamics (MD) simulations are coupled to a coarse-grained representation of the electronic energy. Energy lost or gained by the lattice in each MD timestep is gained or lost by the electronic system. Electronic energy diffusion is modeled via a finite difference solution of the heat diffusion equation. Similar methodologies have been used to model sputtering and laser irradiation [5,6]. We use the model to study the lattice and electronic temperature evolution and the residual defect numbers for 10 keV cascades in Fe, for a range of electron–phonon coupling strengths.

2. Method

Electronic effects are represented in the simulations by coupling the MD cell to a coarse-grained model for the electronic energy. A cubic lattice is superimposed on the atomistic simulation cell and the electronic temperature is defined within each coarse-grained electronic temperature (CET) cell. At each MD step the atomistic equations of motions and the heat diffusion equation for the electronic energy are evolved, and energy is exchanged between the atomistic and electronic subsystems. The electronic simulation cell extends well beyond the atomistic simulation cell and the electronic temperature (T_e) is fixed at 300 K at boundaries of the extended cell. The velocity ($\mathbf{v}_i$) of atom i (mass m), subjected to a force $\mathbf{F}i$ due to interactions with surrounding atoms, evolves according to a Langevin equation, which is solved numerically using standard MD techniques.

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

Here the friction term γ_i represents energy loss by both electron–lattice interactions and electronic stopping. Electronic stopping is implemented for atoms with kinetic energy greater than a cutoff E_{cut}, taken to be twice the cohesive energy (8.63 eV for Fe). The magnitude of the random force $\widetilde{\mathbf{F}}(t)$ is determined by the *local* electronic temperature, that is T_e of the CET cell that atom belongs to. We refer to this as the *inhomogeneous* Langevin thermostat. The evolution of the electronic temperature is described by the heat diffusion equation:

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$$C_e \frac{\partial T_e}{\partial t} = \nabla(\kappa_e \nabla T_e) - g_p(T_e - T_a) + g_s T'_a \quad (2)$$

Here C_e and κ_e are the electronic specific heat and the electronic thermal conductivity, respectively. T_e is the local electronic temperature and T_a is the effective local atomic temperature, calculated from the average kinetic energy of the atoms belonging to a CET cell. T'_a also has dimensions of temperature, but it is related to the kinetic energies of atoms with energies greater than the electronic stopping threshold. The second and third terms on the right hand side of (2) represent energy exchange with the lattice via electron–ion interactions and electronic stopping, respectively. In [7] we demonstrate that energy loss/gain balance is ensured if:

$$g_p = \frac{3Nk_B\gamma_p}{\Delta V m} \quad (3)$$

$$g_s = \frac{3N'k_B\gamma_s}{\Delta V m} \quad (4)$$

Here ΔV is the volume of the CET cell, N is the number of atoms in the CET cell, N' the number of atoms with velocities higher than E_{cut} and k_B is Boltzmann's constant. The electronic stopping coefficient γ_s/m may be obtained directly from the electronic stopping curves of TRIM [8] and for Fe it is found to be of the order of 1 ps^{-1}. Estimates of γ_p have been obtained from experiments for some metals but no experimental values are available for Fe. However from the model outlined in [9] we expect χ ($\chi = \gamma_p/m$) for Fe to be in the range 1–2 ps^{-1}. In this work we explore a wide range of χ (0.5–30 ps^{-1}) in order to investigate the sensitivity of the results to the coupling strength.

The model has been implemented in DL_POLY [10] and a series of 10 keV cascades for Fe, interacting via the Dudarev–Derlet potential [11], was performed. The atomistic simulation cell had 250000 atoms, with periodic boundary conditions. The electronic temperature was defined on 10^6 CET cells and the central cube of 729 CET cells was coupled to the atomistic cell. The remainder of the CET cells served to transport electronic energy away from the central cell, which represents the electronic heat sink of 'the rest of the system'.

Very high energy radiation events are known to decompose into a number of sub-cascades, as they create a small number of high-energy knock-on events. Between such events the moving atom loses energy to the lattice via three mechanisms, low energy cascades, phonon creation and electronic excitations. Using our methodology we can include the effect of the latter, by initiating the cascade with an elevated electronic temperature along a column of the simulation cell, terminating at the knock-on atom. We investigate the effect of the electronic loss for 60 keV cascade simulations, using a simulations cell with 986078 Fe atoms and χ = 1.5 ps^{-1}. Three simulations were performed, with a homogeneous thermostat, an inhomogeneous thermostat and an inhomogeneous thermostat with an initial T_e of 15000 K, to represent electronic loss caused by the atom slowing from 500 keV to 60 keV.

3. Results and discussion

The primary aim of the current study is to examine the effect of the strength of the electron–phonon interactions on the residual defect numbers, and also compare the results with simulations that do not include the energy returned to the lattice from the electronic system. These simulations are representative of situations where the electronic energy is transported rapidly from the cascade region (high thermal conductivity) and this is modeled using a homogeneous Langevin thermostat. The results are presented in detail in [12] therefore here we summarize the more significant findings. We find that T_e rises to a maximum value, which increases with the coupling strength, in a few hundred femtoseconds. This is followed by a rapid decay where the cooling rate is dominated by thermal conduction out of the cascade region. In the later stages the rate of cooling decreases, particularly for strong coupling, as there is significant energy exchange between the lattice and the electrons.

The mean number of residual defects is summarized for all the simulations in Fig. 1. We note the non-monotonic relationship between the coupling strength and the defect number, particularly for the homogeneous thermostat. For high coupling strength the rapid removal of energy from the cascade by interaction with the cold electrons inhibits the formation of the thermal spike (molten zone) and the peak number of defects is strongly reduced, resulting in fewer residual defects. For intermediate coupling there is some quenching of the thermal spike but the rapid cooling of the cascade freezes in more defects, therefore the residual defect number increases. The defect numbers for low coupling tends towards the value obtained for constant energy (NVE) simulations (15.5 ± 1.5).

The inhomogeneous simulations have lower numbers of residual defects than the homogeneous simulations for all coupling

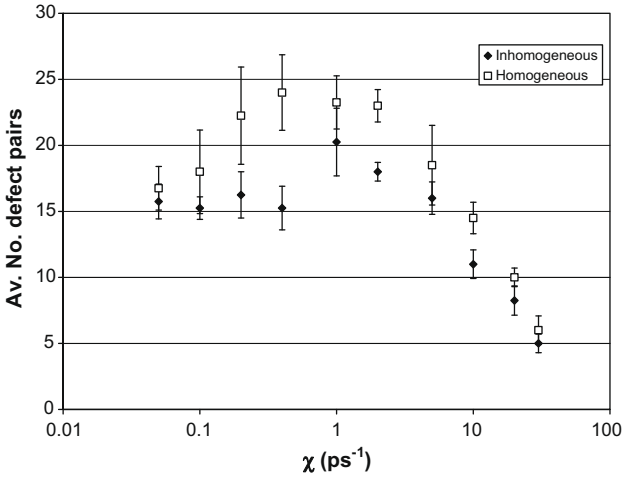

Fig. 1. The mean number of stable residual defect pairs for 10 keV cascade simulations with inhomogeneous (filled diamonds) and homogeneous (open squares) thermostats and a range of coupling constants. The error bars represent the standard error for four independent simulations.

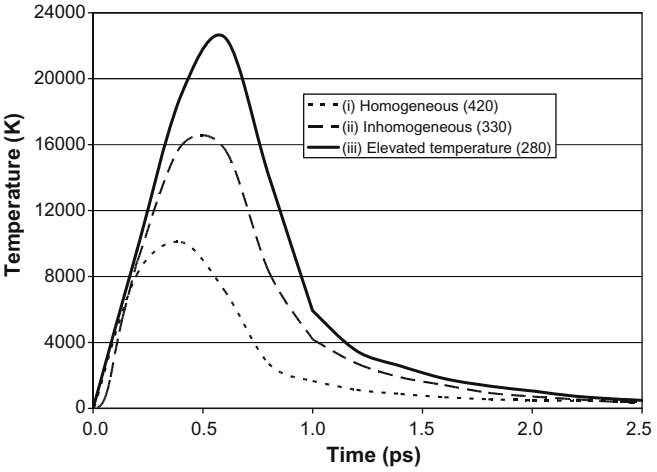

Fig. 2. The time evolution of the total defect numbers for 60 keV cascade simulations with a homogeneous thermostat (solid line), inhomogeneous thermostat (broken line) and inhomogeneous thermostat with initial electronic temperature elevated in a column of the simulation cell (dotted line). The numbers in brackets in the key show the defects numbers at the end of the simulations.

strengths. This is due to two effects. Firstly the inhomogeneous simulations include an additional friction term to represent electronic stopping at high energies. Secondly the inhomogeneous simulation is thermostatted with the local electronic temperature, which results in slower cooling of the cascade and increased defect recombination.

The effect of enhanced electronic temperature is even more apparent for high PKA energies. The defect evolutions for the three 60 keV cascades are plotted in Fig. 2, with the residual defect numbers included in the key. We note that the elevated electronic temperature simulation, which was initiated with a electronic temperature of 15 000 K in a half column of the electronic simulation cell, has the highest maximum number of defects (largest thermal spike) and the homogeneous thermostat has the lowest maximum defect number. However the trend is reversed for the residual defect numbers, with the elevated temperature simulation having the lowest number of residual defects. Thus the increased thermostatting temperature both increases the maximum disorder and increases defect recombination, with the net result that a reduced number of residual defects are created.

4. Conclusions

We have included the effects of electronic energy losses and redistribution in cascade simulations by coupling the atomistic simulations to a coarse-grained model to describe electronic energy transport. We have found a non-monotonic relationship between the number of residual defects and the strength of the electron–ion coupling. Strong coupling tends to quench the cascade and inhibit the formation of the full thermal spike. Intermediate coupling increases the cooling rate and freezes in additional defects. Thermostatting with the local electronic temperature enhances defect annealing. These effects are enhanced for high (60 keV) PKA energies. Inelastic scattering raises the electronic temperature along the path prior to the cascade and including this effect further enhances defect annealing, resulting in a smaller number of residual defects. Thus we conclude that a complete description of a damage cascade in metals should include the effects of electronic energy transport and redistribution.

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