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Assessment of the relation between ion beam mixing, electron–phonon coupling and damage production in Fe

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ABSTRACT

As a step in the process of assessing the reliability of interatomic potentials for iron, we compare experimental measurements of ion beam mixing with values obtained from molecular dynamics simulations. We include the electron–phonon coupling (EPC) model by Hou et al. [Q. Hou, M. Hou, L. Bardotti, B. Prével, P. Mélinon, A. Perez, Phys. Rev. B 62 (2000) 2825] in the simulations and consider a range of coupling strenghts. Three different iron interatomic potentials are used. We discuss the effect of the coupling on the primary damage and how the damage is influenced by different velocity minima for applying electron stopping.

1. Introduction

The reliability of iron interatomic potentials is not yet established, since the description of primary damage often varies from one potential to another. Although the variation when comparing all potentials is very large [2], we have recently shown that when only the modern potentials that describe the energetics of interstitials reasonably are included in the comparison, the discrepancy is much smaller [3]. In that study, however, the uncertainty in how the transfer of energy from the atomic to the electronic subsystem (electronic stopping S_e [4] and electron–phonon coupling (EPC) [5]) should be treated was not considered. Unfortunately there are large uncertainties in how these should be treated at low (of the order of 1–10 eV) atom kinetic energies [6–8]. To reduce the uncertainties in the damage production, S_e, and EPC, one could benefit from comparisons to experimental quantities that directly depend on the cascade development. One of these quantities is the ion beam mixing (IBM).

IBM is simply the athermal relocation of atoms from their lattice sites by ion irradiation. This mixing is experimentally measurable by for instance observing the broadening of a marker layer under ion irradiation [9]. In iron, only two of these experiments have been done, yielding mixing efficiencies of 4.5—4.6 Å^5/eV [9] and 7.2, 8.1 Å^5/eV [10,11], respectively.

The mixing crucially depends on the cascade development, since ion irradiation produces multiple cascades. In these, the energy end heat distributions are consequential, implying that correct descriptions of electronic stopping S_e and electron–phonon coupling (EPC) are required. S_e slows down ballistic atoms and thus reduces the cascade region and with that also the mixing. However, uncertainties regarding this quantity exist, e.g. the lowest energy at which the electronic friction should be applied is debated [7]. EPC also reduces the mixing, since a coupling between the electrons and the lattice results in a fast distribution of the heat from the hot cascade core to the cooler electronic gas. This suppresses the liquid region in which the atoms can redistribute and mix. In metals, the thermal conductivity is handled predominantly by the electrons in the beginning of a cascade [5,12], which indicates that the EPC could play an important role in iron.

When modelling cascades with molecular dynamics (MD) simulations methods, the EPC is not taken into account in the conventional MD algorithms [13], but can be added with various schemes [1,14–19]. Studies of the effect of EPC on cascade damage in a

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range of coupling strengths in Fe have been done [16,20,21] with the conclusion that a strong coupling has an influence on the resulting damage. However, the actual coupling strength in iron has not been determined, hence, the importance of including EPC models in cascade simulation in iron is still unclear.

Here we carry out a systematic study of the role of the EPC and S_e in iron cascades by reproducing an IBM experiment with molecular dynamics methods. We use three different iron potentials and we assess how the quantities affect the primary damage.

2. Methods

2.1. Simulating the ion beam mixing

The Kim et al. iron IBM experiment [9] was done at 6 K with 650 keV Kr ions irradiating Fe, with Pt and Au used as tracer. Pt tracers were also used in the other experiment [10], where iron matrices were irradiated by 150 keV Ar ions. Temperatures in the range 18–345 K were used, resulting in two different values (a lower for 20 K and a higher for 29–345 K). The large amount of oxygen impurities in this experiment resulted in large uncertainties, hence, the Kr experiment can be considered more accurate and therefore we chose to simulate that one.

The IBM simulations were done in two steps. First, molecular dynamics (MD) range calculations were performed to obtain the recoil spectrum $n(E)dE$ of the Kr ions irradiating Fe. The spectrum and the deposited nuclear energy was calculated in the range 200—600 Å from the surface, and the marker layers in the corresponding experiment were deposited at a depth of 400 Å. (The thickness of the markers in the experiment was 5–15 Å.) The angle between the beam and the normal to the surface was $\Theta = 10^\circ$ in both the experiments and the simulations and ϕ was additionally varied randomly in the 0–360° interval in the simulations.

Full MD simulations (at 300 K) were used to simulate cascades caused by self-recoils, the energies of which ranged from 0.5 to 200 keV. At least 10 cases for each energy were simulated, the exception being the 200 keV recoils, of which only 6 events were simulated. Three different Fe potentials were used: AMS [22], DD-BN [23,3] and MEA-BN [24,3].

From the cascades, the square of the total atom displacement, R^2, was obtained. This corresponds to the difference between the positions of the atoms at the end and the beginning of a cascade, i.e.

$$R^2 = \Sigma_i (r_i(t) - r_i(t=0))^2. \tag{1}$$

A function was fitted to the data points in order to be able to inter- and extrapolate. The function takes both the low and high (sub-cascade formation) energy dependency into account.

$$R^2(E) = \frac{aE^{3/2}}{b^{1/2} + E^{1/2}} \tag{2}$$

The experimentally measured normalized mixing efficiency is defined as

$$Q_{exp} = \frac{Dt}{\Phi F_{D_n}}, \tag{3}$$

where D is an effective diffusion coefficient for mixing, t is the implantation time, Φ the ion fluence and F_{D_n} the deposited nuclear energy per unit depth [9]. The unit of the efficiency is $Å^5/eV$. Using the atomic definition of the diffusion coefficient, $D = \frac{<r^2>}{6t}$, this is equal to the simulated mixing

$$Q_{sim} = \frac{R^2}{6n_0 E_{D_n}}, \tag{4}$$

where n_0 is the atomic density (in a BCC material, the atomic density is $n_0 = 2/a_0^3 \cdot a_0^{\text{DD-BN}} = 2.86$ Å, $a_0^{\text{MEA-BN}} = 2.89$ Å and $a_0^{\text{AMS}} = 2.86$ Å at 300 K). E_{D_n} is the deposited nuclear energy.

The total cumulative mixing efficiency resulting from the ion irradiation is obtained by

$$Q_{sim}^{tot}(E_0) = \frac{\int_0^{E_0} R^2(E) n(E) dE}{6n_0 E_{D_n}}, \tag{5}$$

where E_0 is the ion energy (650 keV for Kr, 150 keV for Ar) and $n(E)dE$ is the above mentioned primary recoil spectrum. This method for relating simulated and experimental values of the mixing efficiency has previously been employed successfully for different materials [25].

2.2. The electron–phonon coupling

The EPC coupling model of Hou [1] was implemented into the cascade simulations. Details of the model is found in [1,5,12] and is here only described briefly. The electronic system is considered as a heat bath of temperature T_e, and when ignoring the phonon diffusion, the change in the temperature of the ionic system is expressed as

$$\frac{dT_i(t)}{dt} = -\alpha(T_i(t) - T_e), \tag{6}$$

where (using the Sommerfeld free electron theory)

$$\alpha = \frac{\Theta_D L n e^2 k_B Z T_e}{2 m_e \kappa \epsilon_F}. \tag{7}$$

Θ_D is the Debye temperature, L = Lorentz number, n = electron density, e = the electron charge, k_B = Boltzmann's constant, Z is the valence, m_e = the electron mass, κ = thermal conductivity and ϵ_F = the Fermi energy. The values for these parameters in iron are found in Table 1. The time constant for the coupling is $\tau = \alpha^{-1}$ and a large time constant indicates a strong coupling.

The electron–phonon energy exchange can be described as a damping force. This force, acting on atom i with the velocity $\mathbf{v}_i$, can be written as

$$\mathbf{F}_i = -\mu \mathbf{v}_i, \tag{8}$$

where

$$\mu = m_i \alpha \frac{T_i - T_e}{T_i}. \tag{9}$$

In order to avoid singularities as the velocities are approaching zero, this expression is written as

$$\mu = m_i \alpha \frac{T_i - T_e}{\left[T_i^2 + (T_e/20)^2)\right]^{1/2}}. \tag{10}$$

The factor 1/20 was chosen to be compatible with the time steps used in MD [12].

This damping is included in the MD code together with the electronic stopping which also acts as a damping force. The two forces have to be joined at some suitable velocity. The electronic stopping dominates at high velocities and should not be applied to low-energy ions (otherwise the linear dependence on velocity quenches any simulations down to 0 K). Therefore, below a velocity corresponding to the cohesive energy of iron, only damping due to

Table 1
Constants used in the calculation of the EPC time constant of iron [26,27].

Θ_D	420 K
L	2.61 W ohm/K^2
n	$17.0 \cdot 10^{28}$ m^{-3}
Z	2
κ	0.8 $W/$(cm K)
ϵ_F	11.1 eV
m_{Fe}	55.85u