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Effect of valence holes on swift heavy ion track formation in Al_2O_3

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ABSTRACT

This paper focuses on effects of electrons and valence holes on structure modifications in swift heavy ion (SHI) tracks in dielectrics. To investigate this problem a multiscale model is constructed which consists of (a) Monte Carlo modeling of a SHI penetration and secondary electron cascading; (b) molecular-kinetic approach for low-energy electrons spatial redistribution after finishing of ionization cascades; (c) molecular dynamics modeling of lattice excitation due to its coupling with the relaxing electron ensemble and subsequent atomic dynamics on picoseconds timescales. The model is applied to 167 MeV Xe ion irradiation of solid Al_2O_3. We found that lattice heating by excited electrons does not exceed ~200 K, which is not sufficient to form an ion track observed in experiments. Structure transformations appear in the numerical simulations only when the excess energy accumulated in valence holes is taken into account.

1. Introduction

A swift heavy ion (SHI, $M > 20$ a.m.u., $E > 1$ MeV/nucl) loses a large part of its energy on excitation of the electron subsystem of a target in the nanometric vicinity of its trajectory (>95%, 1–40 keV/nm along the projectile path) [1]. This energy loss occurs via target ionization resulting in generation of fast electrons. Propagation of these electrons from the SHI trajectory produces new ionizations forming next generations of electrons and holes. During the subsequent relaxation of the excited electron ensemble, a part of its excess energy is transferred to target atoms resulting in transient lattice excitation in an ion track.

Dissipation of the excess energy of delocalized electrons in SHI tracks occurs on shorter times than those of atomic vibrations and, thus, the characteristic time of the collective modes of a lattice [2]. Therefore, an adequate description of energy transfer from excited electrons to the lattice in a SHI track requires a more general approach than that based on electron–phonon coupling [3–6]. Such an approach must take into account possibilities of realization of various limit cases of dynamical response of lattice atoms during their interaction with the relaxing electron ensemble in a track.

The presented microscopic multiscale model describes the kinetics of excitation of a dielectric in a SHI track up to picoseconds after the projectile passage. It consists of three different approaches combined together [7,8]. At the first stage, up to tens of femtoseconds after the projectile passage, a Monte Carlo (MC) [9–11] model is applied to simulate initial excitations of the electron subsystem by an SHI as well as further kinetics of fast electrons appearing due to ionization. At longer times (>10 fs), spatial spreading of electrons is accounted for by means of the molecular-kinetic method. The energy and momentum exchange of excited electrons with the lattice is calculated tracing the atomic trajectories with the molecular dynamics (MD) method up to picoseconds after the SHI passage.

The model is applied to estimate lattice heating of Al_2O_3 in the track of 167 MeV Xe ion. The simulations demonstrate that heating of Al_2O_3 due to coupling of its lattice to delocalized electrons is too low (~200 K) to produce structure transformations detected in experiments [12]. Such structure transformations appear in the numerical simulations only when the lattice is additionally supplied with the excess energy accumulated in valence holes (simulated track has a diameter of 2 nm vs 1.3 nm track observed in the experiment [12]).

2. Multiscale model

We apply an iterative scheme to estimate the spatial and temporal dependencies of the energy transferred into the lattice of Al_2O_3 from the excited electrons in a SHI track. First, the MC model

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is used to determine the initial radial distributions of the parameters characterizing an excited state of the ensemble of electrons: their concentration and the energy density. These initial distributions are used to restore the initial local equilibrium distribution functions of electrons at different distances from the axis of the track coinciding with the SHI trajectory. After finishing of ionization cascades, the molecular-kinetic method provides the changes of the concentration and the energy of low-energy electrons at different radii from the ion trajectory [8].

Using the dynamic-structure factor (DSF) formalism described below, the rates of the energy exchange of excited electrons with the lattice at different distances from the ion trajectory are calculated resulting also in decrease of the excess energy of the electron ensemble. The energy transferred from electrons to the lattice in different track regions is distributed among the different species of atoms by increasing their kinetic energy in accordance with their mass fractions.

MD tracing of atoms enables us to obtain the transient lattice DSF and, thus, realistic cross sections of the electron–lattice energy exchange in a relaxing SHI track. The same MD also models the lattice relaxation and spatial dissipation of the excess energy of the atoms after cooling of the electron ensemble [7,8]. The characteristic time of cooling of delocalized electrons in a SHI track due to their spatial spreading is shorter than the time of thermalization of the nonequilibrium lattice [13]. Therefore, we use the kinetic temperature of lattice atoms to estimate lattice heating during this exchange [14]:

$$T_{kin}(\mathbf{r},t)=\frac{1}{P}\sum_{\alpha=1}^{P}T_{kin,\alpha}(\mathbf{r},t)=\frac{1}{P}\sum_{\alpha=1}^{P}\frac{M_\alpha}{3k_B(Q_\alpha-1)}\sum_{n=1}^{Q_\alpha}(\mathbf{v}_n-\mathbf{v}_\alpha^c)^2, \qquad (1)$$

Here P is the number of different types of atoms of a target ($P=2$ for Al_2O_3); k_B is the Boltzmann constant; Q_α are the numbers of atoms of each type α in a volume where the kinetic temperature is defined; $\mathbf{v}_n$ is the velocity of an α-atom of number n; M_α, $\mathbf{v}_\alpha^c$ are the masses and velocities of the center of mass of α-atoms in a cell which this α-atom n belongs to.

For estimation of the valence holes contribution to lattice excitation we use the simplest model of an instant energy deposition to the atomic system with subsequent lattice relaxation by means of MD. This crude procedure does not assume any particular physical mechanism governing such energy exchange, and is used merely for a 'zero-order' "upper" estimation of the effect of valence holes.

The details of different modules of the model are described in the next subsections. We would like to emphasize that the model does not require any knowledge in advance about the experimental track radius, it has a full predictive capability.

2.1. Monte Carlo simulation of the initial kinetics of the electron ensemble in a swift heavy ion track

The details of the numerics of the MC model used to describe the excitation of the electronic subsystem of a target are presented in Refs. [9–11]. At the first step, the model simulates a passage of an ion and its energy losses which result in creation of the first generation of excited electrons. The ion trajectory within our simulation slice of 10 nm is assumed to be a straight line along the Z-axis because of negligible energy transfer in a single ion–electron scattering event compared to the kinetic energy of a projectile due to the large difference in their masses.

The model is based on the cross sections of interaction of excited free electrons and an SHI with the electron and atomic subsystems of a target. A simple but efficient approach describing interaction of an incident projectile with a dynamically coupled system of particles is based on (a) the one-particle approximation for a projectile; and (b) the assumption of weak interaction of this projectile with the scattering system (the first Born approximation) [11]. In this case, the differential cross section is factored into the cross section describing scattering of a projectile on an isolated particle of the scattering ensemble, and the dynamic structure factor (DSF) describing a collective response of this ensemble to the transferred energy and momentum.

The fluctuation dissipation theorem links the DSF of the ensemble of charged particles with the complex dielectric function (CDF) of this ensemble (a target). This allows to use the experimental optical data for construction of the CDF-based cross sections used in the MC approach [10,11]. The CDF formalism allows us to reconstruct all the effects of the whole valence band of Al_2O_3 crystal, so we do not need to describe "partial" effects from valence electrons of the different kinds of atoms.

The mean free paths of electrons calculated within the CDF-formalism agree well with the NIST database as shown in Ref. [11] for high electron energies down to ~50 eV. As it is known from the original work [15], the same formalism also yields a correct limit for low electron energies (below a few eV, due to a strong static screening). Potential problems can arise only in the intermediate energy range between a few eV and ~50 eV. In this region, however, we use the same cross section assuming that possible deviations should not be too strong.

Propagation of electrons generated in an ion track is modeled even-by-event, accounting for the secondary impact ionizations leaving holes in deep and valence shells, and/or elastic scattering of electrons on lattice atoms. Within this manuscript the term 'elastic scattering' refers to the exchange of kinetic energy only, without ionization of the target atoms, while 'inelastic' refers to an ionization event. The transferred energy ΔE_e (and the corresponding zenith scattering angle θ) of an ionized electron are calculated from the differential cross section in accordance with the energy and momentum conservations [11]. The azimuth angle of scattering φ is chosen randomly between 0 and 2π. Auger decays of deep-shell holes are also modeled.

In this paper we do not take into account a possible effect of the potential of the positively charged track core because of its very rapid and nearly complete neutralization (on timescales < 1 fs) [16]. The remaining long-living part, according to Schiwietz et al. [16], gives only a slight shift in the energy spectrum of delocalized electrons generated in a SHI track, thus having a weak effect on their spreading.

The MC procedure is iterated for ~10^4 times to obtain a trustworthy statistical averaging (error < 10^{-4}). The averaged spatial distributions of the electrons concentration and their energy

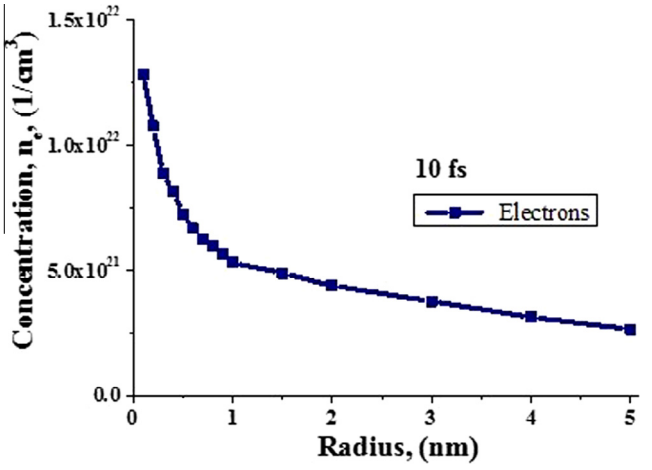

Fig. 1. Radial distribution of the electron concentration in a track of 167 MeV Xe in Al_2O_3, 10 fs after the ion impact.

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