Nuclear Instruments and Methods in Physics Research B 326 (2014) 163–168

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research B

journal homepage: www.elsevier.com/locate/nimb

Excitation and relaxation of olivine after swift heavy ion impact

S.A. Gorbunov [a], N.A. Medvedev [b], R.A. Rymzhanov [c], P.N. Terekhin [d], A.E. Volkov [d,a,c,*]

[a] Lebedev Physical Institute of the Russian Academy of Sciences, Leninskij pr. 53, 119991 Moscow, Russia
[b] Center for Free-Electron Laser Science at DESY, Notkestr. 85, 22607 Hamburg, Germany
[c] Joint Institute for Nuclear Research, Joliot-Curie 6, 141980 Dubna, Moscow Region, Russia
[d] National Research Centre 'Kurchatov Institute', Kurchatov Sq. 1, 123182 Moscow, Russia

ARTICLE INFO

Article history:
Received 29 June 2013
Received in revised form 4 August 2013
Accepted 23 September 2013
Available online 24 January 2014

Keywords:
Swift heavy ion track
Dynamic structure factor
Complex dielectric function
Molecular dynamics
Monte-Carlo
Electron-lattice coupling

ABSTRACT

A multiscale model was developed to describe excitation and initial relaxation of an insulator after an impact of a swift heavy ion (SHI) decelerated in the electronic stopping regime. This model consists of a combination of three methods: (a) Monte Carlo simulations of the nonequilibrium kinetics of the electron subsystem of a solid at the femtosecond scale after the projectile passage. The complex dielectric function (CDF) is used to construct the cross sections for the MC model taking into account a collective response of the electron ensemble to excitation. (b) A molecular-kinetic approach describing further spatial spreading of electrons after finishing of ionization cascades up to hundred femtoseconds. And (c) molecular dynamics simulations of a reaction of the lattice on the excess energy transferred from the relaxing electron subsystem at the picosecond time scale. The dynamic structure factor (DSF) formalism is used to calculate the electron-lattice energy exchange in a general way which is valid for subpicosecond timescales, beyond the phononic approximation of the lattice dynamics. The calculations were performed for 2 GeV Au ion in olivine, demonstrating a heating of the lattice up to 700 K in the nanometric scale picoseconds after the projectile passage.

1. Introduction

A swift heavy ion (SHI, $M > 20$ a.m.u., $E > 1$ MeV/nucl) spends the larger part of its energy (>90%, up to 50 keV/nm along the projectile path) for excitation of the electron subsystem of a solid target in the nanometric vicinity of the ion trajectory (ion track) [1]. This energy loss is accomplished with target ionizations resulting in generation of fast electrons (δ-electrons). Propagation of δ-electrons from the SHI trajectory produces new ionizations forming next generations of electrons and holes. During the subsequent relaxation, a small part of the excess electronic energy is transferred to target atoms resulting in transient lattice excitation in an ion track.

Dissipation of the excess electronic energy in SHI tracks may occur at shorter times than the characteristic time of the atomic vibrations and, thus, than the time of emergence of collective modes of the lattice dynamics (phonons) [2]. Therefore, an adequate description of energy transfer from the excited electron subsystem to the lattice in a SHI track needs a more general approach than that based on the commonly used electron–phonon coupling approximation [3–6]. Such an approach must take into account realization of various limiting cases of the dynamic correlations of lattice atoms during their interaction with the relaxing electron ensemble in the nanometric vicinity of the SHI trajectory.

In this paper we used a microscopic multiscale model [7] to describe the kinetics of the excitation of the lattice of an insulator (an olivine crystal) in a SHI track up to picoseconds after the projectile passage. This model consists of three different approaches combined together.

At the initial stage, the Monte Carlo (MC) approach [8] is applied to simulate the excitation of the electron subsystem of a target. Ionization cascades resulting from scattering of fast electrons generated in a track due to the initial material ionization by an ion, as well as Auger decays of holes at deep atomic shells, are simulated with the MC code up to tens of femtoseconds after the projectile passage. At the longer times (>10 fs), spatial spreading of electrons is modeled with the molecular-kinetic method. The accompanying energy and momentum exchange with the lattice is also calculated. The tracing of the atomic trajectories is realized with the molecular dynamics (MD) method up to picoseconds after the projectile passage.

The above mentioned models rely on the cross sections describing interaction of an ion and fast electrons with the electronic and atomic subsystems of a target. The used approach describing interaction of an incident projectile with a dynamically coupled and

* Corresponding author at: National Research Centre 'Kurchatov Institute', Kurchatov Sq. 1, 123182 Moscow, Russia. Tel.: +7 499 196 91 78; fax: +7 499 196 16 12.
E-mail address: a.e.volkov@list.ru (A.E. Volkov).

0168-583X/$ - see front matter © 2014 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.nimb.2013.09.028

structured system of particles is based on (a) one-particle approximation for the traced projectile; and (b) the assumption of a weak interaction of this projectile with the scattering system (the first Born approximation). In this case, the differential cross section is factored into the cross section of scattering of the projectile on an isolated particle of the ensemble and a term describing a collective response of the coupled scattering system to the transferred energy and momentum [9]. For the SHI track problem the above mentioned approximations are justified for both, (a) an incident ion interacting with the electronic subsystem of a target, and (b) for excited electrons.

Applying this approach, we constructed the cross sections of inelastic scattering of an SHI and fast electrons on the electron ensemble of a target based on the complex dielectric function $\varepsilon(\mathbf{k},\omega)$ formalism [10] (CDF; $\hbar\mathbf{k}$ and $\hbar\omega$ are, respectively, the momentum and the energy transferred from a projectile into the electron subsystem of a target in a collision under consideration, and $\hbar$ is the Plank constant). The CDF takes into account a collective response of the electron ensemble on an excitation of a target and can be restored from the experimentally measured optical coefficients.

The formalism of the dynamic structure factor $S(\mathbf{k},\omega)$ [9] is used to take into account a collective response of lattice to energy and momentum transferred from the relaxing electron subsystem in a SHI track. MD tracing of atoms enables us to calculate the DSF [11] and, thus, realistic cross-sections of the electron-lattice energy exchange on the femto- to pico-second scales. The same MD also traces the subsequent relaxation and spatial dissipation of the excess energy in the lattice. The fluctuation–dissipation theorem [12] states that in a local equilibrium the DSF and CDF formalisms are equivalent (e.g. for the electron ensemble $S_e(\mathbf{k},\omega) = -(\hbar k^2/4\pi^2 e^2 n)\mathrm{Im}\varepsilon_e^{-1}(\mathbf{k},\omega)$.

In contrast to previous multiscale approaches of SHI track modeling [13–17], the present one is relying neither on phonon (low excitation) nor on plasma (extremely high excitation) approximations of the lattice state for the description of electron-to-lattice coupling. The DSF/CDF formalism includes automatically both limiting cases along with all the possible intermediate states.

2. Monte Carlo model of excitation of the electron ensemble in a SHI track

We use a Monte-Carlo model to describe the initial nonequilibrium kinetics of the electron subsystem of a target in the nanometric vicinity of the ion trajectory [8]. At the first step, the model simulates the passage of an ion and calculates its energy losses as well as the spectra of the first generation of highly excited δ-electrons. The energy transferred during a single interaction of an ion with an electron is negligible comparing to the kinetic energy of a projectile due to the large difference between the masses of an electron and an SHI. Therefore, the first Born approximation can be used for the description of scattering of an SHI on the electronic subsystem of a solid. The elastic scattering of the SHI on target atoms can fairly be neglected. Taking these into account, we calculate the cross-sections of scattering of an ion and fast electrons generated in a track by applying the CDF-based formalism [10], where the cross-section can be expressed via the loss function (imaginary part of the inverse complex dielectric function $\varepsilon(\mathbf{k},\omega)$) as [18]:

$$\frac{\partial^2\sigma_{elec}}{\partial(\hbar\omega)\partial k} = \frac{2Z_{eff}^2(\mathrm{v})e^2}{n_e\pi\hbar^2\mathrm{v}^2 k}\mathrm{Im}\left(\frac{-1}{\varepsilon(\mathbf{k},\omega)}\right). \quad (1)$$

Here σ_{elec} is the cross-section of a particle scattering on the electron subsystem; $\mathbf{k} = \mathbf{k}_i - \mathbf{k}_f$ is the change of the wave vector of a scattered particle, $\mathbf{k}_i$ and $\mathbf{k}_f$ are the initial and final wave vectors of a projectile, $k = |\mathbf{k}|$; $\hbar\omega = E_i - E_f$ is the change of energy of particle, E_i and E_f are the initial and final energies of the scattered particle; n_e and m_e are the concentration and the mass of electrons in the scattering system, respectively. Z_{eff} is the equilibrium charge of a particle penetrating through the electronic system (for an incident electron $Z_{eff} = 1$, for an SHI, the dependence of $Z_{eff}(v)$ on the ion velocity can be obtained from the Barkas formula [8]); e is the electron charge.

The loss function restored from the experimental optical coefficients [19,20], can be expressed in the form of a set of the Drude-type CDF for artificial oscillators:

$$\mathrm{Im}\left[\frac{-1}{\varepsilon(\mathbf{k},\omega)}\right] = \sum_{i=1}^{n_{os}} \frac{A_i\gamma_i\hbar\omega}{\left[\hbar^2\omega^2 - (E_{0i} + \hbar^2k^2/(2m_e))^2\right]^2 + (\gamma_i\hbar\omega)^2}, \quad (2)$$

where E_{0i} is the characteristic energy of an oscillator i, A_i is the fraction of electrons with energy E_{0i}, and γ_i is the i-th energy damping coefficient. The summation is running through the all oscillators n_{os}. The procedure of fitting of these coefficients to experimentally available optical data can be found in Refs. [21–23]. The fitted coefficients for olivine are presented in Table 1. The corresponding *ps*- and *f*-sum-rules [21–23] are satisfied well with these coefficients.

The cross-section (1) can be applied to calculate the electron inelastic mean free path ($\lambda = (n_e\sigma)^{-1}$), and the SHI mean energy loss ($-dE/dx$) [7,21–23]. The calculated electron mean free path in olivine is presented in Fig. 1a where the data from the NIST database are also shown for comparison [24]. The calculated energy losses of Au ions of different energies in olivine are presented in Fig. 1b where they are compared to those obtained from the two standard codes: SRIM-2011 [25] and CasP-4 [26]. The good agreement of both dependencies confirms that the fitting coefficients from Table 1 give realistic cross-sections for electron and ion scattering in solid olivine.

Application of Eq. (1) for ion scattering in the MC code supplies us with the number and the spectrum of the initial δ-electons. Propagation of fast electrons generated in the ion track is modeled event-by-event, accounting for the secondary impact ionizations, leaving holes in deep and valence shells, and/or elastic scattering on lattice atoms.

As will be seen later in Fig. 2, concentration of the excited electrons does not exceed 3×10^{22} cm^{-3} even in the very center of the track, which corresponds to the average electron–electron distance of ~3 Å. On such distance, the energy of screened electron–electron interaction U_{e-e} does not exceed 1 eV (see

Table 1
The coefficients of the loss function in the form of a sum of the oscillator functions, Eq. (2), fitted to the experimental optical coefficients of olivine [20]. The total *ps*-sum is 0.89 (11% deviation from 1), while *f*-sum compares very well with the number of electrons N_e.

Peaks	A_i	γ_i	E_{0i}	*f*-sum (N_e)
Phonons	0.00062	0.045	0.07	–
	0.0025	0.033	0.139	
Valence band	5	4	14	31.91 (32)
	92	8	21	
	198	15	28.5	
	290	32	40	
M-shell of Fe	30	7	55	13.95 (14)
	265	60	75	
L-shell of Mg and L-shell of Si	210	40	110	15.96 (16)
	145	75	160	
K-shell of O	215	200	555	7.88 (8)
L-shell of Fe	210	430	720	8.09 (8)
K-shell of Mg	55	200	1317	1.94 (2)
K-shell of Si	52	1100	1860	1.95 (2)
K-shell of Fe	51	6200	7155	2.02 (2)
Total				83.71 (84)