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Effect of valence holes kinetics on material excitation in tracks of swift heavy ions

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ABSTRACT

A considerable part of the excess energy of the electronic subsystem of a solid penetrated by a swift heavy ion (SHI) is accumulated in valence holes. Spatial redistribution of these holes can affect subsequent relaxation, resulting in ionizations of new electrons by hole impacts as well as energy transfer to the target lattice. A new version of the Monte Carlo code TREKIS is applied to study this effect in Al_2O_3 for SHI tracks. The complex dielectric function (CDF) formalism is used to calculate the cross sections of interaction of involved charged particles (an ion, electrons, holes) with the target giving us ability to take into account collective response of a target to excitations.

We compare the radial distributions of the densities and energies of excited electrons and valence holes at different times to those obtained under the assumption of immobile holes used in earlier works. The comparison shows a significant difference between these distributions within the track core, where the majority of slow electrons and valence holes are located at femtosecond timescales after the ion impact. The study demonstrates that the energy deposited by valence holes into the lattice in nanometric tracks is comparable to the energy transferred by excited electrons. Radii of structure transformations in tracks produced by these energy exchange channels are in a good agreement with experiments.

1. Introduction

Swift heavy ions (SHI, $M \geqslant 20\,m_p$, $E > 1$ MeV/amu) decelerated in the electronic stopping regime lose their energy in solids predominantly (>95%) on excitation of the electron subsystem of the target, reaching up to ~40 keV/nm [1]. Subsequent relaxation of the excess energy of the electronic subsystem accompanied by energy transfer to the lattice may result in nanometric structure and phase transformations along the SHI trajectory [2,3].

Initially produced by an SHI impact, δ-electrons spread from the projectile trajectory dissipating the energy they possess on the femtosecond scale [4–6]. Evolution of the spectra of the excited electronic subsystem determines the kinetics of energy transformations resulting finally in excitation of the material and observable effects. Thus, detailed modeling of the transient kinetics of excited conduction-band electrons and valence-band holes in the nanometric vicinity of SHI trajectory is essential for understanding evolution of the target material properties during irradiation.

Recently, we have shown that the kinetics of valence band holes in insulators plays an important role in SHI track creation [7]. It was demonstrated that coupling of only the excited electrons to the lattice does not provide sufficient energy for a detectable structure transformations in SHI tracks. Only when the excess energy of valence band holes was included, the track radii formed in the modeling were similar to those found in experiments [8]. Therefore, the kinetics of valence band holes in relaxing SHI tracks must be studied in more detail. In particular, Ref. [7] as well as the majority of other works (see e.g. [9,10]) did not consider spatial redistribution of valence holes from the SHI trajectory. On the other hand, it was demonstrated in Ref. [11] that diffusion of created valence holes considerably changes their spatial profile and, therefore, the profile of the excess energy they contain, already on the picosecond timescale after an SHI passage.

In the present work we include the valence-band holes transport into the recently developed Monte Carlo code TREKIS [12]. The described approach is applied to Al_2O_3, analyzing kinetics of the exited electron subsystem in SHI tracks. Radial distributions of electrons and holes and their energy densities around the ion trajectory in Al_2O_3 as well as the energy transferred to the lattice

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during the first 100 fs after passage of a Xe 167 MeV ion are calculated. The results are compared with those of application of the previous version of our model which did not include valence holes transport. The radial distribution of the energy transferred into the lattice is used as input data for classical molecular dynamics simulating subsequent relaxation of the lattice and structure transformations appearing near the ion trajectory. The results of MD simulation coincide well with the experimental data.

2. Model

Within the assumed first Born approximation, a cross section of scattering of an incident particle on dynamically coupled system of scattering centers can be factored into a product of the cross section of scattering on a single (isolated) center and the dynamic structure factor (DSF) [13], which takes into account a collective response of a target to an excitation. For scattering of charged particles, the DSF is then expressed in terms of the loss function (inverse imaginary part of the complex dielectric function, $\varepsilon(\omega, \mathbf{q})$) via the fluctuation–dissipation theorem [14,15]. This results in the following form of the differential cross section of a charged particle interaction with a solid [6]:

$$\frac{d^2\sigma}{d(\hbar\omega)d(\hbar q)} = \frac{2[Z_e(v,q)e]^2}{\pi\hbar^2 v^2}\frac{1}{\hbar q}\mathrm{Im}\left[\frac{-1}{\varepsilon(\omega,\mathbf{q})}\right], \quad (1)$$

where $Z_e(v,q)$ is the effective charge of the projectile penetrating through a solid as a function of its velocity, v, and transferred momentum, $\mathbf{q}$ (for an incident electron $Z_e = 1$, for an SHI see the discussion about Z_e in [12]); $\hbar\omega$ is the transferred energy in the considered scattering event; $\hbar$ is the Plank constant; e is the electron charge.

The CDF can be reconstructed from optical data (refractive index n and extinction coefficient k) in the form of a set of artificial oscillators [15]:

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

where the summation is running through all the oscillators n_{os}; coefficients E_{0i}, A_i and γ_i are carefully chosen to reproduce the optical constants of these materials, which can be found in Ref. [16] for low-energy photons (<30 eV), or in Ref. [17] for photons of higher energies. The algorithm finding these coefficients is thoroughly described in Refs. [12,15,18]. Fitted parameters E_{0i}, A_i and γ_i for alumina were published in [4].

This method allows taking into account collective effects occurring in a solid during a charged particle scattering resolved by photo-absorption experiments [15], e.g. plasmon excitation as well as optical part of lattice collective vibrations (phonons) for an electron (or a hole) scattering.

In the present paper, we divide the reconstructed cross sections of the charged particle interaction with the solid into two nearly independent channels: elastic and inelastic scattering. The elastic interaction means the scattering of an incident particle on the target lattice. In contrast, inelastic processes result in ionization or excitation of target atoms generating new free electrons.

Finally, the calculated cross sections are implemented into asymptotic trajectory Monte Carlo code (TREKIS [12]) using the Poisson distribution for the free-flight distance [6,19] and the mean free path of a projectile scattering. The approach describes: (a) penetration of a swift heavy projectile resulting in ionization of a target and appearance of primary fast electrons (δ-electrons); (b) scattering of these fast electrons on lattice atoms and target electrons as well as the kinetics of all secondary generations of electrons arising during relaxation of the electron subsystem; (c) Auger decays, which are also resulting in production of secondary electrons, as well as radiative decays of deep shell holes emitting photons (which is only a small effect for the energies and the material considered here).

During penetration of a charged particle, the target is assumed as homogeneous atom and electron arrangements with the densities corresponding to the solid densities of the materials, and no orientation effects are taken into account, such as channeling of the SHI. No defects or impurities in the material are included in these MC simulations.

Target electrons are considered as uniformly distributed particles occupying either the deep atomic energy levels [20] or the states in the valence or conduction bands according to the density of states (DOS) of a material (Al_2O_3 [21]). Taking into account large velocities of projectiles, we assume these electrons as point-like particles at fixed positions during their energy and momentum exchange with an SHI (instant collisions).

We assume that a scattering event of an incident electron on the target can result in appearance of only one new electron because multiple electron excitations by a single impact is much less probable. The energy transferred in the collision is determined from the differential cross sections given by Eq. (1). Because of the limited energies, neither ions nor the produced electrons manifest relativistic effects, Cherenkov, or Bremsstrahlung emissions. All the secondary electrons appearing in cascades are modeled in the same scheme as the primary ones.

3. Effect of valence holes motion

In the present work we improve our model TREKIS by including spatial redistribution of valence holes and their interaction with a target. This allows to study the effect of valence holes mobility on the kinetics of the material excitation and relaxation in an SHI track. The main goal of this consideration is to establish the role of valence holes redistribution in the processes of energy transfer from electron subsystem into the target lattice.

3.1. Kinetics of valence holes

We consider a hole as an independent point-like quasi-particle, which occupies an energy level E_h in the valence band, distributed according to the density of states of the material, $D(E)$. In order to realize the MC procedure, the knowledge about a dispersion relation $q(E)$ of the particle in the valence band is necessary. $q(E)$ can be determined from the density of states of the material within the 'effective one-band' approximation [22]:

$$D(E) = \frac{\sigma}{2\pi^2}q^2(E)\frac{dq}{dE}, \quad (3)$$

where σ determines the spin factor which is equal to two for the electron system. In the 'effective one-band' model [22], Eq. (3) can be solved analytically for the assumed isotropic momentum:

$$q(E) = \sqrt[3]{\frac{6\pi^2}{\sigma}\int_0^E d\varepsilon \cdot D(\varepsilon)} \quad (4)$$

which results in an average isotropic dispersion relation between the momentum q and the energy E. Introducing an effective mass of a hole, m_h^{eff}, this dispersion relation can be substituted by "free particle" dispersion relation $E_h = \hbar^2 q_h^2/2m_h^{eff}$, where m_h^{eff} now is a function of the hole energy (E_h). Using this effective mass, the velocity of a valence hole at a given energy state can be determined and implemented into Eq. (1) to calculate the partial cross sections of a hole interaction with a material. The differential cross section in Eq. (1) does not depend explicitly on the effective mass m^{eff} of an incident particle, but is inversely proportional to the squared velocity. Considering the dependence of these cross sections on

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