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The microscopic model of material excitation in swift heavy ion tracks

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The paper presents a quantitative model describing the kinetics of the electronic and ionic subsystems of a target along the trajectory of a swift heavy ion (SHI) decelerated in the electronic stopping regime. Electron-to-lattice energy and momentum transfer is described within the dynamic structure factor (DSF) formalism which takes into account different extreme cases of dynamic correlations in the ionic system of a target in a relaxing SHI track. The DSF approach gives an ability to merge Monte-Carlo and Molecular-Dynamics approaches simulating the kinetics of the electron ensemble and the lattice, respectively, into a unified model. This hybrid model predicts a little heating of the lattice in SHI tracks in LiF crystals that agrees well with the experiment-based estimation of this heating. The developed tool can be used for modeling of the kinetics of highly exited material states in swift heavy ion tracks and/or laser spots.

1 Introduction Swift Heavy Ions (SHI, $M > 20$ a.m.u., $E > 1$ MeV/nucl) lose the largest part of their energy (up to 95%, 1 to 10 keV/nm along the ion path) on excitation of the electronic subsystem of a solid in the nanometric vicinity of their trajectories [1].

The subsequent energy and momentum transfer from the excited electronic subsystem of a target to the ionic one in a SHI track can hardly be described in terms of the macroscopic models because of the nanometric spatial and femto- to pico-second temporal scales of the track kinetics. For example, even in the modern models of the electron-lattice interaction, the dynamic state of the ion subsystem is described in the harmonic (phonon) approximation [2-5]. In fact, these models are based on the underlying assumption that the time of phonons emergence is much shorter than the time of interaction of the dynamically correlated atoms with electrons. But dissipation of the excess electronic energy in a track needs much shorter times than the characteristic time of atomic oscillations and, thus, the time of appearance of phonons [6]. Therefore, application of the electron-phonon mechanism to a description of electron-lattice coupling in SHI tracks is questionable, and a more general model is necessary. Such a model should take into account a possibility of realization of various limit cases of dynamic correlations of lattice atoms during their interaction with the non-equilibrium electron ensemble in nanometric SHI tracks at femto- to pico-second time scales.

The microscopic model presented in this paper consists of three different approaches combined together to describe the target kinetics in a relaxing SHI track. A Monte Carlo (MC) approach [7] is used to simulate initial electron excitations/ionizations stimulated by SHI penetration as well as further kinetics of fast electrons appearing due to these ionizations. This MC is also taking care of the secondary electron ionization cascades and Auger-decays of deep holes. Further diffusive electron transport is accounted for by means of molecular-kinetic method tracing temporal and spatial evolutions of the electron density and the energy density of these electrons in a track. The accompanying energy exchange with the atomic system of a target is calculated with help of the dynamic structure factor (DSF) formalism describing the collective response of target atoms to the energy and momentum transfer from the relaxing electron ensemble [8]. The kinetics of lattice atoms in the nanometric vicinity of the ion trajectory is traced with the Molecular Dynamics (MD) method allowing also self-consistent calculations of the DSF of the atomic ensemble.

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The developed model is used to estimate the lattice heating in tracks of gold ions decelerated in LiF in the electronic stopping regime. The obtained little heating (~100 K) agrees well with the experiment-based estimations of track heating in irradiated LiF crystals [9].

2 The DSF based rate of the electron-lattice energy exchange

The differential cross section describing scattering of an electron by a system of dynamically coupled particles is factored within the first Born approximation into the cross section describing scattering of an electron by an isolated particle and the dynamic structure factor (DSF) $S(\vec{k},\omega)$ describing the collective response of the scattering system to the momentum and energy transfer from an electron [8]:

$$\frac{\partial^2\sigma}{\partial\Omega\partial E} = \left|V(\vec{k})\right|^2 \frac{m^2}{4\pi^2\hbar^5}\frac{k_f}{k_i}S(\vec{k},\omega) \tag{1}$$

Here $\vec{k}_i$ and $\vec{k}_f = \vec{k}_i - \vec{k}$ are the initial and final wave vectors of an electron, respectively; $\hbar\omega = \varepsilon_i - \varepsilon_f = (\hbar^2\vec{k}_i^2 - \hbar^2\vec{k}_f^2)/2m$ is a change of the energy of an electron; $V(\vec{k})$ is the Fourier transform of the interaction potential between an electron and a single particle (lattice atom, in our case).

The DSF of the atomic scattering system

$$S(\vec{k},\omega) = \frac{N}{2\pi}\int dt d\vec{r} e^{i(\vec{k}\vec{r}-\omega t)} G(\vec{r},t) \tag{2}$$

is the Fourier transform of the spatial and temporal pair correlation function $G(\vec{r},t)$ describing the coupled atomic dynamics in the system

$$G(\vec{r},t) = \frac{1}{N}\left\langle \sum_{i,j=1}^{N} \int d\vec{r}' \delta(\vec{r} + \vec{R}_i(0) - \vec{r}')\delta(\vec{r}' - \vec{R}_j(t)) \right\rangle \tag{3}$$

Here, N is the number of scattering atoms. The angle brackets denote the quantum mechanical and statistical averaging over an ensemble of eigenstates of the unperturbed Hamiltonian of the scattering system.

The collective response of the scattering system to a transferred energy and a momentum depends on the temporal and spatial scales of a realized interaction [8]. This effect is revealed in the different limit cases of the spatial and temporal dependencies of the correlation function $G(\vec{r},t)$ [6,8].

The classical approximation of this function has a simple form [10] which can be calculated directly by MD [11]:

$$G(\vec{r},t) = \frac{1}{N}\sum_{i,j=1}^{N} \delta(\vec{r} + \vec{R}_i(0) - \vec{R}_j(t)) \tag{4}$$

The following form of the correlation function is used for description of scattering of an electron on a two component system [12]:

$$\begin{aligned} G(\vec{r},t) &= q_1 q_1 G_{1-1}(\vec{r},t) + q_1 q_2 G_{1-2}(\vec{r},t) \\ &+ q_2 q_1 G_{2-1}(\vec{r},t) + q_2 q_2 G_{2-2}(\vec{r},t) \end{aligned} \tag{5}$$

Here q_1 and q_2 are the model charges of the lattice ions (for example, for LiF q_{Li}=1, q_{F}=-1 [13]). The functions $G_{\alpha-\beta}(\vec{r},t)$ describe the spatial and temporal correlations between of α and β atomic species:

$$G_{\alpha-\beta}(\vec{r},t) = \frac{1}{N}\sum_{i=1}^{N\alpha}\sum_{j=1}^{N\beta} \delta(\vec{r} + \vec{R}_i^{\alpha}(0) - \vec{R}_j^{\beta}(t)) \tag{6}$$

To apply efficiently the DSF formalism to the description of electron-lattice coupling, we divide the area of a track into the cylindrical layers of a thickness larger than the mean free path of electrons. We assume a local equilibrium in the electron subsystem in these layers because of the significant differences between the characteristic times of thermalization of electron excitations (10^{-15} sec) [6], and the energy transfer to the lattice (> 10^{-14} sec).

The application of the Thomas-Fermi dielectric function allows us to use approximations which are favorable for application of the DSF formalism: (a) one-electron approximation, considering screened electron-electron interaction when modeling the state of the electron ensemble, and (b) a screened potential for description of the electron-lattice interaction:

$$V(r) = \frac{Ze^2}{r} e^{-\frac{r}{L_S}}, \quad L_S^{-2} = \frac{e^2 m_e}{\pi^2\hbar^2\varepsilon_0}\int f_{\vec{k}} d\vec{k} \tag{7}$$

Here Z is the ionic charge, L_S is the screening length [3], $f_{\vec{k}}$ is the distribution function of electrons over the states characterized by the wave vectors $\vec{k}$, and ε_0 is the vacuum permittivity.

Taking this into account, we assume that the energy exchange between the lattice and electrons in each cylindrical layer is described by the one-electron kinetic equation where the collision integral depends only on the cross sections describing the electron-lattice interaction in the framework of the DSF formalism [14]. The energy transfer rate from the relaxing electron subsystem of a target to the ionic one is defined in this case as:

$$Q = \frac{4}{(2\pi)^5\hbar}\int f_{\vec{k}_i}(1 - f_{\vec{k}_f})\,\omega\, S(\vec{k},\omega)\left|V(\vec{k})\right|^2 d\vec{k}_i d\vec{k}_f \,. \tag{8}$$

3 Combined microscopic approach describing SHI track excitation

3.1 SHI passage and the electron kinetics in a track

A combined model is used to obtain the temporal and spatial dependencies of the distribution function $f_{\vec{k}}(\vec{r},t)$ of electrons in a SHI track.

At the initial stage, the MC simulates the excitation of the electronic subsystem by a projectile and by fast electrons generated due to ionizations. This MC code has already been tested in experiments investigating the ionization kinetics in SHI track cores in silica [7]. The physical background and details of the numercis used in the MC model can be found in Ref. [7].

At the first step, MC simulates interaction of a SHI with electronic subsystem of a target supplying with the electronic energy losses of the ion as well as with the parameters of the ensemble of the first generation of excited free electrons.

Then, propagation of these electrons is modeled even-by-event, accounting for the secondary impact ionizations, leaving holes in deep and valence shells, and/or elastic scattering on lattice atoms. Production of secondary free electrons due to Auger decay of deep holes is also taken into account in the MC model. Created secondary electrons are modeled in the same manner. The processes of electron spreading and hole decays are traced until times when electron behavior turns from a ballistic to a diffusive one [15] (~10 fs after the projectile passage).

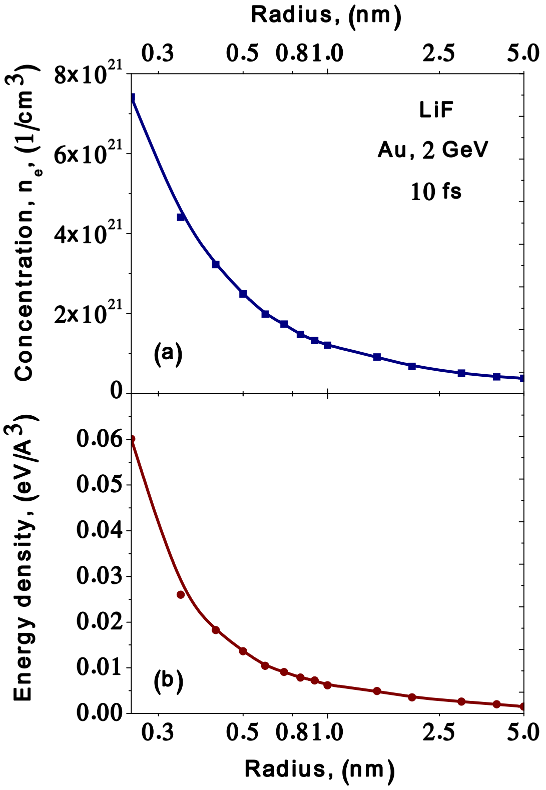

Figure 1 The radial distributions of the electron density (a) and the energy density of electrons (b) in a track of 2 GeV Au ion in LiF.

The MC procedure is iterated for 10^4 times to obtain a trustworthy statistics. The averaged radial distributions of the concentration of electrons and their energy density are then extracted and used as the initial conditions for further modeling. Figure 1 presents these radial distributions obtained with the MC simulations in the layer of thickness of 10 nm along the ion trajectory in a LiF crystal after 10 fs from the ion passage.

Molecular-kinetic method is then used to describe further diffusion of electrons and the energy they accumulated after the time when electrons started to demonstrate the diffusive behavior in the MC simulations. Since all the directions of motion of electrons are equivalent at this stage, the average number of electrons, passing through the area ΔS in time Δt from g^{th} to $(g+1)^{\text{th}}$ cylindrical layer, is

$$\Delta N_{g\to g+1} = \frac{1}{6} n_g < v_g > \Delta S \Delta t \cdot \tag{9}$$

Here n_g is the concentration of electrons in the g-th layer, $<v_g>$ is their average speed.

The knowledge about the current values of the concentration of electrons n_g and their energy density E_g in a g-th layer gives an ability to restore the parameters of the equilibrium Fermi distribution of electrons in this layer, the chemical potential (μ_g) and the temperature (T_g), from the implicit relations in the free-electron approximation [16]:

$$\begin{aligned} n_g &= \frac{2m_e^{3/2}}{\sqrt{2}\pi^2\hbar^3}\int_0^\infty \left[\exp\left(\frac{\varepsilon-\mu_g}{k_B T_g}\right)+1\right]^{-1} \times \sqrt{\varepsilon}\,d\varepsilon, \\ E_g &= \frac{2m_e^{3/2}}{n_g\sqrt{2}\pi^2\hbar^3}\int_0^\infty \left[\exp\left(\frac{\varepsilon-\mu_g}{k_B T_g}\right)+1\right]^{-1} \times \varepsilon^{3/2}\,d\varepsilon \end{aligned} \tag{10}$$

Here k_B is the Boltzmann constant and m_e is the electron mass Eqs. (9) and (10) supplies with the information about temporal variations of the electron concentrations and temperatures characterizing the electron ensembles in any g-th layer.

3.2 MD tracing of atomic dynamics The original MD code is applied to calculate the DSF (Eqs.(2,4,5)) governing the energy transfer from the excited electron subsystem to the ionic one. The code uses the Verlet algorithm with the time-step of 0.5 fs to trace the atomic dynamics for over 40000 atoms. A modified Tosi-Fumi interatomic potential [13], well suiting for LiF, is used in this code.

The kinetic temperature [11, 17, 18] characterizing the non-equilibrium atomic system in a chosen volume is used to evaluate the lattice heating in a track:

$$T_{kin}(\vec{r},t) = \frac{1}{3k_B(N_a-1)} M_a \sum_n (\vec{v}_n - \vec{v}_{mean})^2 \tag{11}$$

Here N_a is the number of particles in the volume. M_a and $\vec{v}_n$ are the mass and velocities of atoms, respectively; $\vec{v}_{mean}$ is the average velocity of atoms in chosen volume. Summation is running over all the atoms contained in the volume. The kinetic temperature coincides with the thermodynamic one in the equilibrium [17].

3.3 Combined model for simulations of the electron kinetics and the atomic dynamics in relaxing SHI tracks (applied to LiF) Simulation of the energy transfer from electrons to the lattice, electron diffusion and the ionic conductivity in the nanometric vicinity of the SHI trajectory is realized within the following scheme:

1. The local equilibrium distribution functions of electrons, obtained out of the MC and molecular-kinetic simulations (Eqs. (9) and (10)), are used to describe the

initial states of the electron subsystem in the cylindrical layers with the thicknesses of 0.5 nm.

2. Then, the rates of the energy exchange between the electronic subsystem and the lattice in these cylindrical layers during a current time step are calculated by Eq. (8) taking into account the DSF of the atomic ensemble.

3. After that, the energy losses of the electronic subsystem in each layer during this time-step are determined taking into account the calculated electron-lattice energy exchange rates there.

4. Next, changes of the concentration and of the energy density of electrons in the layers due to electron diffusion are determined.

5. Then, the equilibrium electron distribution functions corresponding to the modified electron energy density and concentration are restored in each cylindrical layer.

6. These new functions are substituted into the next step of the modeling.

7.) The procedure is repeated over again.

The excess energy transferred from electrons to the lattice in a cylindrical layer at each time step is distributed among the different types of atoms according to their mass fractions by increasing their kinetic energy.

During the cooling down of the electrons in a SHI track (< 100 fs) the dynamics of the atomic system does not change significantly. Therefore, in the present work, the same DSF was used at each step of the model in Eq. (8).

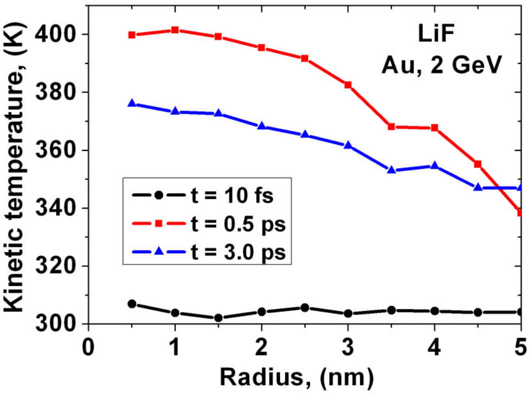

Figure 2 The radial dependence of the kinetic temperature of the LiF lattice at different times after the passage of a 2 GeV gold ion.

The temporal dependence of the radial distribution of the kinetic temperature of LiF lattice in the nanometric vicinity of the trajectory of 2 GeV Au ion is shown in Fig. 2. This figure demonstrates that the largest part of the excess energy transferred into the lattice is localized within the distances $r < 3$ nm from the ion trajectory. The maximum lattice heating in this area does not exceed 100 K which is in a good agreement with the experimental results reported in [9]. This agreement proves an applicability of the presented model to description of material excitation in SHI tracks.

4 Conclusion

A microscopic model was developed which describes all the stages of material excitation in a SHI track: excitation of the electronic subsystem by a penetrating ion, secondary ionizations by fast electrons generated in a track, the kinetics of electrons and deep holes, an energy and momentum transfer from the excited electron subsystem into the ionic subsystem of a target, and the subsequent lattice dynamics. The results of application of this model to LiF crystals irradiated with 2 GeV Au ion demonstrated the upper limit of the lattice heating (~100 K) in a good agreement with the experimental results.

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