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KINETICS OF ELECTRONS FROM PLASMA DISCHARGE IN A LATENT TRACK REGION INDUCED BY SWIFT HEAVY ION IRRADIATION

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Abstract

While passing swift heavy ion through a material structure, it produces a region of radiation affected material which is known as a "latent track". Scattering motions of electrons interacting with a swift heavy ion are dominant in the latent track region. These phenomena include the electron impurity and phonon scattering processes modified by the interaction with the ion projectile as well as the Coulomb scattering between two electrons.

In this paper, we provide detailed derivation of a 3D Boltzmann scattering equation for the description of the relative scattering motion of such electrons. Phase-space distribution function for this non-equilibrioum system of scattering electrons can be found by the solution of mentioned equation.

Key words

swift heavy ions, electronic scattering, latent track, Boltzmann equation, phonon scattering, electron scattering-in/out rate

1. INTRODUCTION

The heavy ions irradiation is of interest for the development of specific structures in the irradiated material. Nanostructured materials provide the opportunity for tailoring physical, electronic, and optical properties for a variety of technological applications, including advanced nuclear energy systems. There is an ever-increasing demand for radiation-tolerant materials that can withstand the extreme radiation environments in nuclear reactors, accelerator-based nuclear systems, and nuclear waste forms. Understanding radiation effects in materials with unique nanostructures is an urgent challenge, since it may hold the key to unlocking the design of tailored materials for advanced nuclear energy systems (1).

In ion beam modification of materials, application of energy of accelerated ions below MeV is considered as the standard ion beam technique. Energy of ions from “high energy implanters” is restricted by their maximum accelerated voltage that usually does not exceed the value of 500 kV. Higher ion implantation energies are achievable mostly by MV range tandem ion accelerators. Obligatory schemes for basic research, over 1 MeV/amu, often referred to asswift heavy ions, are increasingly used for materials modification and synthesis. The typical application range of swift ions is of the order of several hundred MeV to some GeV. At low energies (keV), kinetic energy is effectively transferred to the atomic structure through the nuclear stopping cross section, target atoms are directly displaced from their lattice sites, and defects are produced via atomic collision cascades. For swift heavy ions, electronic energy loss can be more than two orders of magnitude higher than nuclear energy loss, and nuclear energy loss is, therefore, negligible (see scheme in Fig.1). Swift heavy ions (SHI) first transfer their energy to the loosely bound target electrons initiating a cascade of secondary electrons that spreads radially within femtoseconds (1). The mentioned time intervals is much shorter than the time necessary to create defects via lattice relaxation (i.e. 10^{-17} to 10^{-14} s). It has been found that the energy deposited by heavy ions passing through or near the device structure, produces high density plasma of electrons along the particle trajectory. This plasma thermalizes in about 1 ps and the associated electron–hole pairs can be collected by a drift or diffusion in a few hundreds of picoseconds. After some time delay, the energy is gradually transferred into atomic motion through electron-phonon coupling within sub-picoseconds, thereby initiating a thermal spike that can modify the atomic structure of the target. Therefore SHI induce the formation of damaged regions by electron interactions. Along the path of swift ions, the so called “latent tracks” of a diameter of a few nanometers and depths of a few micrometers are formed, which can be visualized by etching (2). SHI irradiation provides new unique methods of materials modification.

There are several models describing this process. Coulomb repulsion of suddenly ionized atoms within the track volume leads to the so-called “acoustical shock” and acoustical excitations are generated in this case. "Ion explosion" model as a probable mechanism of acoustical excitation arising in a solid state during heavy swift ion irradiation was proposed in (3). The so-called “thermalspike” model (4) was developed for the description of the heavy ion track damage creation in metals and oxides (5). The model is based on the existence of a shortliving plasma state in the track volume. It seems, that in using thermal spike model, the radii of damaged region where lattice temperature acceedes the melting point, correlates with the etched nanopore size (6). The thermodynamics of "hot electrons" in the plasma along the heavy swift ion trajectory and relative scattering motion of electrons are studied in the latest works by calculating the non-equilibrium electron distribution (7).

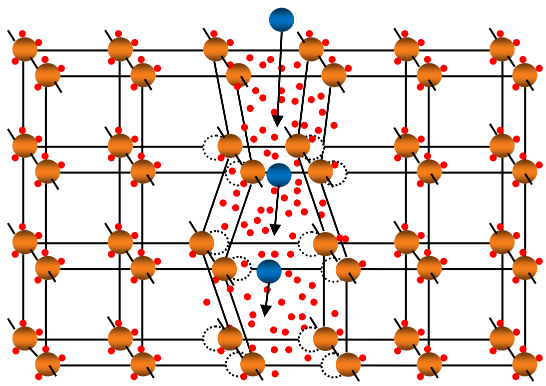

Fig.1 *Formation of latent track in electronic stopping regime.* ● *Heavy ion ,* • *electron*

SHI with energies higher than ~ 1 MeV/amu and masses higher than ~ 20 proton masses stimulate the structural and phase transformations in vicinities of a few nanometers of their trajectories when penetrating various solids. These effects occur in the electronic stopping regime, when the electronic energy loss of a projectile overcomes a threshold (~ 2−5 keV/nm in dielectrics). The radiation damage produced by elastic recoils is orders of magnitude too low to provide the observed structural modifications in tracks (8, 9). However, the processes of creation of radiation defect as well the critical values of ion energy loss have not been explained completely yet. A general model of radiation damage processes based on the mechanisms of elementary excitations has not yet been formulated.

At intermediate transit energies where electronic and nuclear energy losses are both significant (from 0,5 to 50 MeV, or from 1 to a few hundreds of keV/amu), synergistic, additive or competitive processes may affect the dynamic response to irradiation. In recent years, the importance of these ionization effects and the coupled processes has become increasingly recognized for both metals and ceramics (10). The design of radiation tolerant materials, control of materials modification, and creation of defect structures to tailor materials properties demand a comprehensive understanding and predictive models of energy transfer and exchange processes. Understanding the coupling of electronic and atomic processes in this intermediate energy regime is critically important to predict and control material properties in many energy-related technologies (1).

SHI is a novel technique for modification of materials at the molecular and electronic level. This type of irradiation can modify the structure of material in a controlled way leading to changes in their chemical, electronic, electrical, tribological and optical properties (11,12). SHI irradiation deposits the energy in the material in the near surface region mainly due to the electronic excitation (13). SHI interactions with materials bring about remarkable structural, conformational and morphological changes, which result in enhancement in their performance and properties such as conductivity, electrochemical stability, sensitivity, crystallinity, solubility, porosity, density etc.

The emphasis in this contribution is given to the question of plasma electron scattering in the latent track region along ion trajectory, which plays a determining role in the phenomena associated with such radiation.

2. CONTINUITY EQUATION IN THE PHASE-SPACE

A swift heavy ion penetrating a solid predominantly interacts with the electrons of the substrate giving rise to the so-called electronic energy loss. Within 10^{-16} s, this interaction causes a high density of electronic excitation and ionization several nanometres around the ion trajectory. Depending on the degree of excitation and the specific nature of the solid, this electronic excitation may be transformed into atomic motion via different processes.

The continuity equation describes how the density of system changes with time. Non-eqilibrium system of electrons arises in plasma discharge when the heavy swift ion passes through the irradiated material structure. It is necessary to know the distribution function for these "hot" electrons in the area of the radiation damage for statistical analysis of the whole process. Continuity equation for electrons in the standard 6-dimensional phase space can be written in the next form:

$$\int_{(\Sigma)} \vec{j} \cdot d\vec{S} = -\frac{\partial N}{\partial t} \quad , \qquad [2.1]$$

where $\vec{j}$ is the vector of electrons flow in the phase space, N is the number of electrons inside an enclosed integration area Σ and f is non-equilibrium phase-space distribution function for electrons in the latent track region. Distribution function f (i.e. probability density function) depends on the set of six coocrdinates $\{x_i\}=\{x_1, x_2, x_3, x_4, x_5, x_6\}$, i.e. $f = f(\{x_i\})$. The mentioned set of independent generalized electron coocrdinates in classical case obviously consists of componens of position and momentum vectors:

$$\vec{r} = [x,y,z], \ \vec{p} = [p_x, p_y, p_z]. \qquad [2.2]$$

However, this set of classical coocrdinates $\{x,y,z,p_x\ p_y\ p_z\}$ is characterized by dimensional inhomogeneity. Homogenisation of these coordinates is important for physically correct calculations and it can made formally in the following simple way:

$x_1=\alpha x$, $x_2=\alpha y$, $x_3=\alpha z$, $x_4=\beta p_x$, $x_5=\beta p_y$, $x_6=\beta p_z$,
[2.3]
where α and β are constants with the appropriate physical dimensions. Electrons flow vector can be written as:

$$\vec{j}=\vec{v}f \ , \qquad [2.4]$$

where $\vec{v}$ is velocity of electron in phase space and the number of electrons inside an enclosed integration area N obey:

$$N=\int_{(\Gamma)} f dV \ \text{, i.e.} \quad \frac{\partial N}{\partial t}=\int_{(\Gamma)} \frac{\partial f}{\partial t} dV \ . \qquad [2.5]$$

If we consider [2.5] and [2.6] in continuity equation [2.1] and apply Gauss theorem, we get:

$$\int_{(\Gamma)} Div(f\vec{v})dV=-\int_{(\Gamma)} \frac{\partial f}{\partial t}dV \ \text{, thereof :} \ Div(f\vec{v})=-\frac{\partial f}{\partial t} \ . \qquad [2.6]$$

In [2.6], we integrate over the volume of the ara enclosed inside the Σ . 6-dimensional divergence calculated in the phase-spase is on the left side of the equation [2.6]. It can be calculated as:

$$Div(f\vec{v})=fDiv(\vec{v})+\vec{v}\cdot Grad(f)=f\sum_{i=1}^{6}\frac{\partial \dot{x}_i}{\partial x_i}+\sum_{i=1}^{6}\dot{x}_i\cdot\frac{\partial f}{\partial x_i}, \quad \text{where} \ \dot{x}_i=\frac{dx_i}{dt} \ . \qquad [2.7]$$

Formally, the same calculation procedure as in the 3-dimensional case can be applied, i.e.:

$$Div(f\vec{v})=f\frac{\partial(\alpha v_x)}{\partial(\alpha x)}+f\frac{\partial(\alpha v_y)}{\partial(\alpha y)}+f\frac{\partial(\alpha v_z)}{\partial(\alpha z)}+f\frac{\partial(\beta \dot{p}_x)}{\partial(\beta p_x)}+f\frac{\partial(\beta \dot{p}_y)}{\partial(\beta p_y)}+f\frac{\partial(\beta \dot{p}_z)}{\partial(\beta p_z)}+$$

$$+\alpha v_x\frac{\partial f}{\partial(\alpha x)}+\alpha v_y\frac{\partial f}{\partial(\alpha y)}+\alpha v_z\frac{\partial f}{\partial(\alpha z)}+\beta\dot{p}_x\frac{\partial f}{\partial(\beta p_x)}+\beta\dot{p}_y\frac{\partial f}{\partial(\beta p_y)}+\beta\dot{p}_z\frac{\partial f}{\partial(\beta p_z)}$$

and equation of continuity in phase space takes the form:

$$Div(f\vec{v})=f\nabla_{\vec{r}}\cdot\vec{v}+f\nabla_{\vec{p}}\cdot\dot{\vec{p}}+\vec{v}\cdot\nabla_{\vec{r}}f+\dot{\vec{p}}\cdot\nabla_{\vec{p}}f \ . \qquad [2.8]$$

Because it applies:

$$\frac{\partial x_i}{\partial x_j}=\delta_{ij} \ \text{ then } \ \nabla_{\vec{r}}\cdot\vec{v}=\frac{1}{m}\nabla_{\vec{r}}\cdot\vec{p}=0 \ . \qquad [2.9]$$

In addition, we can consider:

$$\nabla_{\vec{p}}\cdot\dot{\vec{p}}=\nabla_{\vec{p}}\cdot\vec{F} \quad \text{and} \quad \dot{\vec{p}}\cdot\nabla_{\vec{p}}f=\vec{F}\cdot\nabla_{\vec{p}}f \qquad [2.10]$$

where $\vec{F}$ is total force acting on plasma electron. Consequently, the 6-dimensional divergence can be rewritten to the next form:

$$Div(f\vec{v})=f\nabla_{\vec{p}}\cdot\vec{F}+\vec{v}\cdot\nabla_{\vec{r}}f+\vec{F}\cdot\nabla_{\vec{p}}f \qquad [2.11]$$

Subsequently, the continuity equation (2.8) is:

$$f\nabla_{\vec{p}}\cdot\vec{F}+\vec{v}\cdot\nabla_{\vec{r}}f+\vec{F}\cdot\nabla_{\vec{p}}f=-\frac{\partial f}{\partial t} \ . \qquad [2.12]$$

Actually, only the Lorentz type force depends on the momentum of electron. But for this force, we can consider:

$$\nabla_{\vec{p}}\cdot\vec{F}=q\nabla_{\vec{p}}\cdot(\vec{v}\times\vec{B})=\frac{q}{m}\nabla_{\vec{p}}\cdot(\vec{p}\times\vec{B})=\frac{q}{m}(\nabla_{\vec{p}}\times\vec{p})\cdot\vec{B} \quad \text{and} \quad \nabla_{\vec{p}}\times\vec{p}=\vec{0} \, . \qquad [2.13]$$

Therefore:

$$\nabla_{\vec{p}}\cdot\vec{F}=0 \qquad [2.14]$$

and equation (2.12) can be written as follows:

$$\vec{v}\cdot\nabla_{\vec{r}} f+\vec{F}\cdot\nabla_{\vec{p}} f=-\frac{\partial f}{\partial t} \qquad [2.15]$$

The force acting on plasma electrons along the latent tracks is the superposition of the forces generated by external fields and collision forces:

$\vec{F}=\vec{F}_{field}+\vec{F}_{coll}$,

Therefore, continuity equation (2.15) passes to the next form of Boltzmann kinectic equation:

$$\vec{v}\cdot\nabla_{\vec{r}} f+\vec{F}_{field}\cdot\nabla_{\vec{p}} f+\vec{F}_{coll}\cdot\nabla_{\vec{p}} f=-\frac{\partial f}{\partial t} \ . \qquad [2.16]$$

Just collision forces have the effect of electrons scattering in the latent track region.

In kinetic theory (14), the evolution of electron population is driven by the collisions undergone with a fixed hackground of field particles and the electron distribution function is governed by differential Boltzmann equation [2.16]. A very famous and important particular case in this field is the so-called Lorentz gas (15), which corresponds to vanishingly small ratio between electron (as a test particle) and the field particle masses. The matter is assumed to be composed of two sub-systems in interaction: the electrons described within the quasi free electron gas model and the atoms following the Debye model.

3. COLLISION INTEGRAL

Scattering processes generated by collision forces $\vec{F}_{coll}$ can change the distribution function *f*. In the semi-classical case, the next formulas can be considered in equation [2.16]:

$$\vec{p}=\hbar\vec{k}\ ,\quad \vec{v}=\frac{\hbar\vec{k}}{m} \qquad [3.1]$$

and scattering process can be effectively described in the *k*-space. *m* is effective mass of electron. Scattering-out and scattering-in processes can be distinguished during the collision of plasma electron in the latent track region. In the scattering-out process, electrons are scattered from the point $\vec{k}$ into the volume $d\vec{k}'$ around any $\vec{k}'$ in the *k*-space. State $\vec{k}$ is occupied with probability $f\left(\vec{k},\vec{r},t\right)$ and state $\vec{k}'$ is available with probability $1-f\left(\vec{k}',\vec{r},t\right)$. Then the number of electrons scattering-out from state $\vec{k}$ per unit time can be obtained as:

$$dn_{out}=Nw\left(\vec{k}\to\vec{k}'\right)f\left(\vec{k},\vec{r},t\right)\left[1-f\left(\vec{k}',\vec{r},t\right)\right]\frac{d\vec{k}'}{\left(2\pi\right)^3}\ , \qquad [3.2]$$

where $w\left(\vec{k}\to\vec{k}'\right)$ is the probability of electron scattering-out from the state $\vec{k}$ to $\vec{k}'$. Thus the total number of electrons scattered out from the state $\vec{k}$ during the infinitesimal time interval *dt* is given by the sum over all possible $\vec{k}'$:

$$dN_{out}=N\left(df\right)_{out}=-Ndt\int w\left(\vec{k}\to\vec{k}'\right)f\left(\vec{k},\vec{r},t\right)\left[1-f\left(\vec{k}',\vec{r},t\right)\right]\frac{d\vec{k}'}{\left(2\pi\right)^3}\ . \qquad [3.3]$$

The minus sign shows that this quantity describes the loss of electrons in state $\vec{k}$. In addition to the scattering-out of the domain $d\vec{k}$, there also exist scattering processes leading to a gain of electrons in volume $d\vec{k}$ in *k*-space. In this scattering-in process, electrons are scattered from the volume $d\vec{k}'$ around any $\vec{k}'$ into the $\vec{k}$ if this state is not occupied. The total number of scattered-in electrons during the infinitesimal time interval *dt* is:

$$dN_{in}=N\left(df\right)_{in}=Ndt\int w\left(\vec{k}'\to\vec{k}\right)f\left(\vec{k}',\vec{r},t\right)\left[1-f\left(\vec{k},\vec{r},t\right)\right]\frac{d\vec{k}'}{\left(2\pi\right)^3}\ . \qquad [3.4]$$

As can be seen from [3.3] and [3.4], the change of the distribution function caused by collisions of electron is:

$$\left(\frac{\partial f}{\partial t}\right)_{coll} = \left(\frac{\partial f}{\partial t}\right)_{out} + \left(\frac{\partial f}{\partial t}\right)_{in} = \vec{F}_{coll} \cdot \nabla_{\vec{p}} f \; . \qquad [3.5]$$

After substituting the right-hand sides of equations [3.3] and [3.4] to the formula [3.5], we get the collision integral:

$$\left(\frac{\partial f}{\partial t}\right)_{coll} = \left[1 - f\left(\vec{k},\vec{r},t\right)\right]\int w\left(\vec{k}' \to \vec{k}\right) f\left(\vec{k}',\vec{r},t\right) d\vec{k}' - f\left(\vec{k},\vec{r},t\right)\int w\left(\vec{k} \to \vec{k}'\right)\left[1 - f\left(\vec{k}',\vec{r},t\right)\right] d\vec{k}' \; . \qquad [3.6]$$

The collision term [3.6] is correlation function of *f*, so it involves an integral where *f* appears twice in the integrand, albeit at different positions or momenta. This collision term is nonlinear, which makes is difficult to solve the Boltzmann kinectic equation [2.16].

4. SEMI-CLASSICAL BOLTZMANN SCATTERING EQUATION FOR PLASMA ELECTRONS IN LATENT TRACK REGION

Because both impurity atoms and lattice ions do not move with the electron center-of-mass, electrons inside a drifting system feel that impurities and ions are oscillating against them due to the Galilean principle of relative motion. This leads to impurity and phonon assisted photon absorption in the system. The relative scattering motion of electrons can be very well described by a Boltzmanns cattering equation (16).

Boltzmann transport equation without a drift term we get from the equation [2.16]:

$$\vec{F}_{field} \cdot \nabla_{\vec{p}} f + \vec{F}_{coll} \cdot \nabla_{\vec{p}} f = -\frac{\partial f}{\partial t} \qquad [4.1]$$

We can define electron scattering-in ($\Lambda_{\vec{k}}^{(in)}$) and electron scattering-out ($\Lambda_{\vec{k}}^{(out)}$) rates by means of collision integrals:

$$\Lambda_{\vec{k}}^{(in)} = -\int w\left(\vec{k}' \to \vec{k}\right) f\left(\vec{k}',\vec{r},t\right)\frac{d\vec{k}'}{\left(2\pi\right)^3} \quad , \quad \Lambda_{\vec{k}}^{(out)} = -\int w\left(\vec{k} \to \vec{k}'\right)\left[1 - f\left(\vec{k}',\vec{r},t\right)\right]\frac{d\vec{k}'}{\left(2\pi\right)^3} \; . \qquad [4.2]$$

As can be seen from [3.5] and [3.6], collision term can be written as:

$$\vec{F}_{coll} \cdot \nabla_{\vec{p}} f = \Lambda_{\vec{k}}^{(out)} f - \Lambda_{\vec{k}}^{(in)}\left(1 - f\right). \qquad [4.3]$$

and Boltzmann transport equation without a drift term [4.1] goes into the form:

$$\frac{\partial f}{\partial t} = \Lambda_{\vec{k}}^{(in)}\left(1 - f\right) - \Lambda_{\vec{k}}^{(out)} f - \vec{v} \cdot \nabla_{\vec{r}} f - \vec{F}_{field} \cdot \nabla_{\vec{p}} f \; . \qquad [4.4]$$

Therefore, the non-equilibrium distribution of electrons from plasma discharge along latent tracks formation f is governed by quasi-classical Boltzmann kinectic equation:

$$\frac{\partial f}{\partial t} = \Lambda_{\vec{k}}^{(in)}\left(1 - f\right) - \Lambda_{\vec{k}}^{(out)} f - \frac{e}{\hbar}\left(\vec{E} + \vec{v} \times \vec{B}\right) \cdot \nabla_{\vec{p}} f \; , \qquad [4.5]$$

where $\vec{k}$ is electron wave vector, *e* is electrical charge of electron $\vec{E}$ is intensity of electircal field and $\vec{B}$ is magnetic induction. In absence of external electromagnetic field, the Boltzmann scattering equation [4.5] taking into account impurity- and phonon-assisted photon absorption and Coulomb electron scattering is:

$$\frac{\partial f}{\partial t} = \Lambda_{\vec{k}}^{(in)(\beta)}\left(1 - f\right) - \Lambda_{\vec{k}}^{(out)(\beta)} f \quad , \; \beta = \text{(impurities), (phonons), (Coulomb potential).} \qquad [4.6]$$

The scattering rates $\Lambda_{\vec{k}}^{(in)(\beta)}$ and $\Lambda_{\vec{k}}^{(out)(\beta)}$ are electron scattering-in and scattering-out rates due to phonons, including phonon-assisted photon absorption which can be obtained after solving the Schrodinger equation for the single electron wave-function in the presence of the ion potential.

5. PRELIMINARY CONCLUSIONS

In this paper, the kinetics of electrons in swift heavy ion irradiated materials was studied by using generalized Boltzmann scattering equation. This equation includes scattering of electrons with phonons and impurities beyond the relaxation-time approximation. The effect of an "incident" ion potential is reflected in modifying the scattering of electrons with impurities and phonons. This drives the distribution of electrons away from the thermal equilibrium distribution to a non-equilibrium one. The electron average kinetic energy increases with the "strength" of the ion potential and "hot electrons" are creating (depending on the magnitude of impact parameter and the charge number of the ion projectile).

Boltzmann scattering equation for description of the relative scattering motion of electrons interacting with a swift heavy ion by including both the impurity- and phonon-assisted photon absorption processes as well as the Coulomb scattering between two electrons was derived. The modification of electron-phonon scattering can result from the electrons interacting with the ion potential through phonon scattering. The question is the solution of mentioned Boltzmann scattering equation. The Chapman-Enskog theory (17-20) is a way to linearize the Boltzmann equation using a perturbation expansion for non-equilibrium distribution function f for some small parameter.

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