Strongly Coupled Nonequilibrium Nanoplasma
Generated by a Fast Single Ion in Solids

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Received 15 April 2009, accepted 30 June 2009
Published online 08 October 2009

Key words Heavy ions, plasma diagnostics, X-ray spectra, plasma relaxation.
PACS 52.27.Gr, 52.50.Gj, 52.65.Yy, 52.70.La

A plasma model for relaxation of a medium in heavy ion tracks in condensed matter is proposed. The model is based on three assumptions: the Maxwell distribution of plasma electrons, localization of plasma inside the track nanochannel, and constant values of the plasma electron density and temperature during the X-ray irradiation. It is demonstrated that the plasma relaxation model adequately describes the X-ray spectra observed upon interaction of a fast ion with condensed target. Preassumptions of plasma relaxation model are validated by the molecular dynamics modeling and simulation.

1 Introduction

Beams of fast ions with certain parameters can be obtained from electro-magnetic accelerator as well as from plasma of short laser pulses (see for ex. [1–4]). Due to absorption features of ion beams, they are widely used for a number of important technical [5, 6] and medical [7, 8] applications. The interaction of a single fast heavy ion with condensed matter leads to the formation of a track. On the initial stage of track formation the excited channel is created with transverse size of about 1 nm as it is illustrated in Fig. 1. This stage can be studied experimentally by measuring X-ray spectral lines generated by radiative decay in autoionized states of multiply charged ions of the target material. These ions appear due to multiple ionization of the target atoms by the Coulomb field of the projectile ion. Some of the target atoms have vacancies in the K shell. The observable X-ray spectrum is emitted as a result of radiative transitions from higher shells (L, M, \ldots) to the K shell.

The simplest model to interpret X-ray spectra is based on the assumption that the relaxation of different ions is independent of each other as if these ions were isolated [9–16]. In this case the intensity of the X-ray spectral line generated by the ion with spectroscopic symbol Z is proportional to the corresponding multiple ionization cross section multiplied by the branching factor $A^Z/(A^Z+1)$, where $A^Z$ is the radiative transition probability and $1/A^Z$ is the autoionization probability. In what follows we recall this model as ‘atomic relaxation’ one.

The atomic relaxation model relates the observed X-ray spectra only to the interaction of the projectile ion with individual target atoms whereas the excited state of the target medium is not taken into account. In other words the atomic relaxation model completely ignores the fact that the plasma-like environment arising after the initial ionization enables a wider range of relaxation processes for the excited ions. For example, owing to the collisions with free electrons, the initially excited $Z$ ion can either be ionized or recombine before filling the K vacancy upon the radiative transition. It means that the initial excitation of the $Z$ ion could actually result in formation of ions with different charge states with K vacancies and, hence, could lead to emission of spectral lines of ions with $Z' \neq Z$. In this case, the observed X-ray spectrum should reflect not only the characteristics of the (projectile ion) - (target atom) interaction but also the parameters (temperature and density) of the plasma formed.

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2 Plasma relaxation model

The conclusion about plasma creation inside the track of a fast heavy ion was drawn quite long time ago [17–19]. Moreover it was noted there that the plasma was strongly coupled (nonideal). Nevertheless, only recently [20] the correspondence between the plasma parameters and the observed X-ray spectra was considered. It was done in the context of the so-called 'plasma relaxation' model. This model is based on the solution of time-dependent equations of the radiative-collisional kinetics while the initial state of the excited medium is described by the conventional atomic relaxation model.

Plasma relaxation model proposed in present work allows to describe the excitation of solids by single fast ions with total energies of $0.1 \div 1.0$ GeV. This model is based on: 1) the conclusion that the solid-density nanosize plasma is created in the area of projectile-target atom interaction [17–19,25]; 2) the solution of the time-dependent equations of the collisional-radiative kinetics to describe the evolution of an initially created excited state. Three time stages of the formation of nonequilibrium strongly coupled solid-state nanoplasma in the region of the track are considered.

1) At the initial stage ($\sim 10^{-2}$ fs) the state of the medium is described by the conventional atomic relaxation model [9–16]. The initial channel of ionized matter is produced with the transversal diameter of several nanometers and electron density of $n_e \sim 10^{23}$ cm$^{-3}$ (for ion charges $Z = 2–6$). Ions with K-shell vacancies are localized in the vicinity of the channel axis. Although their fraction is relatively low, they are responsible for the appearance of X-ray emission spectra.

The initial target state is defined by the multiple ionization of target atoms by a projectile. We consider solids consisted of atoms with nuclear charges $\sim 10–18$, i.e. having fully populated K and L shells and several electrons in M-shell. Particularly, this are the cases of quartz and aluminum solid targets that were investigated experimentally [21–23].

Initially, both free electrons and different multicharged ions with single K-shell vacancy, $n$ L-shell vacancies and fully ionized M-shell are produced in the area of a single fast ion track. The populations $N_n$ of the states with $n$ vacancies in the L shell are proportional to the cross sections $\sigma_n$ of the multiple ionization of the target atom by the field of the projectile ion. In the atomic relaxation model, the multiple ionization cross section, as a rule (see, for example, [9–13]), is determined using the quasi-classical impact-parameter method. According to this method, the multiple ionization cross section can be written in the form:

$$\sigma_n = \sigma(1s) \sum_{k_1+k_2=n} \frac{2!}{k_1!(1-k_1)!} \frac{6!}{k_2!(8-k_2)!} p_{2s}(0)^{k_1} p_{2p}(0)^{k_2} (1-p_{2s}(0))^{(2-k_1)} (1-p_{2p}(0))^{(6-k_2)}$$

(1)

where $\sigma(1s)$ is the cross section of ionization of the 1s electron, $p_{2s}(0)$, $p_{2p}(0)$ are the probabilities of ionization of the 2s or 2p electron at collision with the zero impact parameter, correspondingly, $n = k_1 + k_2$, $k_1$ and $k_2$ are the number of vacancies in the 2s and 2p subshells of the ion created ($n = 0$ for autoionizing states of fluorine-like ions, $n = 1$ for autoionizing states of oxygen-like ions, etc.). Thus, in order to describe the initial...
conditions, it is sufficient to know the probabilities $p_{2s}(0)$ and $p_{2p}(0)$, which are the functions of the projectile ion charge, the target atom nuclear charge, and the projectile ion velocity. Unfortunately, the exact calculation of these quantities is a very complex problem. Furthermore no published data are available on the collisions of $\text{Ni}^{14+}$ ions with the silicon atom or $\text{Mg}^{7+}$ ions with the aluminum atom. Hence it will be assumed below that $p_{2s}(0) = p_{2p}(0) = p_L(0)$ where $p_L(0)$ is the first free parameter of our model.

The duration of the first stage of plasma formation at each track point is actually determined by the time of the multiple ionization of the target atom and can be estimated as $\tau_1 \sim a/v$, where $a$ is the atomic size and $v$ is the projectile ion velocity. This duration varies in the range of $\tau_1 = 10^{-3} - 10^{-2}$ fs for the experimental conditions considered.

2) At the second stage ($\leq 1$ fs) the Maxwell distribution of free electrons is established with the temperature of tens of eV. The electric double-layer with thickness of about interatomic distance is formed around the nanoscale initial channel.

Relaxation processes at second stage were studied by the nonequilibrium molecular dynamics (MD) simulations [27, 28]. The initial structure of ions in the cylinder cross-section is shown in the insertion to Fig. 2. Six ions with charge $Z = 2$ are located at vertices of the hexahedron and the seventh one with $Z = 4$ is in the center of the cell. The cell is periodically extended along $Z$-axis with the period $a$ equals to the edge of hexahedron. Although this is relatively rough approach to the real ion structure in $\text{SiO}_2$, it is appropriate to estimate the required relaxation times and describe the evolution of free electron density and velocities in the central track area.

![Fig. 2](image)

As the plasma is neutral in large, spreading of the electron cloud produces positive charge in the track center and double electric layer on the track surface. The ion motion can be neglected when studying the dynamics of electrons. The stable radial density profiles of electrons obtained by MD simulations for two cases of $T_e = 25$ eV and $T_e = 14$ eV are shown in Fig.2. The first peak at $r = a$ corresponds to the outer ion shell. For both values of the temperature the equilibration time turned out to be about 0.5 fs. It follows from our calculations that the number of electrons remaining in the area $r < 1.5a$ is not less than 85 percent of the initial density. These electrons form the plasma around radiating ions which is dense enough for the collisional relaxation of the ions. Another simulations which account for the nonzero permittivity of the surrounding media ($\varepsilon = 2.25$) result in similar values.

To consider relaxation of the electron velocity distribution function we start simulations from an artificially prepared initial state where all electrons have the same absolute value of the velocity $v_{th} = (3k_B T_e/m_e)^{1/2}$ (see Fig.3). Direction of the velocities is random. Calculations for $T_e = 14$ eV demonstrate that the distribution at $t = 0.3$ fs fits perfectly Maxwellian one for the considered velocity range. This value can be taken as the equilibration time for the electron velocity distribution function. For $T_e = 25$ eV the relaxation time is $t = 0.25$ fs. All these times are lower than the time of X-ray K-shell spectra irradiation by an order of magnitude. So one can assume that the plasma is in equilibrium during the whole time of spectroscopy data acquisition in [21–23].
3) Further evolution (tens of fs) of initially created charge state distribution for the ions with K-shell vacancies is described by the time depended equations of collisional-radiation kinetics:

\[
\frac{dN^Z_k(t)}{dt} = \sum_{m,Z'} K^{Z'\rightarrow Z,k}_{km} N^Z_m(2)
\]

where \( N^Z_k(t) \) is the \( k \)-state population of ions with the spectroscopic symbol \( Z \), and \( K^{Z'\rightarrow Z,k}_{km} \) is the kinetic matrix depending on the plasma density and temperature. The off-diagonal elements of matrix \( K^{Z'\rightarrow Z,k}_{km} \) are the total probabilities of \( (m, Z') \rightarrow (k, Z) \) transitions due to all elementary processes, while the diagonal elements are equal to the total decay probability of \( (k, Z) \)-state with the opposite sign. As the relaxation of excited states occurs in plasma, it is necessary to take into account all the processes occurring due to collisions of ions with free electrons, i.e. collisional excitation and de-excitation, collisional ionization, three-body and radiative recombination, dielectronic capture.

![Fig. 3](image)

**Fig. 3** Distribution of free electron velocities normalized by thermal velocity \( v_{th} = (3k_B T/m)^{1/2} \) with \( T_e = 14 \text{ eV} \) at 0.005 fs (a), 0.3 fs (b). The initial distribution is \( f(v) = \delta(v - v_{th,0}) \). The solid line indicates the Maxwell distribution for \( T_e = 3E_0/k_B \), where \( E_0 \) is the mean kinetic energy of electrons; \( v \) is the distribution step.

### 3 Time-dependent kinetics for X-ray emission spectra simulation

In the most cases the system of equations (2) is solved using the so-called quasi-stationary approximation [27], where the time derivatives for excited levels are assumed to be zero. It provides the solution only for ground states while the populations of excited states are determined from the solution of the substantially simpler system of algebraic equations. This method can be used when the relaxation rates of the excited levels are considerably higher than the relaxation rates of the ground states. The above approximation is not adequate in the case of solid-state plasmas. Therefore the system of equations (2) is solved for the ground and excited (autoionized) states jointly in the present work.

We use the following assumptions, which made possible to decrease the number of equations, thus essentially simplifying the problem. First, the system of equations (2) includes only ground configurations of the ions and their lowest-lying autoionized configurations. Second, it is assumed that the populations of closely spaced levels of an ion are proportional to their total statistical weights. It is related to the fact that the probabilities of collisional transitions between these levels in solid-state plasmas exceed the probabilities of the radiative and autoionized transitions. As a result only one level (namely \( 1s2l^{(n+m)} \)) is introduced instead of the great number of terms related for example to the configurations \( 1s2s^m2p^{2m+1}L_{(2J+1)} \). The simplified scheme of energy
levels, for which the system of equations (2) has been solved, is shown in Fig.4. The kinetic matrix accounts for the following processes in this case: the ionization by an electron impact, three-body recombination, photorecombination, radiative decay, autoionization, and dielectron capture. It is believed that the plasma temperature and density, which determine the X-ray emission, remain unchanged in the course of relaxation processes.

Fig. 4 The scheme of levels and atomic processes taking into account in the plasma model of track relaxation: radiative decay (dashed lines), autoionization (dotted line), dielectronic capture (dash-dotted lines), collisional ionization (thin solid lines), three-body recombination (thick solid lines), and collisional deexcitation (double solid line). Ion charge states from H-like to F-like are taken into account.

Fig. 5 Populations of Si ion autoionization states in dependence on time in plasma at $T_e = 100$ eV for different ion charge states (F-like, O-like etc.)
The values of atomic constants (such as the energies of electronic levels and the probabilities of radiative and autoionized transitions) and the rates of collisional transitions are taken from [24,29]. The solution of the system of the equations (2) makes it possible to follow the relaxation of the initially generated state with the K vacancy in the $Z_0$ ion.

The examples of the calculation results are shown in Fig.5, where the time dependences of the relative populations of autoionizing levels are shown for silicon ions with the different charge states. The redistribution of the excitation among the different charge states turns out to be very significant in both cases. It justifies the importance of the plasma relaxation model. The solution of the system of equations (2) with the initial values of $N_k^Z(0)$ defined from atomic relaxation model makes it possible also to calculate the emission spectrum observed during the passage of the heavy ion beam through the condensed medium. Denote by $I^Z$ the total intensity of the array of the unresolved satellite transitions caused by the radiative decay of the autoionizing states of an ion with the spectroscopic symbol $Z$. It is emitted during the lifetime of the plasma $\tau_{\text{plasma}}$. The values of $I^Z$ are calculated from the formula

\[ I^Z = \int_0^{\tau_{\text{plasma}}} N_k^Z(t) A_k^Z dt, \]

where $A_k^Z$ is the probability of the radiative decay of the autoionizing state $k$. As it is seen from Fig.5 the establishment of equilibrium populations for the autoionizing states takes about 100 fs, but the relaxation for the most abundant ions is completed in 10-20 fs. Then the populations of their autoionizing states become relatively low. As the intensity of spectral line is proportional to the population of autoionizing states, the main contribution to X-ray radiation of plasma results from the times $\tau = 10 \div 20$ fs, when the plasma is not only extremely nonequilibrium but also strongly non-stationary. Accordingly, the value of $\tau_{\text{plasma}} = 20$ fs is used in our spectra simulations. It is necessary to consider the group of dielectronic satellite lines for every ion charge state $Z$. Such group consists of a number of radiation transitions (up to 40 for C-like Si ion), which are concentrated inside a narrow spectral range of about 0.05 Å. Usually these transitions are unresolved and registered as a spectral peculiarity with the overall profile width defined by the distribution $g^Z A^Z(\lambda)$ rather than by broadening of individual lines. To simulate emission spectra we take the spectral functions describing the shape for a group of unresolved transitions from the experiments [21–23]. The samples of the modeling spectra for different values of plasma temperature $T_e$ are presented in Fig. 6. It is seen that the spectra are strongly temperature sensitive and consequently they can be used to determine the plasma electron temperature.

![Emission spectra of Si multicharged ions calculated with use of atomic and plasma relaxation models. Simulations are made for $T_e = 5$ eV, 20 eV and 200 eV. The value of parameter $p_L(0) = 0.34$ was chosen for every cases here. Spectra are shifted along Y-axes.](image)
4 Determination of plasma nanochannel parameters using X-ray spectra

Similar results were obtained for the interaction of fast heavy ions with condensed targets (solid or aerogel) were carried out recently at the linear heavy ion accelerator facility UNILAC (GSI, Darmstadt, Germany) [21–23]. Among other, the interaction of Ni$^{+14}$ and Mg$^{+7}$ fast ions with low (0.15 g/cc) bulk density quartz aerogel and solid Al targets was investigated, correspondingly. Enough thickness of the target sample provided the full internal stopping of the projectile ions. The use of the aerogel target allowed to expand the track length up to 2 mm which provided the spatial resolution along the track for registered spectra. Also it was shown in [21–23] that the inner nanoscale structure of aerogel did not influence stopping and media excitation processes in other respects.

In the experiments [21–23] the ions with the energy of 11.4 and 5.9 MeV/amu were focused into the 2 mm spot at the edge of a target. The target was exposed during 2-3 hours by a beam current of 0.2 ± 0.5 μA. Such a low current provided the average time interval of 10 ps between single ion propagation. Thus, the measured spectrum might be considered as an average over the independent acts of a single heavy ion interaction with the cold target matter and burning the germ of a track. The X-ray spectra of the excited target matter were registered by means of FSSR spectrometers based on spherically bent crystal dispersion element [24]. The spectral ranges 6.1 - 7.3 Å and 7.9 - 8.3 Å were embraced by use of quartz ($2d = 8.512$ Å) and mica ($2d = 19.938$ Å) crystals, respectively. Simultaneously, the dielectronic satellites of the resonance line for every charge state of the Si and Al target ion were registered in each of the cases. The spectra were registered with 1D spatial resolution along the direction of the projectile ion propagation. Examples of the spectra observed are presented in Fig. 7.

![Fig. 7 Examples of Si (a) and Al (b) X-ray spectra excited by different fast ions with different mass and energy stopped in solid targets, measured in [21–23].](image)

The modeling spectra are compared with those observed experimentally [21–23] for the interaction of projectile Ni$^{+14}$ ions with the quartz aerogel in Fig.8. We see that although only two fitting parameters $p_L(0)$ and $T_e$ are used, the experimental data and plasma model simulation results are in good agreement for all 6-8 satellite groups whereas even the best fit for the former atomic relaxation model fails to describe the experimental results (dotted lines in Fig.8). Analogous results have been obtained for the interaction of projectile Mg$^{+7}$ ions with the solid Al target.
Fig. 8 Comparison of experimental spectra (black solid lines) obtained in [21–23] with modeling results. The spectra were radiated by Si ions inside aerogel SiO$_2$ target excited by Ni projectile ions with energy of 11 MeV/amu (a) and 3 MeV/amu (b). The modeling according the plasma model (thick solid lines) is done for $T_e = 14$ eV, $p_L(0) = 0.33$ (a) and $T_e = 25$ eV, $p_L(0) = 0.34$ (b). For the atomic model simulation (dotted lines) the values of parameter $p_L(0) = 0.335$ (a) and $p_L(0) = 0.375$ (b) were chosen.

The results of the proposed plasma relaxation model are in good agreement between the simulations and the experimental data for the X-ray K-shell spectra of solid quartz and Al targets excited by 100 - 600 MeV single ions. It confirms the validity and self-consistency of the plasma relaxation model and allows to propose the X-ray spectroscopy method for plasma temperature measurement in the fast heavy ion track.

Spectra simulation using plasma relaxation model permits to obtain the values of $p_L(0)$ as well. They change very weakly from 0.33 to 0.34 as the projectile ion energy decreases from 11 to 3 MeV/amu. The values of $p_L(0)$ and their energy dependence for the interaction of Ni$^{14+}$ ions with Si targets has not been described before.

In contrast to $p_L(0)$, the plasma temperature increases significantly with projectile ion slowing down to $\sim 3$ MeV/amu – from 14 to 25 eV for silicon excited by Ni(58) projectiles, and from 25 to 40 eV for aluminum excited by Mg(26) projectiles. This effect illustrates the fact that the linear energy loss of the projectile ion is increasing along with the stopping process from 11 to 3 MeV/amu [23, 25].

On the last step, the electron-electron nonideality parameter $\Gamma = (4\pi n_e/3)^{1/3} e^2/(k_BT_e)$, the formal number of electrons in the Debye sphere $N_D = 4\pi r_D^3 n_e/3$, $r_d = (k_BT/4\pi n_e e^2)^{1/2}$ and the degeneracy parameter $\Theta = N_e(k^2/mT_e)^{3/2}$ were calculated employing obtained values of plasma density and temperature. By this way the estimations of $\Gamma = 0.43 \pm 1.3$, $N_D = 0.13 \pm 0.69$, $\Theta = 0.03 \pm 0.18$ were done. So, it allows us to conclude that the electron gas in heavy ion track in solids is non-degenerate and essentially strongly coupled (nonideal). By other words, the strongly coupled plasma nanochannel is created on the time scale of tens of femtoseconds after single fast ion propagates through solid media.

5 Conclusion

The plasma relaxation model is considered to describe the excitation of solids by single fast ions with total energies of 0.1-1.0 GeV. Three time stages of the formation of nonequilibrium strongly coupled (nonideal) solid-state nanoplasma are studied in the region of the track.

The state of the medium is described by the conventional atomic relaxation model at the initial stage ($\sim 10^{-2}$ fs). The initial channel of ionized matter is produced with the transversal diameter of several nanometers and electron density of $n_e \sim 10^{23}$ cm$^{-3}$ ($Z = 2 - 6$). Ions with K-shell vacancies are localized in the vicinity of the channel axis. Although their fraction is relatively low, they are responsible for the appearance of X-ray emission spectra.
Maxwell distribution of free electrons is established at the second stage ($\leq 1$ fs) with the temperature of tens of eV. The electric double layer with thickness of about interatomic distance is formed around the nanoscale initial channel. Further evolution (tens of fs) of initially created charge state distribution for the ions with K-shell vacancies is described by the time-dependent equations of collisional-radiation kinetics. The ions are stable in the crystal lattice so that the band structure is retained (though disturbed) while the matter is still deeply ionized.

The nanoplasma lifetime is defined by recombination rates. As it follows from kinetic calculations, $t_{\text{plasma}}$ is about several hundreds fs. Extremely low beam current of 0.2-0.5 $\mu$A was used in the experiments considered. It corresponds to 10 ps time interval between single ions in the flow. It means that each fast ion interacts with cold solid matter. In the case when time interval between single ions is about (or smaller) plasma lifetime, the plasma relaxation model has to be improved taking into account hydrodynamic expansion, density and temperature gradients.

X-ray K-shell spectra of solid quartz and Al targets excited by 100 - 600 MeV single ions [21–23] are explained with a good agreement between the proposed model simulation and the experimental data. Both the agreement and molecular dynamics modeling confirm the validity and self-consistency of the plasma relaxation model. For the experimental conditions the electron density and temperature are found to be equal to $4 \cdot 10^{23}$ cm$^{-3}$ and 10-50 eV respectively. Thus the number of electrons in the Debye sphere is in the range of $0.14 \div 0.32$, so the plasma is strongly coupled.

The X-ray spectroscopy method is proposed to measure plasma temperature in the fast multi-charged ion track. The obtained results can be used to study further stages of heavy ion track evolution: the melting of overheated ionic lattice in the presence of hot electrons, two-temperature hydrodynamics of track expansion, shock wave propagation, recombination, cooling, the formation of a latent track and finally a change in the properties of the material with accumulation of tracks.

Acknowledgements The authors appreciate B. Doyle, H. Hjalmarson, J. Aidun, M.M. Basko, B.M. Smirnov, and V.S. Vorobyev for prolific discussions and suggestions. The work was partly supported by RFBR Grant No.07-08-00738-a, ISTC Grant No.3504, Programs of Fundamental Research No.12 and No.27 of the Presidium of RAS and Federal Special Program Vorobyev for proli...