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Electron–lattice coupling after high-energy deposition in aluminum

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ABSTRACT

This paper presents an analysis of the parameters of highly-excited electron subsystem of aluminum, appearing e.g. after swift heavy ion impact or laser pulse irradiation. For elevated electron temperatures, the electron heat capacity and the screening parameter are evaluated. The electron–phonon approximation of electron–lattice coupling is compared with its precise formulation based on the dynamic structure factor (DSF) formalism. The DSF formalism takes into account collective response of a lattice to excitation including all possible limit cases of this response. In particular, it automatically provides realization of electron–phonon coupling as the low-temperature limit, while switching to the plasma-limit for high electron temperatures. Aluminum is chosen as a good model system for illustration of the presented methodology.

1. Introduction

High energy deposition into a solid by swift heavy ions decelerated in the electronic stopping regime (SHI, $M > 20\,m_p$, $E > 1$ MeV/amu, m_p is a proton mass) allows to achieve extreme levels of excitation of its electron subsystem. The temperature of the electron ensemble can rise up to several Fermi energies in the nanometric vicinity of the ion trajectory (SHI track) at the femto- to pico-second timescale after an ion passage [1,2]. Similar levels of electronic excitations are reached on the micrometer scale during irradiations of solids with femtosecond free-electron lasers (FEL) [3–6]. Subsequent relaxations of the excited electron subsystem results in energy and momentum transfer into the lattice that may lead to unusual nanometric structural and phase transformations in an irradiated material [1–6].

Taking into account fast and large increase of the temperature of delocalized electrons appearing after high energy deposition, the Two Temperature (Thermal Spike) Model (TTM, TSM) is often used to describe possible lattice heating by the ensemble of hot electrons [1,2]. The relative "simplicity" of TTM provides its popularity in the society [1,2].

However, macroscopic models or fitting procedures are often applied to determine the key parameters of TTM, e.g. the dependence of the electron–lattice coupling factor on the temperature of the ensemble of delocalized electrons. Taking into account the ultrashort temporal and spatial scales of TTM in SHI tracks, such choosing of the parameters may stimulate reasonable doubts in results of application of the model to simulations of lattice excitation (see, e.g. [7,8] or [9,10]).

This paper is aimed to supply researchers using TTM with these parameters calculated rigorously. Aluminum was chosen as a model system for demonstration of these dependencies. The main efforts are focused on the dependence of the electron–lattice coupling factor on the temperature of the ensemble of delocalized electrons generated in a SHI track. The electron–phonon mechanism is often used to describe electron lattice coupling in SHI tracks/laser spots. However, the time of cooling of the electronic subsystem of a solid in a nanometric SHI track is shorter than, or on the order of, the characteristic time of atomic oscillations in a lattice [7–10]. This makes an application of the electron–phonon mechanism questionable for the description of interaction of hot electrons with a lattice in a SHI track. We compare results of applications of the electron–phonon approximation with the general formulation of electron-lattice coupling based on the dynamic structure factor (DSF) formalism [11]. DSF takes into account in a quantitative way effects of all the spectrum of spatial and temporal correlations in the atomic dynamics on lattice excitation. In particular, it automatically provides realization of electron–phonon mechanism as the low-temperature limit, while switching to the plasma-limit for high electron temperatures.

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The molecular dynamics (MD) procedure is developed to calculate the DSF and to simulate the kinetics of lattice excitations at the sub-picosecond timescales as well as to obtain the dependence of the electron–lattice energy transfer rate on the electron temperature. Additionally, the other necessary parameters for the TTM, namely the electron heat capacity and the screening parameter as functions of electron temperature are obtained taking into account the realistic density of states of solid aluminum.

2. Electron–lattice energy transfer rate, electron heat capacity and screening in aluminum

Due to the screening effect and high Fermi energy, the kinetic energies of conduction-band electrons in aluminum are larger than the energies of their interaction, and the one-electron approximation can be used to describe the state of this electron ensemble. The rate of electron–lattice energy exchange, Q_{e-i}, is given by the moment of the one-electron kinetic equation [9]:

$$Q_{e-i} = \frac{\hbar^3}{2\pi^3 m_e^2} \int f_{\mathbf{k}_i}(1 - f_{\mathbf{k}_f}) \hbar\omega \frac{k_i}{k_f} \frac{\partial^2 \sigma}{\partial\Omega \partial(\hbar\omega)} d\mathbf{k}_i d\mathbf{k}_f. \quad (1)$$

Here σ is the cross section of an electron scattering on the ion subsystem; $f_{\mathbf{k}}$ is the distribution functions of electrons ($f_{\mathbf{k}} = f_{\mathbf{k}}^{eq}$ is the Fermi function in case of local equilibrium); Ω is the solid angle of electron scattering; $\mathbf{k}_i$ and $\mathbf{k}_f$ are the initial and final wave vectors of an electron; $\hbar\omega = \frac{\hbar^2\mathbf{k}_i^2}{2m_e} - \frac{\hbar^2\mathbf{k}_f^2}{2m_e}$ is the change of the energy in the free-electron approximation, which works very well for aluminum; m_e is the free-electron mass.

The first Born approximation is applicable when describing coupling of a lattice with the excited ensemble of delocalized electrons in a SHI track [10]. Within this approximation, the differential cross section is factored into the cross section of electron scattering on an isolated atom and the "charge–charge" dynamic structure factor (DSF) of a target. DSF describes effects of spatial and temporal correlations in positions and dynamics of lattice atoms on electron scattering [11]:

$$\frac{\partial^2 \sigma}{\partial\Omega \partial(\hbar\omega)} = |V(\mathbf{k})|^2 \frac{m_e^2}{4\pi^2\hbar^5} \frac{k_f}{k_i} S(\mathbf{k}, \omega). \quad (2)$$

Here $V(\mathbf{k})$ is the spatial Fourier transform of the interaction potential between an electron and a single atom of a target; $\mathbf{k} = \mathbf{k}_i - \mathbf{k}_f$ is the change of the wave vector of a scattered electron.

The Fourier transform of the atomic spatial and temporal pair correlation function $G(\mathbf{r}, t)$ determines the DSF [11]:

$$S(\mathbf{k}, \omega) = \frac{N}{2\pi} \int dt d\mathbf{r} \exp[i(\mathbf{k}\mathbf{r} - \omega t)] G(\mathbf{r}, t). \quad (3)$$

were N is the number of scattering atoms.

In the classical approximation of the lattice dynamics, $G(\mathbf{r}, t)$ is reduced to the simple form [12]:

$$G(\mathbf{r}, t) = \frac{1}{N} \left\langle \sum_{i,j=1}^{N} \delta(\mathbf{r} + \mathbf{R}_i(0) - \mathbf{R}_j(t)) \right\rangle. \quad (4)$$

where $\mathbf{R}_i(0)$ is a coordinate of an i-th atom at the initial time, $\mathbf{R}_j(t)$ is a coordinate of a j-th atom at the time instance t, $\langle \ldots \rangle$ is a statistical averaging over the atomic ensemble. The following correction should be introduced into the classical DSF $S_{cl}(\mathbf{k}, \omega)$ in order to satisfy the necessary quantum–mechanical asymmetry [13–16]:

$$S(\mathbf{k}, \omega) = \frac{(\hbar\omega/T_l)}{1 - \exp[-(\hbar\omega/T_l)]} S_{cl}(\mathbf{k}, \omega). \quad (5)$$

where Tl is the lattice temperature

Combining Eqs. (1)–(5), the following formula for the energy transfer rate can be obtained:

$$Q_{e-i} = -\frac{4}{(2\pi)^5\hbar^2} \int d\mathbf{k}_i d\mathbf{k}_f E_{\mathbf{k}_i} |V(\mathbf{k})|^2 \left[f_{\mathbf{k}_f}^{eq}(1 - f_{\mathbf{k}_i}^{eq}) S(-\mathbf{k}, -\omega) - f_{\mathbf{k}_i}^{eq}(1 - f_{\mathbf{k}_f}^{eq}) S(\mathbf{k}, \omega) \right], \quad (6)$$

where $E_{\mathbf{k}} = (\hbar^2\mathbf{k}^2)/2m_e$ is the energy of an electron. The Fermi function $f_{\mathbf{k}}^{eq}(T_e)$ is used to describe the electron ensemble at different temperatures.

Eq. (6) demonstrates that the calculation of the energy transfer rate needs the electron temperature dependencies of the interaction potential (screening length $L_S(T_e)$) and the chemical potential of the electron ensemble. Realization of TTM model also requires knowledge about such dependencies of the electron heat capacity, $C_e(T_e)$, and the electron heat conductivity. The last one is beyond the scope of the present paper.

The density of states (DOS, $D(E)$) of the ensemble of delocalized electrons in a material determines the forms of these dependencies. We took the realistic DOS of aluminum from [17], where it is normalized to 3 electrons per aluminum atom.

Below we present the dependencies of the electron heat capacity and the screening lengths on the temperature of the electron ensemble in the conduction band of aluminum. The temperature dependence of the chemical potential $\mu(T_e)$ we calculated coincides exactly with that given in [17].

The electron heat capacity is determined by:

$$C_e(T_e) = \int_0^\infty \frac{\partial f(E, T_e, \mu(T_e))}{\partial T_e} D(E) E dE. \quad (7)$$

Numerical solution of Eq. (7) gives the temperature dependence of $C_e(T_e)$ presented in Fig. 1. This dependence is close to that obtained from DOS of a free electrons gas. Deviations between these dependencies occur at low temperatures, where effects from peculiarities of DOS structure are more pronounced. The electron heat capacity in [17] underestimates the temperature dependency of $C_e(T_e)$.

The screened Coulomb potential (Yukawa potential [18]) was chosen to describe the interaction of an electron with a lattice atom:

$$V(r) = Z\frac{e^2}{r} e^{-r/L_S}. \quad (8)$$

Here e is the electron charge, $Z{\cdot}e$ is the charge of a lattice ion, L_S is the screening length determined according to [19–21] as:

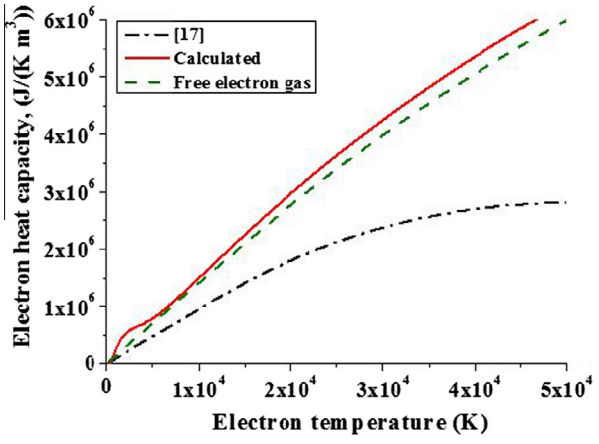

Fig. 1. Calculated electron heat capacity in aluminum vs those calculated in [17] and free electron gas approximation.

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