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A ballistic transport model for electronic excitation following particle impact

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ABSTRACT

We present a ballistic model for the transport of electronic excitation energy induced by keV particle bombardment onto a solid surface. Starting from a free electron gas model, the Boltzmann transport equation (BTE) is employed to follow the evolution of the temporal and spatial distribution function $f(\vec{r},\vec{k},t)$ describing the occupation probability of an electronic state $\vec{k}$ at position $\vec{r}$ and time t. Three different initializations of the distribution function are considered: i) a thermal distribution function with a locally and temporally elevated electron temperature, ii) a peak excitation at a specific energy above the Fermi level with a quasi-isotropic distribution in k-space and iii) an anisotropic peak excitation with k-vectors oriented in a specific transport direction. While the first initialization resembles a distribution function which may, for instance, result from electronic friction of moving atoms within an ion induced collision cascade, the peak excitation can in principle result from an autoionization process after excitation in close binary collisions. By numerically solving the BTE, we study the electronic energy exchange along a one dimensional transport direction to obtain a time and space resolved excitation energy distribution function, which is then analyzed in view of general transport characteristics of the chosen model system.

1. Introduction

The excitation of the electronic degrees of freedom following the impact of a keV particle onto a solid surface manifests in three different experimentally accessible observables: the formation of secondary ions within the flux of particles released ("sputtered") from the surface [1], the emission of electrons into the vacuum ("external" electron emission) [2,3] and the flux of excited charge carriers through a buried internal energy barrier realized, for instance, by the insulating film of a metal-insulator–metal junction ("internal" electron emission) [4]. In order to understand the results of such measurements and arrive at a prediction of those quantities, we proposed a model to calculate the excitation of electronic degrees of freedom resulting from electronic friction of moving particles and autoionization following close binary collisions in a particle impact-induced atomic collision cascade [5–7]. As one of the essential ingredients of such a model, the rapid transport of excitation energy away from the spot of its generation was described in terms of a diffusive approach involving a nonlinear diffusion equation, where the electron energy diffusivity was coupled to the local and temporal lattice disorder, yielding a four-dimensional excitation energy density profile which may then be parametrized in terms of an elevated time and position dependent electron temperature. The resulting electron temperature profiles were then employed to obtain external electron emission yields by means of a slightly modified Richardson-Dushman approach [8,9] or to assign ionization probabilities to each sputtered atom according to the so-called substrate excitation model [10].

In contrast to the rather successful calculation of *external* electron yields, a straightforward adoption of the thermionic emission approach to calculate *internal* electron emission yields turned out to give results underestimating experimental data by orders of magnitude [11]. This discrepancy may be explained in terms of the large electron mean free paths within the first femtoseconds after the projectile impact. Thus, the applicability of the diffusion model must be put into question especially at that particular time interval, which coincides with the period where most of the (internal and external) electron emission is assumed to take place.

Therefore, it was concluded that the ballistic nature of the transport process probably has to be taken into account within the model. One attempt was done in terms of a hybrid model [12] combining diffusive and ballistic transport of the excited electrons. First, for a set of layers in different depths below the surface, electron temperatures are calculated according to the standard diffusion model. Then, each layer is regarded as a source of hot electrons according to the Richardson-Dushman

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model. These hot electrons are now assumed to undergo a ballistic transport, which is phenomenologically described via an effective mean free path λ for inelastic energy losses preventing them from being able to traverse the buried tunneling barrier. Depending on the traveling length x towards the barrier, the probability to arrive at the metal–insulator interface and contribute to the internal emission yield is then described by $\exp(-x/\lambda)$. With this approach, it was shown that the calculated internal electron emission yields are of the right order of magnitude as measured experimentally. However, this hybrid model is not capable of treating directional effects as induced, for instance, via a variation of the projectile angle of incidence, arising from the extremely anisotropic excitation spectra found in [13].

In order to tackle this problem and take the treatment of electronic excitation transport one step further, the present work employs the classical Boltzmann transport equation (BTE) enabling a more fundamental investigation of the ballistic transport characteristics of electronic excitation energy generated in the first few femtoseconds after the projectile impact. Different initial distribution functions $f(\vec{r},\vec{k},t=0)$ for the occupation probability density of an electronic state with wave number $\vec{k}$ at position $\vec{r}$ and time t are implemented representing different excitation mechanisms such as electronic friction (generating a thermal-like distribution [14–17] and electron promotion (generating single electron excitations above the Fermi level [7,18]). The latter, which in the following will be referred to as "'peak excitation"', is considered to model the fate of a single electron excitation and investigate the competition between ballistic transport and thermalization via electron–electron scattering. The temporal and spatial evolution of the distribution function $f(\vec{r},\vec{k},t)$ obtained this way can be analyzed with respect to the transport of excitation energy, and the results will be compared to the formerly used diffusive transport model.

2. Model

Once excitation energy has been generated within an atomic collision cascade, we can model its dynamics by means of the Boltzmann transport equation (BTE)

$$\left[\frac{\partial}{\partial t}+\frac{\hbar\vec{k}}{m_e}\cdot\nabla_{\vec{r}}\right]f(\vec{r},\vec{k},t)=\left.\frac{\partial f}{\partial t}\right|_{e-e}. \tag{1}$$

for the distribution function $f(\vec{r},\vec{k},t)$ describing the occupation probability density of an electronic state with wave vector $\vec{k}$. The left hand side of Eq. (1) represents the variation of $f(\vec{r},\vec{k},t)$ in space and time due to ballistic transport, whereas the right hand side of Eq. (1) takes into account the changes of the distribution function due to electron–electron scattering. Note that electron phonon collisions are neglected here, since the relevant timescale for these interactions is of the order of picoseconds [19], whereas the typical timescale of electron emission which is targeted here is only a few femtoseconds [8,9].

In detail, the variation of f due to electron–electron scattering, where electrons with initial wave vectors $\vec{k}$ and $\vec{k}_1$ are scattered into states with wave vectors $\vec{k}_2$ and $\vec{k}_3$ and vice versa, is given by

$$\begin{aligned}\left.\frac{\partial f}{\partial t}\right|_{e-e}=&\frac{2\pi\Omega^2}{\hbar}\int_\Omega d^2k_1\int_\Omega d^2k\ \ |M_{ee}(\Delta k)|^2\delta(\epsilon(\vec{k})+\epsilon(\vec{k}_1)-\epsilon(\vec{k}_2)\\&-\epsilon(\vec{k}_3))\times[f(\vec{k}_2)f(\vec{k}_3)(1-f(\vec{k}))(1-f(\vec{k}_1))\\&-f(\vec{k})f(\vec{k}_1)(1-f(\vec{k}_2))(1-f(\vec{k}_3))].\end{aligned} \tag{2}$$

In Eq. (2), M_{ee} denotes the electron–electron scattering matrix element in the k-space and Ω is the volume of an individual discretization cell. The essential physical input entering M_{ee} is the Fourier transform of a screened Coulomb electron–electron interaction potential

$$V(\vec{r})=\frac{e^2}{4\pi\,\epsilon_0\,r}e^{-\kappa r}. \tag{3}$$

with κ denoting the characteristic inverse screening length as the only free parameter of the model. For simplicity, we use the Thomas–Fermi screening length [20]

$$\kappa_{TF}=\frac{2\cdot m_e\cdot e^2}{\epsilon_0\,\hbar^2}\left(1-\exp\left(-\frac{\hbar^2\pi n}{m_e k_B T_e}\right)\right) \tag{4}$$

yielding $\kappa = 3.6\ \text{Å}^{-1}$ for the electron density n given below. A more sophisticated determination of the screening length would be a self consistent calculation of this quantity according to the equation

$$\kappa^2=\frac{e^2 m_e}{\pi^2\hbar^2\,\epsilon_0}\int_0^\infty dk\ f\left(k,t\right). \tag{5}$$

We checked the influence of κ on the calculated diffusion coefficient D (see below) in a range of $[2\ \text{Å}^{-1},5\ \text{Å}^{-1}]$ and find diffusion coefficients varying from 14.7 to 15.3 cm^2/s, indicating a rather weak influence with regard to the goal of the present work.

Using a plane wave approach for the electrons we obtain

$$M_{ee}=\frac{e^2}{2\Omega\,\epsilon_0}\frac{1}{\sqrt{(\Delta k)^2+\kappa^2}} \tag{6}$$

with $\Delta\vec{k}=\vec{k}_1-\vec{k}_2=\vec{k}_3-\vec{k}$ denoting the transfer of momentum between the electrons.

The delta-function $\delta(\epsilon(\vec{k})+\epsilon(\vec{k}_1)-\epsilon(\vec{k}_2)-\epsilon(\vec{k}_3))$ entering Eq. (2) ensures that in the sum over all mathematically possible collisions only those collisions are considered, which are in accordance with energy conservation. In addition, according to Pauli's principle the transition from an initial electronic state with wave vector $\vec{k}_i$ to a different final electronic state with wave-vector $\vec{k}_f$ scales with the probability that the initial state is occupied and the final state is unoccupied, leading to an additional factor $f(\vec{k}_i)(1-f(\vec{k}_f))$.

The numerical integration of this six-dimensional Boltzmann equation turns out to be computationally too expensive. Therefore, we restrict our calculations to one dimension in real space and two dimensions in the k-space, where one direction of the k-space is oriented along the transport direction in real space (x-axis) and the other direction is perpendicular to the transport direction. Note that this refers to a two-dimensional treatment in k-space, and therefore the characteristics of a two-dimensional electron gas need to be used in order to describe the ground state properties of the electronic system. The real space is discretized in cells of length $\Delta x = 3$ Å in the space domain, and the k-space is discretized into 61×61 k-vectors per spatial cell with $\Delta k = 0.054\ \text{Å}^{-1}$. This discretization is a trade-off between cpu-power and the resolution of the k-space. Note that Δk is small compared to the inverse screening length $\kappa = 3.6\ \text{Å}^{-1}$ of the screened Coulomb potential. Due to the finite spacing of the k-grid, fluctuations of the totatl energy in the system may occur. We have therefore repeated a few calculations using a finer spacing and found no significant differences with respect to the presented results.

The ground state properties of the system were set to describe a generic two-dimensional electron gas with a Fermi energy of $E_F = 5.0$ eV, corresponding to an electron density of $n = 2.1\times10^{19}\ \text{m}^{-2}$ and a wave vector $k_F = 1.2\ \text{Å}^{-1}$, which are approximately characteristic for silver at temperature $T = 0$ K. In order to describe a localized excitation of the system, two types of initial conditions for the distribution function $f(\vec{r},\vec{k},t)$ are implemented at a particular point in real space, namely i) a Fermi–Dirac-distribution at different elevated electron temperatures and ii) a 0 K Fermi–Dirac distribution additionally exhibiting one or more non-thermal single electron excitations in terms of selected k-states occupied above E_F.