Nuclear Instruments and Methods in Physics Research B 303 (2013) 55–58

Contents lists available at SciVerse ScienceDirect

Nuclear Instruments and Methods in Physics Research B

journal homepage: www.elsevier.com/locate/nimb

Computer simulation of internal electron emission in ion-bombarded metals

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ARTICLE INFO

Article history:
Received 23 July 2012
Received in revised form 19 December 2012
Available online 5 January 2013

Keywords:
Internal electron emission
Kinetic excitation
Metal–insulator–metal junctions
Ion bombardment

ABSTRACT

We present a computer simulation study of internal electron emission in ion-bombarded metal–insulator–metal (MIM) junctions. The computational approach consists of (i) a molecular dynamics part describing the particle kinetics upon projectile impact, (ii) the computation of kinetic electronic excitation as well as its transport and (iii) a thermionic model to calculate the flux of electrons from the top electrode to the bottom electrode of the MIM. The results are compared to recent experiments and discussed in terms of different transport models for the description of hot electron propagation in metals.

1. Introduction

The bombardment of a metal surface with keV-ions initiates a sequence of atomic collisions near the surface. This atomic collision cascade propagates in space and time on scales of a few nm and ps, respectively. The moment the ion is set into motion initializes the time scale of our simulation. Depending on the bombarding conditions, particles may be released from the surface in the course of the cascade. This process is usually called *sputtering* [1].

However, the bombardment does not only affect the nuclear degree of freedom, but is also connected with electronic excitation processes resulting from the movement of the target atoms [2] which manifest as physical phenomena such as kinetic electron emission [3] or secondary ion formation [4]. Furthermore, excitations with energies below the vacuum level can be experimentally observed by means of thin metal–insulator–metal (MIM) junctions [5,6]. In these experiments, the top electrode of a MIM-device is bombarded with ions, and part of the excited electrons propagate towards the buried oxide layer which effectively acts as an energy barrier. Hot carriers overcoming this barrier can be detected as an "internal emission current" between the top film and the bottom electrode. In this context, the terminology "internal electron emission" has been established [7].

In previous publications, we have developed a hybrid computer simulation model capable to treat the atomic particle dynamics within the cascade, the kinetic excitation mechanism as well as the transport of excitation energy away from the spot of generation. This model has been successfully employed to describe ion-induced kinetic electron emission and also secondary ion formation for silver self-bombardment with projectile energies of a few keV. These computational studies suggest the rough physical picture of the kinetic electron emission process to be dominated by the very initial electronic excitation peak (0–30 fs) directly following the particle impact, whereas the formation of secondary ions results from the evolution of the surface electron temperature at later stages of the collision cascade.

Within the time interval of about 1 ps after the projectile impact, the time and space evolution of the atomic collision cascade leads to a strong perturbation of the lattice. The loss of periodicity in the lattice generated that way leads to a more localized character of electrons in the region influenced by the projectile impact (called the "cascade volume") [8]. Analysis shows that on time scales of about hundred fs after the projectile impact, the cascade volume becomes completely amorphized.

Applying the model to the bombardment of an amorphous crystal target, it is possible to reproduce experimentally measured "external" kinetic electron emission yields into the vacuum in a quantitative manner [9]. While external emission represents a surface phenomenon and therefore probes the electronic excitation state in close vicinity of the surface, it is of great interest to determine how well the same concept is suited to describe the internal emission phenomenon which occurs at depths of the order of 10 nm below the surface. In the present study, we therefore extend the model to the prediction of internal emission yields across a buried oxide layer of a MIM device. In particular, we expect this process to be highly sensitive to how the initial excitation – which is predominantly generated close to the surface – propagates down to the oxide interface. The comparison of the calculated results with experimentally measured internal emission yields then

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http://dx.doi.org/10.1016/j.nimb.2012.12.023

allows a critical assessment of the validity of the transport mechanism incorporated in the model calculations.

2. Methods

The hybrid computer simulation model employed here consists of four parts, namely: (i) a molecular dynamics description of particle dynamics, (ii) the implementation of kinetic excitation mechanisms, (iii) the description of the transport of excitation energy and (iv) the description of the internal electron emission. In the following, each point will be briefly described.

2.1. Molecular dynamics

We use a standard molecular dynamics scheme to describe the particle dynamics. In short, the Newtonian equations of motion are numerically integrated for all atoms of the system (projectile + target atoms). For the silver self-sputtering system considered here, we employ a well-established semi-empirical many-body potential designed by Kelchner et al. [10].

2.2. Implementation of kinetic excitation mechanisms

The electronic friction mechanism is described on the basis of the Lindhard-Scharff-model of electronic stopping [11]. In this picture, the electronic system of the metal is assumed to be a homogenous quasi-free electron gas. Electronic excitations are supposed to originate from direct electron-atom scattering involving the conduction band electrons, leading to an effective friction force experienced by the moving atoms. Following Lindhard et al. [12], the electronic energy loss dE per track length dx of a particle moving with velocity v is given by the expression:

$$\frac{dE}{dx} = -Kv, \tag{1}$$

which can be transformed into the time domain yielding

$$\frac{dE}{dt} = -Kv^2 = -AE. \tag{2}$$

In Eqs. (1) and (2), A and K constitute material parameters which are calculated from Ref. [12] for a given target–projectile combination. For the Ag–Ag model system, the corresponding values are $A = 2.88 \times 10^{12}$ 1/s and $K = 258.9 \times 10^{-15}$ kg/s. The electronic energy loss of each moving atom calculated according to Eq. (2) is assumed to be instantaneously fed into the electronic system at the present position of the particle. In consequence, the entire ensemble of moving cascade atoms represents a time- and space dependent source of excitation energy. The total electronic excitation source rate is given by summing over all moving atoms (each with kinetic energy $E_{\mathrm{kin}}^{(i)}$) including the projectile, i.e.

$$\frac{dE(\vec{r},t)}{dt} = A\sum_i E_{\mathrm{kin}}^i(t)\cdot\delta(\vec{r}_i(t)-\vec{r}). \tag{3}$$

The second excitation mechanism incorporated into our model is electron promotion in violent atomic collisions. For two silver atoms within a silver solid, the quasi-diabatic molecular orbital evolving from the atomic 4d-level is strongly shifted upwards in energy with decreasing interatomic distance r and energetically crosses the Fermi level E_F at $r \approx 1.5$ Å. Once this internuclear distance is undercut in a binary collision, the electron may undergo a resonant transition into a free conduction band state of energy E^*, thereby generating an electron with an excitation energy $E^* - E_F$. In addition, the energy E_h of the d-hole generated this way must be considered as an (indirect) source of excitation energy.

2.3. Transport of excitation energy

Once an electronic excitation has been created, it is strongly delocalized and rapidly spreads within the solid. Possible methods to describe this transport process include statistical [13], ballistic [14] or diffusive approaches. In our model, we treat the transport by means of a nonlinear diffusion equation with an excitation energy diffusivity D constituting the essential model parameter.

At the surface, a Neumann-type boundary condition is applied in order to prohibit the flux of excitation energy from the solid into the gas phase. In order to simulate an infinite extension of the electronic system parallel to the surface, we use pseudo-infinite boundary conditions at the lateral boundary planes of the cascade volume as described in great detail elsewhere [8].

This procedure finally yields a full four-dimensional excitation energy profile $E(\vec{r},t)$ which can be converted into an electron temperature $T_e(\vec{r},t)$. This can then be used to calculate ionization probabilities of sputtered particles, kinetic external or internal electron emission yields as discussed below.

2.4. Description of internal electron emission

Fig. (1) illustrates the experimental geometry of the ion-bombarded MIM junction. The metal–insulator interface at depths of the order of 10 nm below the surface can be considered as an effective energy barrier for the electrons. For Ag|AlO_x|Al MIM structures frequently used in experiments, the barrier height Φ_B is found to be approximately 2.5 eV. Experiments show that only a small minority of the electrons detected in the bottom electrode originate from tunneling processes [15]. More specifically, the interpretation of experimental data on measured internal emission yields in terms of simple ballistic tunneling theory reveals that the majority of the detected current results from "over-the-barrier" transmission at energies close to the barrier edge. Therefore, it appears reasonable to calculate the internal electron emission yield by means of a thermionic emission model of the Richardson-Dushman type as has been successfully employed for the calculation of external electron emission yields [9]. With Φ_B denoting the barrier height, the internal electron emission yield γ_{int} for a MIM-device with an interface located at depth z_0 below the surface can then be numerically calculated for each impact as [16]

$$\gamma_{\mathrm{int},k} = \frac{1}{e}\int_0^{t_c}\int_A j_{e,k}(x,y,z_0,t)\,dA\,dt, \tag{4}$$

with

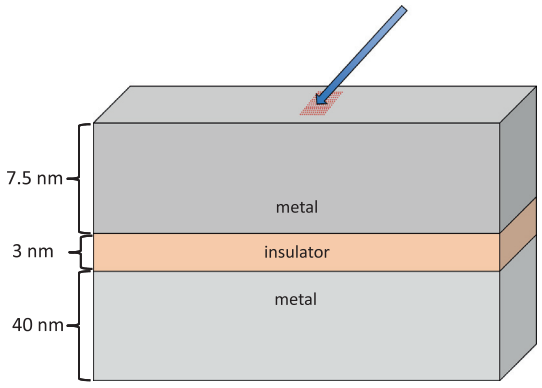

Fig. 1. Scheme of experimental geometry of a metal–insulator–metal junction bombarded with ions.