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Atomistic self-sputtering mechanisms of *rf* breakdown in high-gradient linacs

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ABSTRACT

Molecular dynamics (MD) models of sputtering solid and liquid surfaces – including the surfaces charged by interaction with plasma, Coulomb explosion, and Taylor cone formation – were developed. MD simulations of self-sputtering of a crystalline (1 0 0) copper surface by Cu^+ ions in a wide range of ion energies (50 eV–50 keV) were performed. In order to accommodate energetic ion impacts on a target, a computational model was developed that utilizes MD to simulate rapid atomic collisions in the central impact zone, and a finite-difference method to absorb the energy and shock wave for the collisional processes occurring at a longer time scales. The sputtering yield increases if the surface temperature rises and the surface melts as a result of heat from plasma. Electrostatic charging of the surface under bombardment with plasma ions is another mechanism that can dramatically increase the sputtering yield because it reduces the surface binding energy and the surface tension. An MD model of Taylor cone formation at a sharp tip placed in a high electric field was developed, and the model was used to simulate Taylor cone formation for the first time. Good agreement was obtained between the calculated Taylor cone angle (104.3°) and the experimental one (98.6°). A Coulomb explosion (CE) was proposed as the main surface failure mechanism triggering breakdown, and the dynamics of CE was studied by MD.

1. Introduction

Energetic ion collisions with solid targets are an important area of research in basic science [1–8], as well as in numerous industrial applications [9–16]. Tech-X Inc.[1] has been developing multi physics finite-element packages, including TXphysics, OOPIC, and, most recently, VORPAL, that are capable of simulating the dynamics and interaction of metal plasma with the surfaces in various *rf* structures. As does any finite-element package, these computational tools need the plasma–surface interaction characteristics, such as the sticking, reflection, and sputtering coefficients, that ultimately define the atomic flux from the surface to the plasma (i.e., fueling the latter) as a boundary condition. Since the sticking and sputtering yield coefficients are the atomistic characteristics of the plasma ion interaction with the surface, they can be calculated directly by either Monte Carlo or molecular dynamics methods.

Self-sputtering processes are also of fundamental interest in the development of high-gradient *rf* accelerators [1]. At least three research and development efforts are independently studying the behavior of high-gradient *rf* structures for accelerators. The Neutrino Factory and Muon Collider Collaboration (NFMCC) is looking at developing low-frequency structures for muon cooling [2–6], the International Linear Collider is optimizing the performance of 1.3 GHz superconducting *rf* structures aimed at the design of a 1 TeV superconducting electron/positron collider [7], and the High-Gradient RF Collaboration is studying high-frequency ($f > 10$ GHz) structures aimed at an electron–positron collider operating at energies higher than 1 TeV [8].

Understanding self-sputtering is also important for cathode discharge applications such as surface coating and modification [9–11], explanation of the unipolar arc development [12,13], detection of fast ions and particles [14,15], formation of columnar defect in alloys for fast particle registration [16], integrated circuits development based on copper connectors, and thin-film deposition development [17,18].

Experimental studies of the self-sputtering were carried out in [17] and discussed in [18,19]. Sputtering yields of metal ion beams, with energies from 10 to 500 eV, on polycrystalline Au, Cu, Ag, Cr, and Al films were measured in situ by using crystal microbalance techniques. A momentum-transfer model was developed that predicts the sputtering yield in the form

$$Y = K(E/\lambda)\left[\frac{m_1 m_2}{(m_1+m_2)^2}\right], \quad (1)$$

where K is the coefficient depending on the material, E is the ion energy, and λ is the penetrations depth (range) of ion in the target ($\lambda \approx 10$ Å at 300 eV for all studied metals). Eq. (1) was found to be appli-

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cable in both high-ion and low-ion energy ranges, with an accuracy of ~20% [17].

Physical and ion-enhanced chemical etch yields by noble gas and metal ions were fitted by a square root function down to the threshold energy for self-sputtering of metals, noble gas sputtering of metals, Si, SiO_2, and ion beam etching of Si in [18]. Energetic ion-surface collisions used in thin-film technology imply both favorable effects such as promoting epitaxial growth and deleterious effects, since such collisions can introduce structural and/or compositional defects in the film. Both processes depend on the ion energy; and, therefore, optimization of the growth processes by selecting ion energies for fabrication of high-quality thin films is a high demand. Analysis of ion collisions was, provided for a wide variety of projectile ion/target material at very low ion energies. A universal dependence was proposed that describe the etching rate for semiconducting, insulating and metal surfaces above the threshold energies.

The sputtering yield Y, which is the number of neutral atoms ejected per incoming ion, can be expressed according to Sigmund's theory as follows:

$$Y(E) = C_{\mathrm{pt}} S_n\left(E/E_{\mathrm{pt}}\right), \tag{2}$$

where C_{pt} and E_{pt} are constants that depend on the projectile ion p and target material t, and $S_n(\varepsilon)$, $\varepsilon = E/E_{\mathrm{pt}}$, is a universal function, the nuclear stopping cross-section, depending on the reduced energy ε. C_{pt} can be expressed via the surface binding energy U, and $S_{\mathrm{n}}(\varepsilon) \sim E^{1/2}$, for $\varepsilon \leqslant 0.02$. Steinbrüchel [18] proposed a formula based on a detailed comparison to experimental data:

$$Y(E) \approx A\left(E^{1/2} - E_{\mathrm{th}}^{1/2}\right), \tag{3}$$

where A and E_{th} are constants of the incoming ion/target material.

As mentioned above, the sputtering yield is a boundary condition for finite-element plasma simulation codes and can be calculated by atomistic simulation methods, such as Monte Carlo or molecular dynamics [19]. Monte Carlo (MC) is the main method for sputtering yield calculations and was developed in a series of papers [20,21]. A Monte Carlo code T-DYN was developed by Biersack et al. [20]. The standard MC code simulates the event of an ion collision with the virgin target, without defect accumulation in the target from ion impacts. This code was applied to various ion/target material combinations in [21,22].

A self-sustained self-sputtering occurring during high-current pseudospark operation ($\approx 10^4$ A/cm^2, $I > 10^3$ A) is shown in [22] to be a possible mechanism for superdense glow. The mean-free-path for ionization of cathode material sputtered in the low-current hollow-cathode phase can be shorter than the cathode–anode gap distance, and ionized atoms can return to the cathode surface, self-sputtering with a yield greater than one. The self-sputtered cathode atoms become ionized in the beam of electrons accelerated in the cathode sheath. As was shown in [22], a large fraction of the discharge current at the cathode surface can be carried uniformly over the surface by ions, and a very high electron emission density is not required to maintain the high current.

A review of the method of MD computer simulation and results obtained for the physics of sputtering is given in [19]; the physical input (such as the interatomic potentials, coupling to the electronic system), reliability, and computer time requirements of simulations are discussed. MD results obtained after 1992, the time of the last review, are presented, with an emphasis on results that are difficult to obtain by other theoretical or computational means: sputtering from high-energy–density zones (spikes), cluster emission, formation of surface topography and their influence on sputtering, and chemical effects.

MD simulations of self-sputtering and sticking coefficients of low-energy ion were reported in [23]. Sputter yields Y and sticking coefficients S are essential inputs for evolution studies of feature profiles. MD simulations are used to compute sputter yields and sticking coefficients for Cu ions impinging on a Cu surface at various incident energies $15 < E_{\mathrm{i}} < 175$ eV and incident angles $0 < \theta_{\mathrm{i}} < 85°$. Threshold energies for sputtering E_{th} are also predicted and shown to vary with θ_{i}. Abrams and Graves [23] showed that for energies below the experimental threshold for physical sputtering ($E_{\mathrm{th(expt)}} \sim 60$ eV), a yield between 0.01 and 0.1 Cu/ion still can be obtained for some off-normal angles of incidence. A dependence $Y \propto E_{\mathrm{i}} - E_{\mathrm{th}}$ was found to be valid below $E_{\mathrm{th(expt)}}$ when Y is a maximum with respect to θ_{i} (at $\theta_{\mathrm{i}} = 45°$). It was also shown that a dependence $Y \propto {E_i}^{1/2} - {E_{\mathrm{th}}}^{1/2}$ is more suitable at other incidence angles [23].

MD simulations were also used to calculate scattering and penetration of normally incident hyper thermal ($5 < E_{\mathrm{i}} < 400$ eV) of Ne, Ar, and Xe atoms off a Cu crystal [24]. The authors showed that the incident energy was efficiently deposited in the solid and that the deposited fraction depends mostly on the projectile mass, and less on the bombarding energy. At low energy, the larger part of the nondeposited energy is taken away by the reflected projectile. Above the sputter threshold, an amount between 2% and 6% of the incident energy was carried by the sputtered particles. The results compared well with experiment. Gades and Urbassek [24] demonstrated that a realistic interaction between the projectile and the target atoms influences the energy deposition at energies below around 100 eV.

Several researchers have used MD simulations to study Coulomb explosion events occurring at energetic heavy and highly charged ion collisions with solid targets [14,26,27].

One of the goals of the simulations we present here is to verify and benchmark the sputtering yields used as an input file in the TXphysics and OOPIC software packages developed at Tech-X Inc. We also describe a preliminary interface between Vorpal and MD that we have constructed based on the exchanges of input/output files from one simulation level to another.

2. Simulation models

This section focuses on the sputtering and Coulomb explosion models developed for simulation studies.

2.1. Sputtering model

The evolution of the atomic system was determined by the solution of the set of the classical equations of motions for all the atoms in the system. The copper target was represented by crystalline (1 0 0) copper substrates, with the sizes dependent on the ion kinetic energy. All substrates were prepared by equilibration at room and elevated temperatures. Additional simulations were used to prepare a liquid copper surface for self-sputtering simulations.

The energetic ions were placed at a certain distance z_{i} above the target's surface (see Fig. 1), where the distance to the surface z_{i} was varied in three Cartesian directions within approximately one lattice parameter in order to randomize the collision event for further averaging. The ions were directed toward the target at an impact angle of 5°, in order to prevent channeling mode. The ion energies were selected in an interval from 50 eV to 50 keV, in order to compare with the existing sputtering yield data.

The size of the target is critical for the correct calculation of the sputtering yield since the shock-waves resulting from impacts have a velocity of ~5 km/s in copper and therefore are reflected from the basic cell's boundaries within 1–2 ps. If no special measures are taken to suppress or mitigate these waves, they can drastically change the calculated result. Therefore, a parallelepiped