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Computer simulation of sputtering induced by swift heavy ions

P. Kucharczyk*, A. Füngerlings, B. Weidtmann, A. Wucher

Fakultät für Physik, Universität-Duisburg-Essen, 47048 Duisburg, Germany

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ABSTRACT

New experimental results regarding the mass and charge state distribution of material sputtered under irradiation with swift heavy ions suggest fundamental differences between the ejection mechanisms under electronic and nuclear sputtering conditions. In order to illustrate the difference, computer simulations based on molecular dynamics were performed to model the surface ejection process of atoms and molecules induced by a swift heavy ion track. In a first approach, the track is homogeneously energized by assigning a fixed energy to each atom with randomly oriented direction of motion within a cylinder of a given radius around the projectile ion trace. The remainder of the target crystal is assumed to be at rest, and the resulting lattice dynamics is followed by molecular dynamics. The resulting sputter yield is calculated as a function of track radius and energy and compared to corresponding experimental data in order to find realistic values for the effective deposited lattice energy density. The sputtered material is analyzed with respect to emission angle and energy as well as depth of origin. The results are compared to corresponding data from keV sputter simulations. As a second step of complexity, the homogeneous and monoenergetic lattice energization is replaced by a starting energy distribution described by a local lattice temperature. As a first attempt, the respective temperature is assumed constant within the track, and the results are compared with those obtained from monoenergetic energization with the same average energy per atom.

1. Introduction

The sputtering of atoms from a solid by ion bombardment is of great practical interest in areas related to the fabrication technology of semiconductor devices and to surface analysis techniques such as Secondary Ion Mass Spectroscopy (SIMS). In many applications, sputtering is induced by the impact of low energy ions with energies in the keV regime. In this case, the energy transfer between projectile and target is exclusively dominated by mostly elastic collisions, with the recoiling target atoms undergoing further collisions, and target atoms may be released from the surface at the end of such a collision cascade. In addition to the collisional "nuclear" energy loss $(dE/dx)_n$, the projectile experiences an electronic energy loss $(dE/dx)_e$ by generating electronic excitations within the target material. In the low energy regime, this electronic stopping has only a small influence on the particle ejection mechanism itself, but may influence the excitation or charge state of the sputtered particles [1,2].

At high impact energies, where a swift heavy ion impinges with a kinetic energy of the order of MeV/u, the situation changes. Here, the interaction with the nuclear system becomes negligible, and the slowing down of the projectile is caused almost solely by electronic stopping. Depending on the target material, the electronic excitation generated this way may rapidly spread and dissipate into the bulk of the irradiated sample without coupling to the atomic lattice. In other cases, the excitation stays localized around the trajectory of the incoming ion for a long enough time to efficiently couple energy into the lattice. In this scenario, electronic sputtering occurs if the lattice becomes energized enough to allow the evaporation of atoms. The sputter yield Y, i.e., the average number of atoms ejected from the surface per incident ion, observed for solids under such irradiation conditions is generally larger than would be expected from purely collisional sputtering [3].

To understand the energy exchange processes following the impact of a swift heavy ion, which ultimately lead to material modification and emission following the primary electronic excitation, thermodynamic models have been developed. These models are based on the assumption of a local thermal equilibrium described by a time and position dependent temperature both in the electronic and the lattice sub-systems of the solid. Moreover, it is assumed that the transport of energy within both systems is governed by diffusion. In the MeV/u energy range of interest here, the dominating energy transfer occurs via electronic stopping of the projectile, with the nuclear energy loss being negligible at these energies. The primary excitation generated in the electronic sub-system is rapidly shared and thermalized between the electrons on a femtosecond time scale, leading to an elevated electron

* Corresponding author.
E-mail address: pawel.kucharczyk@uni-due.de (P. Kucharczyk).

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temperature T_e, while the nuclear system is still at a low lattice temperature T_a and mainly heated via electron-phonon coupling. The spatio-temporal evolution of electron and lattice temperatures is then described by two coupled non-linear heat flow equations, leading to the generation of a thermal spike in the lattice sub-system. It is generally believed that track formation is observed when T_a exceeds the melting temperature of the material, whereas sputtering, i.e., the ejection of (sub-)surface material into the vacuum above the surface, occurs if the mean excitation energy per atom becomes comparable to the sublimation energy.

A thermodynamic description of particle emission from a thermal spike was published by Sigmund and Claussen [4]. Within this model, energy is assumed to be instantaneously deposited into the nuclear system within a cylindrical track volume centered around the trajectory of the impinging ion, producing a locally enhanced lattice temperature which spreads and dissipates according to the law of heat conduction in a continuum [5]. Particle emission is then described as the thermally activated evaporation from an ideal gas bounded by a planar surface barrier, where the evaporated flux is integrated over the spatio-temporal temperature distribution at the surface. In this work, the authors derived expressions for the total sputter yield as well as the emission energy distribution of the sputtered particles, with a central result being the prediction that the sputter yield should scale with the square of the initially deposited energy per unit track length. Assuming a constant conversion efficiency from electronic to lattice excitation energy, this would mean that the sputter yield should scale with $(dE/dx)_e^2$, a relation which has indeed been observed for fast ions incident on low-temperature condensed-gas solids [6]. A similar dependence is measured for insulators like oxides as well, while the yields measured for ionic crystals are generally found to increase more steeply [3]. For metallic samples, on the other hand, systematic investigations of this kind are still lacking.

A number of studies have been performed to test the concept of thermal spikes by using molecular dynamics (MD) to follow the nuclear motion within the target on a microscopic scale. As a result of such simulations, Bringa et al. [7] have shown that the assumption of a diffusive energy transport within the irradiated material is probably incorrect. Instead, they show that the particle motion is governed by a pressure pulse building up in the energized track region. Based on these results, Jakas et al. proposed a fluid dynamics approach to model the ejection process [8] in a way similar to the gas flow model published by Urbassek and Michl [9]. While the first simulations were performed on weakly bound systems using the pairwise additive Lennard-Jones potential to describe the interaction between the target atoms, it was later shown that the results were transferrable to metallic systems as well, provided the length and energies are scaled to the respective interatomic distances and binding energies [10]. Since these early studies involving an instant lattice energization as a starting point for the MD simulations, more sophisticated approaches have been developed where the time dependent nature of the local energy transfer to the lattice atoms is acknowledged. One possible approach is to employ a parametrized energy-time distribution derived, for instance, from a two temperature model (TTM) treating the energy coupling between electronic and nuclear subsystems. In order to further the details of the energy transfer processes, the TTM can be coupled to a Monte Carlo approach [11] describing the initial generation and redistribution of electronic excitation energy prior to electronic thermalization. Models of this kind have been successfully employed to describe structural modifications in insulators [12] and semiconductors [13,14]. Probably the most advanced simulations available today describing the interaction of a swift heavy ion with a solid combine a TTM-calculation of the electron temperature profile with an MD simulation which explicitly couples the atomic motion to the momentary electron temperature [15]. While models of this kind have successfully been applied to describe SHI-induced structural modifications [16], they have not yet been applied to electronic sputtering phenomena. An alternative concept to describe the lattice dynamics following the impact of a swift heavy ion is given by the Coulomb explosion model originally proposed by Fleischer et al. [17]. Within this model, target atoms along the ion trajectory become ionized by the projectile, and the resulting repulsion between the ionized atoms leads to track formation and particle emission. The dynamics as a consequence of an initially repulsive Coulomb force between the ionized atoms have also been followed by MD simulations and were shown to produce similar results as the thermal spike calculations [18].

In our group, we have recently started to perform sputtering experiments with swift heavy ions impinging onto various metal, semiconductor and insulator targets. In these experiments, we investigate the mass spectrum of secondary neutral and charged particles emitted from the bombarded surface due to the projectile ion impact. While the secondary ions are directly detected using a Time-of-Flight (ToF) spectrometer, neutral particles are post-ionized using a pulsed VUV laser beam to render them accessible for mass spectrometric analysis. In order to facilitate a quantitative comparison of secondary ion and neutral sputter yields, particular emphasis is put on the fact that both species are detected under otherwise identical experimental conditions regarding detection efficiency as well as the sampled solid angle and emission velocity intervals. Comparing the measured signal of post-ionized secondary neutral species with those of the corresponding secondary ions, it is possible to determine the ionization probability, i.e., the probability that a sputtered particle is emitted in a positively or negatively charged state.

One of the most interesting results of our studies is the fact that relatively large neutral sputter yields are observed for a few metallic targets irradiated by swift heavy ions with energies of about 5 MeV/u [19]. At these energies, amorphous tracks were previously found to form only in Ti and Zr [20]. These observations are notable, because metals are assumed to be insensitive to electronic energy-loss due to their high electron mobility. For the special case of a dynamically sputter cleaned polycrystalline indium sample being irradiated by 4.8 MeV/u gold ions, however, it was found that significant electronic sputtering of mostly neutral In atoms and In_n clusters occurs, with the ionization probability of the emitted indium atoms (leading to the formation of In^+ secondary ions) being about one order of magnitude larger than that determined in-situ under bombardment with 5 keV argon ions [21]. Moreover, the ionization probability measured under SHI impact was found to be practically insensitive of surface oxidation induced by deliberately exposing the sample to O_2. In contrast, secondary ion formation observed under keV argon ion bombardment exhibits the well-known oxygen matrix effect, leading to a drastic enhancement of the ionization probability by more than two orders of magnitude at the oxidized surface. From these experiments, it was concluded that the emission and ionization processes of sputtered particles must be fundamentally different under electronic and nuclear sputtering conditions, respectively. In particular, we suppose that the depth of origin of the sputtered particles must differ in a characteristic way, leading to the apparent insensitivity of electronically sputtered material to the chemistry of the uppermost surface layer.

In the present paper, we therefore model the emission process of (sub-) surface particles following a sudden energization of a certain lattice volume within a solid such as that generated by the impact of a swift heavy ion. The goal is to calculate the sputtering yield, i.e., the average number of atoms ejected from the solid, as a function of lattice excitation parameters such as the extension and energy content of the energized region. By comparing the resulting yield values with those observed in our experiments, we then determine “reasonable” excitation parameters, which are then used to calculate characteristics of the particle emission process such as the emission energy, angle and depth-of-origin distributions of the ejected particles. The resulting emission characteristics will then be compared with those prevailing under keV impact induced nuclear sputtering conditions in order to illustrate possible differences in the ejection mechanism under nuclear and