Atom Ejection Mechanisms and Models

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In a series of publications the authors have been exploring atom ejection from clean and chemically reacted copper and nickel surfaces under argon ion bombardment at 600 eV ion energy by classical trajectory simulations. The calculations generate a variety of experimental observables including relative sputtering yields, energy and angular distributions for the ejected atoms, multimer yields and surface damage information. The method provides a necessary first step in the development of rigorous ejection models, prior to the inclusion of ionization effects. A general conclusion from all of the calculations is that surface morphology - the location of specific atoms with respect to each other - effectively controls the ejection mechanisms for a specific system. A comprehensive comparison between the predictions of the simulations and experimental SIMS results obtained from clean and reacted surfaces has been published [1]. These comparisons are most successful when ratios of variables between different crystal faces or adsorbates in different coverages are used. Computed yields are sensitive to the crystal structure, coverage, binding energy and site symmetry of adsorbates. A recent investigation was concerned with the Cu(111)/Ar^+<111> system as a function of the ion energy [2]. This report summarizes and extends that work.

Details of the classical dynamical procedure have been described elsewhere [3]. In general, we solve Hamilton's equations of motion in time for a model microcrystallite consisting of four layers with ~90 atoms/layer. The Ar^+ ion is aimed toward an impact point in an irreducible symmetry zone of the surface, and the resultant collision cascade continues until the remaining atoms have insufficient energy to escape from the surface. The process is then repeated for a number of additional impact points. The original results were obtained with sets of 80 impact points trajectories. The model is discussed in greater detail in ref. 3.

1. Summary of Reported Results

Previous results [1-3] indicate that the computed yields compare quite favorably with the available experimental data. Most of the atom yield has been found to come from the first atomic layer of the target. The second layer contributes <10 percent and the third layer ~1 percent. The experimental ejected atom yield, atoms/ion, is a surprisingly poor measure of the yield from an individual ion trajectory. The distribution of atoms ejected/single ion (ASI) contains valuable information. Ejected atom energy distributions and various parameters associated with multimer formation were presented. Multimer yield ratios have been calculated which can be directly compared with experimental results.

All of the results support a theory of atom ejection which is a combination of the transparency of the lattice coupled with dynamic processes in the first few layers of the target surface. There is no support for a model which depends upon the flux of atoms or energy from below the surface.

2. Atoms Ejected/Single Ion

An examination of the details of individual ion collision events indicates that some ions eject surprisingly large numbers of atoms, even at low ion energies, and some eject none. A study of ASI as a function of ion energy gives valuable insight into the atom ejection mechanism.

The earlier ASI studies were limited by the small number of trajectories calculated at each energy. While 80 trajectories are sufficient for yield and multimer studies they give very 'noisy' ASI distributions. To remedy this situation 480 additional trajectories were calculated at 8.0 keV. The original impact points were distributed as uniformly as possible over the irreducible symmetry zone. The additional points were chosen to allow a sensitivity analysis of the original set. This was accomplished by shifting the entire grid of points by small displacements in six different directions. Thus, the relative orientation and separation between impact points was maintained and a set of points near each of the original points was examined. Standard deviations were calculated for many of the data previously reported from the seven data sets, (see Table 1).

Table 1 Data at 8.0 keV for the Cu(111)/Ar^+<111> System

Atom Yield	10.9	$\pm$ 0.3	atoms/ion
Atom fraction in multimers	0.21	$\pm$ 0.02	
Fractional yield, layer 2	0.087	$\pm$ 0.005	
Fractional yield, layer 3	0.0012	$\pm$ 0.0002	
Dimer/monomer ratio	0.085	$\pm$ 0.007	
Trimer/monomer ratio	0.011	$\pm$ 0.004	
Quadrimer/monomer ratio	0.006	$\pm$ 0.002	
Trimer/dimer ratio	0.13	$\pm$ 0.01	
Quadrimer/dimer ratio	0.075	$\pm$ 0.002	

The largest multimer encountered in this series was a nomomer, Cu_9, which was bound with 0.6 eV/atom. All of the smaller multimers also occurred, but the yields were too small to be meaningful.

The ASI distribution function at 8.0 keV is shown in Fig.1. Note that there are impact points which have no yield. The function appears to consist of two components, a narrow one centered near an ASI of 11, and a broad distribution from about 2 to 25. With this curve as a guide the 80 point ASI functions can also be interpreted. The first component appears in the energy range from 3.0 to 10.0 keV, where the yield is largest. The maximum ASI also occurs in this region; both 5.0 and 10.0 keV produced values which exceed the ASI of 25 shown in the Figure. The low energy and high energy distribution functions appear to contain only the broad component. At 0.6 keV the maximum is 19. The two functions are remarkably similar. Above 10.0 keV the maximum ASI is decreasing with energy, which is not the result expected from the statistical theories.

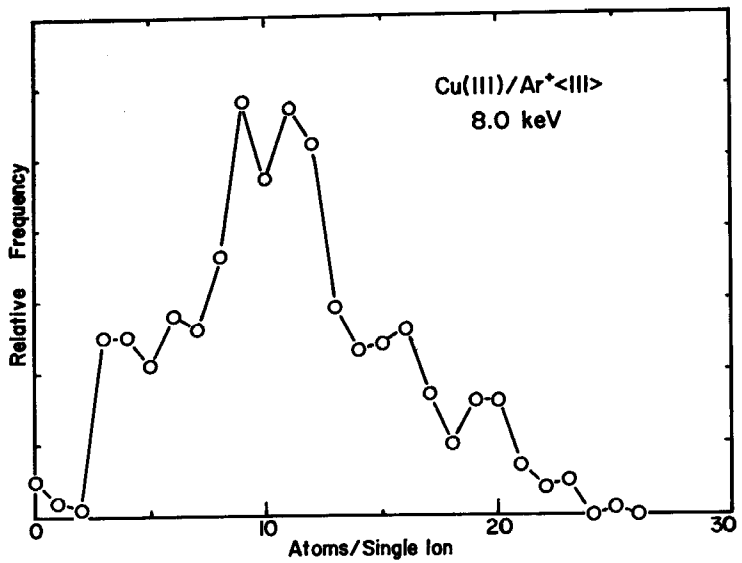

Fig.1 ASI Distribution Function for Cu(111)/Ar^+ 111 at 8.0 keV. The most probable ASI, 9, occurred 58 times in 560 trajectories

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New Models of Sputtering and Ion Knock-On Mixing

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We have recently developed new models of sputtering [1], ion emission [2], and ion knock-on mixing [3] which provide quick and accurate estimates of neutral and ion sputtering yields and of the broadening observed in a sputter-profiling experiment. In this abstract, we briefly summarize the major features of the sputtering and ion emission models.

We separate the sputtering process into bulk and surface effects. The relative sputtering yield, i.e., variations with ion energy, mass and incident angle, is determined by a bulk effect model which is illustrated in Fig.1.

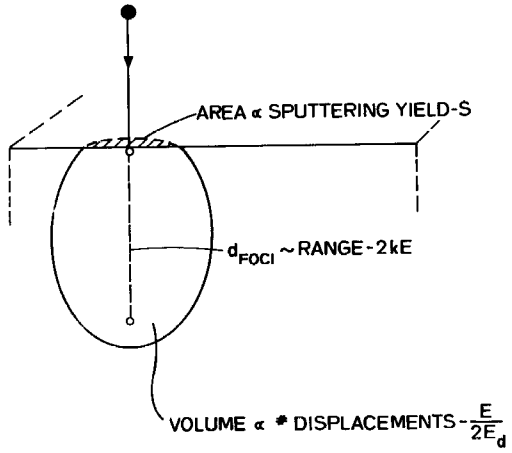

Fig.1 The incident ion creates a disturbed statistical region in the bulk. Its intersection with the surface defines an affected area which is proportional to the yield

This model leads to a simple algebraic equation [1] which accurately predicts the sputtering yield as a function of the ion energy, mass, and incident angle over a broad range of these parameters. In Fig.2, the mass dependence of the model is checked against the experimental data of EERNISSE [4].

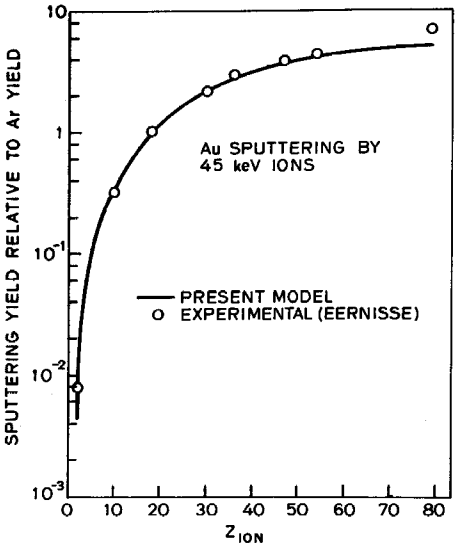

Fig.2 Au is bombarded by 45keV ions (data of EERNISSE [4]). Solid curve is the prediction of the present model

A scale factor B, defined as the fraction of displaced surface atoms that actually escape, is needed to obtain absolute sputtering yields. This factor is a property of the target surface only and is the only parameter requiring adjustment in the model. B may be determined from comparison to experimental data in the literature. See Fig.3.

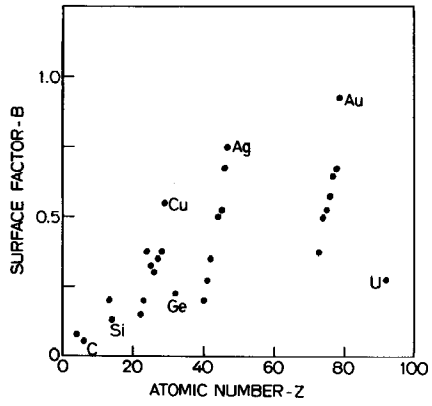

Fig.3 Values of B, the fraction of displaced surface atoms that escape, derived from the experimental data of ROSENBERG and WEHNER [5]

It appears that B is proportional to $n\sqrt{M}$ where n is the number of valence electrons and M is the atomic mass. This is consistent with an ion neutralization model (e.g. [6]) in which displaced ion cores at the surface are neutralized with a probability inversely proportional to their emitted velocity.

We propose that unneutralized ions must overcome an additional image force. The majority of emitted ions have energies of only a few eV and cannot surmount the image force barrier. This model may now be used to predict the ion yield I and sputtering yield S as follows: We let N_T be the total number of displaced surface atoms as predicted by our algebraic equation; N_N is the number of neutrals and N_I the number of ions such that $N_T = N_N + N_I$. We now have $S = BN_T \cong N_N$ and we assume that $I = \delta N_I = \delta(1 - B)N_T$ with $\delta \ll 1$. We thus obtain the following equation for the yields:

$$I/S = \delta(1 - B)/B \qquad (1)$$

The factor (1 - B)/B compares very favorably with the values of I/S measured by PRIVAL [6]. This model provides a simple explanation of the correlation between high sputtering yields and low ion yields.

The observed increase of both positive and negative ion yields upon oxidation [7] may be related to a reduction in the image force. Ions which surmount the image potential come from the broad high energy tail of the emitted atom distribution. For this reason, the average energy of emitted ions is greater than the average energy of emitted neutrals. The factor δ varies inversely with the image potential. We note also that the I/S dependence of the ion yield observed by DELINE [8] is similar to the (I - B)/B dependence observed here.

Finally, we comment on the physical content of the model described herein. The ellipsoid illustrated in Fig.1 is a mathematical construct which allows us to estimate the amount of energy deposited near the surface. The remarkable agreement of the model with experiment over a broad range of ion energies, masses, and incident angles indicates that we are using a valid statistical approach. Crystalline and other second order effects are, of course, not included in the model. The validity of the sputtering model is discussed further in [1].

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