Computer Simulation of Sputtering. DON E. HARRISON, JR., Dept. of Physics and Chem., Naval Postgraduate School, Monterey, CA, 93940, BARBARA J. GARRISON, Univ. of Calif., Dept. of Chem., Berkeley, CA, 94720, NICHOLAS WINOGRAD, Purdue Univ., Dept. of Chem., W. Lafayette, IN, 47907.

Classical trajectory simulations have developed to the point that it is feasible to model the cascade produced by an ion impact event. The results are sufficiently close to the experimental data that the computer model serves to interpret and explain the experiments. The ability to follow the dynamics of each individual atom in the cascade leads naturally to pictorial interpretations of a single sputtering event. Statistical analysis of data obtained from the results of many similar impact events produces numbers which can be directly compared to the experimental data. The comparisons are good indications of the reliability of the model approach.

The model has been developed and extensively tested in connection with theoretical studies of sputtering(1-3). Preliminary investigations of cluster formation from pure metal targets(4-6) indicate that the transition to systems of interest to the SIMS experimentalist can be made easily. Investigations in progress at this time demonstrate that oxygen atoms can be placed on the target surface without unduly complicating the computations or greatly increasing the computation time.

We model the ion impact event using classical dynamics. Extensive testing(4) has shown that to contain all of the important cascade events the target microcrystallite must consist of at least 250 atoms. The positions and momenta of the primary ion and all of the lattice atoms are developed in time during the trajectory or collision cascade. The trajectory computation is terminated when the momentum has dissipated through the microcrystallite and no more atoms can be ejected. In practice the calculation is stopped when the most energetic particle remaining in the crystal has 2 eV of kinetic energy. The final positions and momenta can then be used to determine particle yields, the energy distribution, the angular distribution and information about the formation of molecular clusters from the ejected atoms. Each trajectory is different because the ion is initially directed toward a different point on the target surface. Between 100 and 200 points must be sampled on each target orientation, at each energy and angle of incidence, to obtain statistically meaningful samples.

The classical dynamics are controlled by the pair potentials between the colliding particles. A repulsive potential is required for the ion-atom interactions. The atom-atom interactions are both repulsive and attractive at different collision energies. The potential functions are the heart of the calculations when close agreement must be maintained between the calculated results and experimental data. On the other hand, much useful pictorial and interpretive data can be obtained from potential functions which are only approximately correct.

The model computations are done using single crystal targets oriented to expose the low index surfaces. They indicate that the ejection mechanisms are very sensitive to the target face exposed. For example, the atom struck by the ion is often emitted from the (110) face, but very rarely from the (100) and (111) faces(4). An overwhelming majority of the ejected atoms come from the surface layer of the crystal for all three faces, and ejection from below the second layer is rare.

Some years ago the Cu/Ar^+ system was chosen for baseline investigation by computer simulation. The computational model is a decendent of the pioneering effort on the computer simulation of radiation damage(7). Early work(8) with Cu/Cu^+, rapidly developed into studies of the two element Cu/Ar^+ system(1-3). During this period the integration scheme was developed and refined(1,9), adjustable parameters such as the surface binding energy were replaced by complete interatomic potential functions(2) which span both the repulsive and attractive portions of the interaction. The model simulation gives excellent agreement with all experimental data available for the Cu/Ar^+ system. The model originally developed for relatively high energy (> 1 keV) studies on the (100) surface was found to work, without adjustment, on the other two low index faces and at greatly reduced ion energies(4). Sputtered atom energy distributions agree very well with experimental results(4). Initial studies on a single crystal orientation(3) showed that the model produces dimers, Cu_2, and trimers, Cu_3, over a wide range of ion energies and impact points on the crystal. Similar behavior has now been observed from the other two low index faces(4,6), and a sputtered pentamer, Cu_5, has been studied in some detail(5). Formation of

dimers from surface nearest-neighbor atoms is not common from the (100) and (110) faces, but often occurs from the (111) face(6). Thus the molecular recombination mechanisms are orientation sensitive. The multimer yield ratios agree with data from experiments on Ni surfaces(10).

On the basis of these results the strengths and weaknesses of the computer model method can be evaluated with confidence. As far as they go, the model results agree well with the experimental data. Because of the nature of the model no attempt has been made to include information about the charge state of an ejected particle. It is possible to state with some assurance the mechanisms which form clusters and the points of origin in the crystal from which the individual atoms were emitted. The computer model gives atomic point of origin information that cannot be confirmed by experiment, but these correlations at the atomic level give insight which will aid in the interpretation of data. Thus the computer models and SIMS experiments are synergistic. In the simple systems studied to date the combination gives much more information than either taken separately.

The two element Cu/Ar^+ system has also given insight into the importance of the interatomic potential functions to the entire calculation. At the present stage of development of the method the only adjustable parameters remaining in the model are those which characterize the potential functions. One can argue that binary potential functions do not fairly represent the actual interatomic interaction in a crystal, but as the collision interaction energies increase the potential approximation improves and this is the only computationally feasible approach at this time. The choice of two-particle potential functions does not limit the computer model to binary collisions, because three atoms can be simultaneously in collision. Some experimentally measurable quantities, the total sputtered atom yield for example, are sensitive to the choice of potential function. The energy dependence of the yield was used to establish the potential parameters now in use. Other useful information, such as the surface point of origin of the atoms, is quite insensitive to the potential function. The actual number distribution of multimers formed depends upon the particular choice of a gas-phase potential function for the Cu_2 molecule, but the existence of multimers does not.

Currently we are studying oxygen reacted copper surfaces(11). Using spectroscopically known molecular potential functions, O_2 and CuO molecules form above the target surface. The presence of the oxygen atoms make a small change in the energy distribution of the sputtered Cu atoms, enhancing a second peak which appears as a shoulder on the maximum of the clean surface distribution.

The pure metal simulations produce "spot patterns", a representation of the angular dependence of the emission of target atoms, which are in excellent agreement with corresponding experimental patterns. By selecting only those particles with rather high kinetic energy, > 20 eV, the fraction of particles ejected along preferred cystallographic directions is considerably enhanced. The lower energy particles tend to have a diffuse angular distribution since they are ejected late in the trajectory when much of the surface order is no longer present.

The adsorbate atoms were placed in a c(2x2) array above the Cu(100) surface plane. Since appropriate pair potentials for CuO at the lattice surface are unavailable, we have assumed a form similar to the copper potential with a binding energy adjusted to yield a reasonable adsorption energy. The height above the surface for the A-top site and for the 4-fold and 2-fold bridge sites were calculated from their equilibrium separation distance (6) to be 2.55, 1.74 and 2.13 Å, respectively.

The angular distributions calculated from the three candidate registries are quite different, particularly when only the high energy sputtered atoms are collected. Thus it appears that a collaboration between simulation and angular dependent SIMS can determine the registry of regular adatom structures on metal surfaces.

To summarize, in the course of the investigation of sputtering by classical trajectory simulation a great deal of information has been gathered about the model, the (Cu/Ar^+) and the $(Cu(O)/Ar^+)$ systems. The success of the model encourages its application to more complex problems.

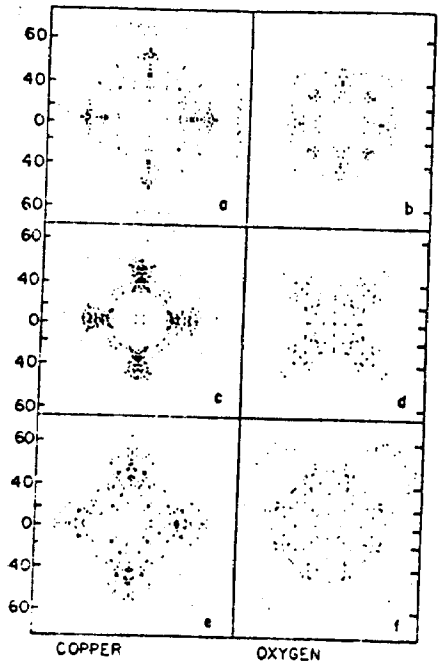

Figure 1. Angular distributions for the copper and oxygen originating from a c(2x2) structure with oxygens in an A-top (a,b), 4-fold bridge site (c,d) and 2-fold bridge site (e,f). The kinetic energy of the copper atoms is >20 eV for each case, while the kinetic energy for the oxygen atoms is between 20 and 50 eV. The binding energy for oxygen on copper is arbitrarily assumed to be 0.75 eV. The numbers on the ordinate refer to the polar deflection angle given in degrees.

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