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Simulating Cluster-ion Impacts

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Abstract. Although cluster-ion interactions may superficially appear to resemble those in nuclear physics, there are important differences. First, at the energies of interest the deBroglie wavelength of an atom in the cluster is so short compared to atomic dimensions that the atom's trajectory in a solid is classical. Thus, one may use the Born-Oppenheimer approximation in which the particles all obey Newton's equations of motion and only the interaction potentials reflect the quantum mechanical character of the system. Thus, we can treat many atomic interaction processes in the semi-classical limit; in this paper I shall, for example, indicate how in this way one can include the effect of atomic excitation in collisions. Second, the cross sections involved are much larger than those for nuclear interactions, i.e., the mean free paths of the "reaction products" are so short that large collective effects always occur. This means that the calculations must deal with large numbers of interacting particles. For the case of cluster ions at MeV energies this has required us to develop new computational strategies, e.g., the use of massively parallel computers. By using such simulations strategically we are able both to optimize the design of experiments and to interpret their results less ambiguously.

INTRODUCTION

The interaction of molecular (cluster) ions with targets yields a rich variety of complex phenomena, and as a consequence , designing experiments so that they elucidate the mechanisms involved is not readily achieved by trial and error. For this reason simulation techniques provide a possible means for assuring efficient observation and interpretation. In the present context, i.e., energetic cluster ion bombardment, the intent is not to attempt a fully complete description of reality; rather, the hope is that by means of a cleverly chosen caricature of the experiment to expose its essence.

For large clusters with energies of MeV/atom the Born-Oppenheimer approximations expected to be appropriate; thus, the trajectories of both the atoms in the cluster and the solid evolve classically, and the quantum behavior of the electrons is buried in the ion-atom and atom-atom potentials. Choosing these potentials can be done at many levels of sophistication; that is too

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extensive a subject to describe in detail here. The general form of such a potential contains a repulsive core that results from the Coulomb field of the nuclei shielded by the electrons in tightly bound shells and the Pauli repulsion that occurs when these electron orbits in the colliding atoms are brought into proximity. A reasonable approximation to these cores is the Molière potential, which is based on a Thomas-Fermi estimate (1). Ab initio calculations of the core potential have also been done, but since they usually quite closely resemble the Molière estimate, they are not really worth the extra effort.

At greater distances the atom-atom potentials are attractive, their magnitude and shape can be determined by the properties of the target, e.g., sublimation energy, elastic constants, crystal structure, etc.. In the least sophisticated simulations these are taken to be potentials between individual pairs of atoms; somewhat more realistic interactions are taken from effective-medium recipes. A popular form of the latter that is often used is the Embedded Atom Method (EAM) (2), which exploits the Hohenberg-Kohn demonstration that the local potential felt by an atom is a function only of the local electron density (3). Thus, the EAM (and its related forms) includes some many-body interactions and deals more accurately than pair potentials with regions of limited symmetry, e.g., surfaces, crystalline imperfections, localized clusters of atoms, etc. In low energy ion bombardment in which the trajectories of ejected atoms are determined in detail, it has been demonstrated that EAM potentials give results that are in significant better agreement with the data (4).

When the energy of an incoming cluster is in the keV/atom range, collisions between cluster constituents and target atoms cause the inner electronic shells of a colliding pair to come into contact. This provides the opportunity for electronic excitation to take place. Including such effects can be accomplished within the method used to integrate the Newtonian equations of motion for the many-body system composed of target and cluster atoms. Generally, in such molecular dynamics (MD) simulations one must employ a variety of tricks to reduce these integrations from their natural scale of N^2 (where N is the total number of atoms involved, 10^3-10^5) to something more like $N\ lnN$. In addition, one often uses an adaptable increment in time, in which shorter time steps are used only when a pair of atoms is undergoing a hard collision (where large accelerations occur). When such a colliding pair reach a distance of closest approach at which electronic excitation can occur, a probability of excitation can be included as a test (5). If the excitation is predicted to happen, the appropriate amount of excitation energy is extracted from the interaction by separating the pair slightly, i.e., letting them "slide" down the steep repulsive core potential just enough to remove the excitation energy (6,7). Then the system is allowed to continue its classical evolution.

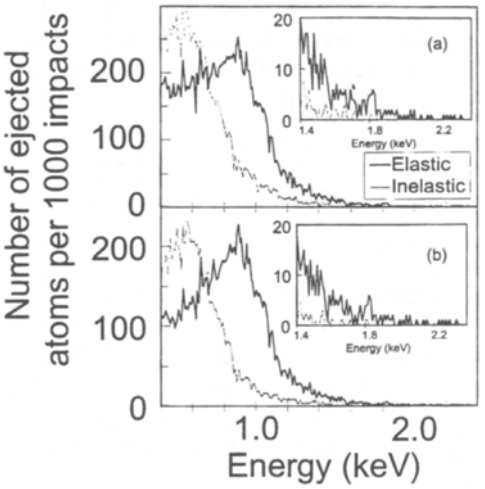

FIGURE 1. Energy distribution of (a) all rebounding cluster atoms and (b) excited, rebounding cluster atoms for an Al_{63} cluster impacting on an Au(111) target at 1.5 keV/atom [from ref. (9)].

For the case of keV/atom incident clusters the effects of such electronic excitation are predicted to be insignificant — with one interesting exception. If the cluster contains atoms that are lighter than the atoms of the target, an appreciable fraction of the light cluster atoms rebound from the surface. Some of them have energies greater than they had initially, which is due to scattering of incoming cluster atoms with some from the front of the cluster that have already rebounded — as in a colliding beam accelerator (8). These collisions involve small distances of closest approach and hence a high probability of inelastic scattering due to electron promotion.

In Figure 1 are shown the calculated energy spectra of rebounding Al atoms predicted for 1.5 keV/atom Al_{63} cluster ions incident on an Au surface. The inclusion of electronic excitation lowers dramatically the peak in the expected energies of these reflected atoms (9).

MeV/ATOM CLUSTER ATOMS

Although all the techniques described above can be applied at higher incident energies, difficulties appear that are both technical/calculational and fundamental. We are dealing with the former at the present time; the latter represent a frontier that we are just beginning to define, which will be

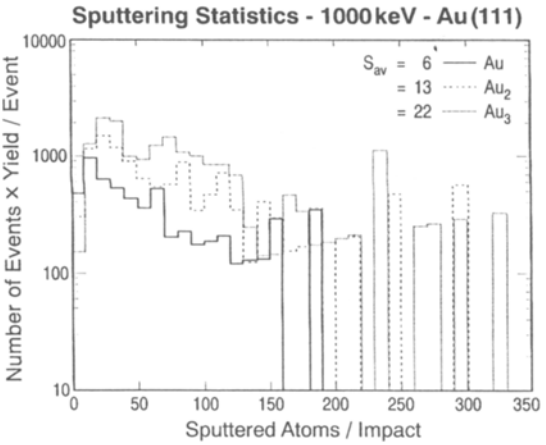

FIGURE 2. The contribution to the sputtering yield as a function of multiplicity for Au_n^+ on an Au(111) crystal.

discussed in the last section.

Higher incident velocities require shorter time steps in the integration of the equations of motion. The fact that the ion-atom interaction cross sections are smaller at higher energies means that fewer collisions occur; however, those that do occur can have enormously higher energy transfers. Thus, we have poor statistics, i.e., many simulations yield no collisions, but a rare hard collision can set many more atoms in motion. Figure 2 shows the frequency of occurrence of events in which N Au gold atoms are ejected ("sputtered") from an Au target by Au_n^+ cluster ions at 1 MeV/atom. In this figure the frequency of occurrence is weighted by the number of atoms sputtered in the event, this gives a clearer picture of how events of different multiplicity contribute to the total yield of sputtered atoms. For keV/atom single-ion sputtering this sort of distribution is very Poisson-like; here the contribution is roughly equal for each multiplicity. (Note that the average number of sputtered atoms for incident Au_3^+ in this figure is 22; approximately half the events yield no sputtered atoms. In this situation the variance is more significant than the average.)

Dealing with such strange statistics is a computational nightmare, in that one has to run many simulations in order not to miss the few that have exceptionally large multiplicities. We have dealt with this problem by using the 512 node Intel Paragon at Caltech. (each node is equivalent to a high performance work station like an IBM RS-6000.) One node manages the 511 others: it loads a different simulation into each; when a particular node

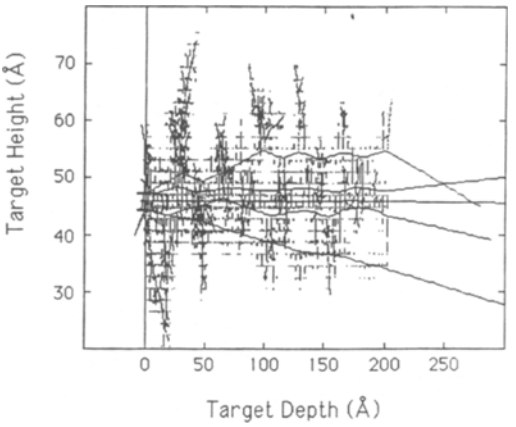

FIGURE 3. Side view of a 100 keV/atom Au_5^+ cluster impact on a Au(100) target showing movement between channels and recoiling lattice atoms for the five atoms from the cluster.

finishes, the manager node extracts its results and provides it with a new simulation to perform. In this way all nodes are kept busy, and any node that has a large event can take as long as it needs to finish (10).

In these sputtering simulations, it is not necessary to use targets that are thicker than ~ 20 atomic layers. However, if one wants to simulate how the trajectories of the atoms in a cluster evolve as they move through a target, then target thicknesses of 10^2-10^3 atomic layers must be used. We have done this in a few cases; for example, a molecular dynamics (MD) simulation of 100 keV/atom Au_5^+ in Au is shown in Figure 3. Here we show not only the paths of the cluster atoms but also those of the target atoms that have been set into motion. An MD simulation of this sort takes several days to run, which is a bit too long even on the Paragon to accumulate a statistically significant number of events.

If one doesn't need to keep track of the motion of the all the moving target atoms, a different calculational technique we can be used — the binary collision approximation. In this method one can just move each incident atom on straight-line trajectories between collisions, which occur for different collision impact parameters at each mean-free-path distance along an ion's path. This type of calculation runs very quickly; it is the basic method used in popular codes like TRIM (11). The price one pays, however, is that since each incident atom is stepped forward in distance to its next collision, the separate atoms in the cluster are not locked to a common time step; hence,

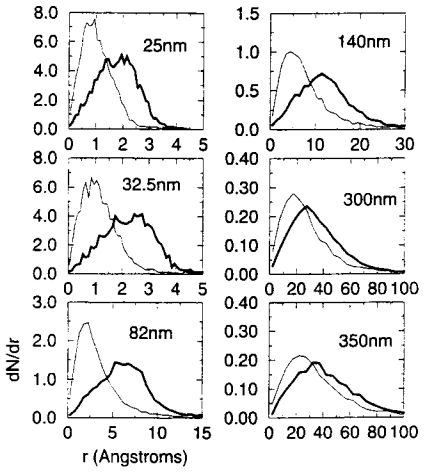

FIGURE 4. The radial distribution of exit points for 2.0 MeV/atom C_5 clusters on amorphous carbon targets of various thicknesses (2.5 to 350 nm). The heavy line is for simulations that include the partially shielded Coulomb interactions of the cluster atoms; the light lines neglect these interactions [from ref. (12)].

one cannot easily include their mutual interactions.

Recently, we have developed a hybrid computer code that runs as fast as in the binary collision approximation, but includes the interaction of the cluster atoms (12). In Figure 4 are shown the radial distribution of exit points for 2 MeV/atom carbon atoms from C_{10} incident on amorphous carbon targets of various thicknesses. The heavy curve includes the partially shielded Coulomb interactions of the cluster atoms; the light curve neglects them. As you can see, for thin carbon targets there is a significant difference (12).

In our simulations for low energy bombardment we have discovered that there is a significant probability for rebounding or sputtered atoms from high multiplicity events to recombine above the target surface to form highly excited atomic clusters of target atoms (13,14). In Figure 5 we show the number of atoms and internal excitation energy of a 5 eV/atom Au_{108} cluster rebounding from a barrier. Here, the original cluster fragments rapidly, and we follow the largest target fragment as it then evaporates an atom at a time, which is the dominant cooling mechanism on these time scales (13).

The size distribution of clusters from low energy sputtering was a mystery until we plotted the distribution versus multiplicity for our simulation of 3 keV Ar^+ on Cu (14). This showed convincingly that when viewed from this perspective, that simple combinatoric estimates are in excellent agreement with the experimental data. Thus, our next project will include the estimation

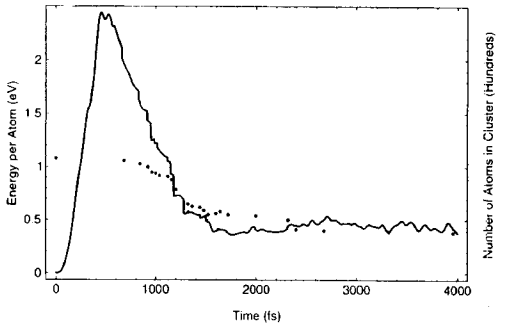

FIGURE 5. The thermal energy per atom in the largest target fragment is plotted versus time (solid line) for a "head-on" impact of a (100)-Au_{108} cluster (5 eV/atom) on a two-dimensional barrier. The number of atoms in this fragment is also plotted (dots, units of one hundred). This figure was taken from reference (13).

of emitted cluster yields from the very large events we find in the Au_n^+ on Au simulations.

THE FUTURE

The basic difficulty that occurs at MeV/atom cluster energies is that we have left out a major piece of the physics. At MeV/atom energies the dominant portion of the energy transferred from the cluster to the target is due to energy loss to the target electrons. Although we include such drag effects on the moving atoms, and as we discussed earlier we can include deep electronic excitations as well, we have not included the effect of this heating of the target electrons on the atom-atom potentials. Since MeV/atom C_{60} projectiles give $\sim$ 50 keV/nm to target electrons, this is not a negligible effect. For example, 20 MeV C_{60} bombardment of Ti has been shown to leave enormous damage tracks — much larger than those of GeV Pb ions (15).

Calculations that include these electronic heating effects are likely to be much more difficulty than anything we have attempted so far. The transport of secondary electrons, the Auger electrons from the decay of deep electronic vacancies, and the high plasma frequency of the target material all contribute to the complexity of the process. All must, however, be included if we are to deal realistically with the sputtering and damage phenomena involved. Since we have already seen experimental indications of qualitatively new phenomena that occur (15), such simulations are definitely worth our continued efforts. In this way we hope to continue to guide the course of the next generation of experiments in this field.

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