Many-Body Effects in Atomic-Collision Cascades

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Molecular-dynamics simulations have been used to identify two cooperative atom-ejection mechanisms which increase sputtering yield. Their effects are analogous to the "thermal spike," "shock wave," and "reduced binding energy" sputtering mechanisms. They are examples of nonlinear, many-body effects in cascades which go beyond collisions between randomly moving particles.

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When metal surfaces are bombarded with high-energy molecular ions, many experiments have determined that the average number of atoms ejected *per nucleus,* the *molecular ion sputtering yield,* is significantly greater than the corresponding *atomic ion yield* for the constituent atoms.[1-4] This "molecular ion effect" is usually ascribed to "nonlinear effects" in the target's atomic collision cascade.

Most authors agree that nonlinear processes correlate strongly with high—energy-density atomic-collision cascades. Because a comparison between the cascades created by heavy atomic and diatomic ions appears to allow direct control over the energy density of similar cascades, evidence of correlations between high energy densities and nonlinearities obtained by this approach are deemed particularly instructive.

The term "nonlinear" implies different effects to different people. For example, deviations of experimental high-energy heavy-ion sputtering yields from the linear theory of sputtering values have been attributed to nonlinear effects, which are described as collisions between moving atoms.[5] Recent simulation studies which examined the time development of the ratio of the number of these nonlinear collisions to the total of all "collisions" do not support this conjecture.[6] In this context a collision was defined to be an interaction in which the potential energy exceeds a specified *positive* threshold.

Even in amorphous solids, the theory of nonlinear cascades is a dauntingly complex problem; so all theoretical approaches to the problem require simplifying assumptions. Additional atom-ejection mechanisms, called here the "supplemental" theoretical models, attempt to predict an "excess yield," beyond that which would occur in a "normal," i.e., low—energy-density, cascade. The oldest such model is the *thermal spike.* In it excess ejections come from a long-duration locally overheated target region. The thermal-spike model, including related variants, has an extensive literature with analyses still being produced.[7,8] Another approach assumes that the collision cascade generates *shock waves.*[9,10] This collective motion of particles then produces moving regions of high temperature and pressure, which eject atoms upon reaching the surface. A third, less completely developed, model suggests that excess ejections can be explained in terms of a *reduced surface binding energy*[5] in surface regions which have been severely disrupted by the high—energy-density cascade.[2,11]

At present, direct experimental support for these proposed models is limited, and key discriminating experiments are unidentified. Experimental verification is complicated by the large uncertainty of various parameters, which make theoretical predictions imprecise. Zalm and Beckers[12] point out some difficulties of interpretation inherent in these models. In this environment, molecular-dynamics computer simulation becomes an appropriate research tool.

The calculations which led to this paper are part of a research program undertaken to identify the source(s) of discrepancies between simulated and experimental ejected-atom yields. Although these discrepancies do not negate the proven effectiveness of simulations as research tools, any unresolved difference is necessarily a source of concern. The mechanisms described below have been identified over a wide range of ion energies and ion-target systems; so the total-yield difficulty is not relevant here. The simulations do not support any of the proposed models in detail, but there are similarities to the supplemental theories. Each contains elements of truth, but all oversimplify the actual situation.

These simulations investigated heavy-atom targets, sputtered by heavy atomic and diatomic ions at low and high energies. For comparative purposes, results were obtained from two logically dissimilar simulations: one is a form of TRIM.SP[13,14] modified to follow lateral motion, the other a recently developed *hybrid* program, QDRIM, which incorporates a multiple-interaction (MI) treatment (as developed in QDYN[15,16]) for the *single-crystal surface zone* of the target with a TRIM.SP treatment of particles which penetrate into an underlying *amorphous zone.* More details of QDRIM will be published elsewhere.[6] Because of its binary-collision (BC) logic, TRIM.SP necessarily assumes a dilute, low—energy-density, *linear* collision cascade. Both simulations used Molière potential functions. As in QDYN,[15] an attractive well is added to the Au-Au potential in QDRIM's surface zone. Inelastic (electronic) energy losses were omitted from all calculations.[17]

The upper proton of Fig. 1 shows the sputtering yield

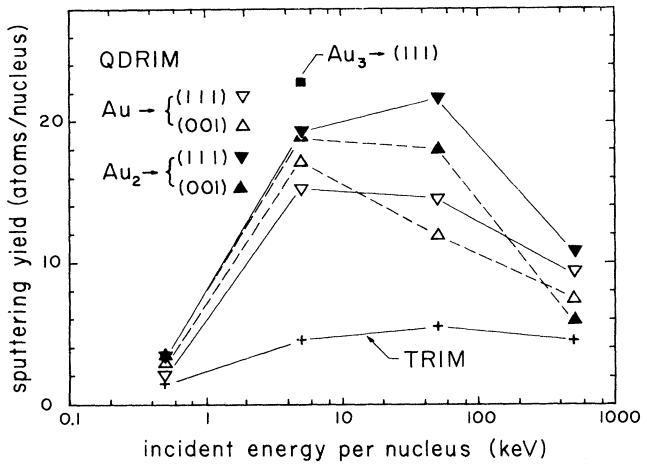

FIG. 1. Sputtered-atom yields, *normalized to atoms/nucleus,* for Au and Au_2 "ions" on Au(111) and (001) surfaces from QDRIM, and for Au "ions" on "amorphous" Au from TRIM. QDRIM yields from "amorphous Au" are not significantly different. One point indicates the yield/nucleus of a triatomic, Au_3, "ion."

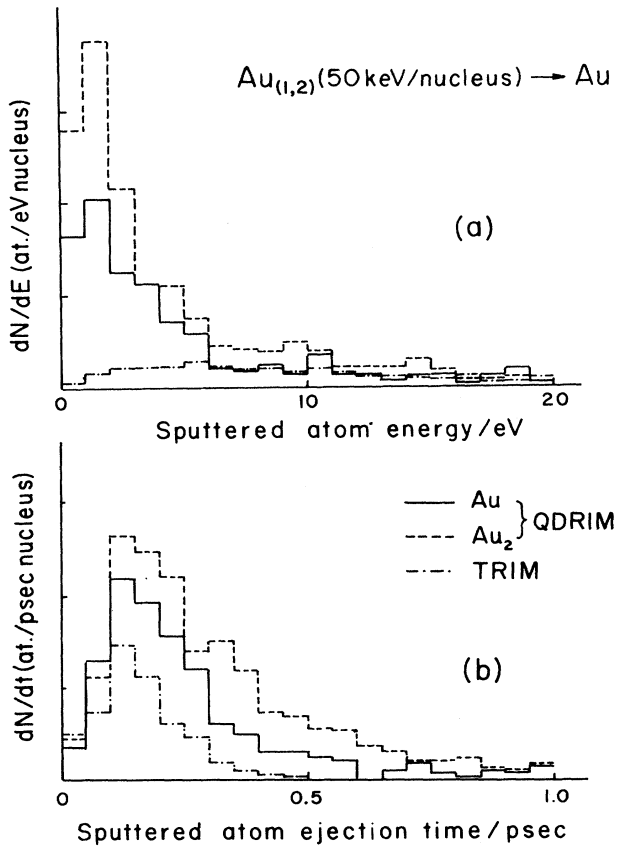

FIG. 2. (a) Ejected-atom energy distributions calculated by QDRIM, for Au and Au_2 from crystalline Au, and by TRIM, for Au from amorphous Au. (b) Atom time-of-ejection distributions, normalized to the same number of incident nuclei, for the same calculations.

per nucleus as a function of the bombardment energy, calculated by QDRIM, for Au^+ and Au_2^+ "ions" (projectiles) on (111) and (001) surfaces of an Au target. Similar computations, performed with an amorphous Au QDYN zone, show no significant deviations from these results. The lower part of Fig. 1 shows comparable calculations performed using TRIM.SP in an amorphous target of the same surface area as the QDRIM calculations. The TRIM calculations were constrained in this way to minimize the differences between the two computations.

Although the simulations differ only in the surface-zone treatment, QDRIM's sputtering yields are strikingly higher than TRIM.SP's. Still, QDRIM's molecular-ion excess ejection is smaller than the experimental effect. The *yield ratio (per nucleus),* $Y(Au_2)/Y(Au)$, never exceeds 3, while experimental ratios up to 5 exist for comparable ion energies and ion-target combinations.[2] TRIM logic simulations cannot calculate molecular-ion yields directly, but yield increases from approximations of molecular-ion bombardments are comparable to the QDRIM results. The point indicated with a square was computed for Au_3 to show that a larger ionic cluster does not significantly increase the yield/nucleus.

Figure 2 shows the ejected-atom energy and ejection-time distributions for atomic and diatomic "ion" bombardment at 50.0 keV/nucleus from QDRIM and TRIM.SP, normalized to the same number of bombarding nuclei. Both energy and time distributions are quite different. The ejection-time distributions from the MI calculations have longer tails than TRIM's, and QDRIM's diatomic-ion distribution's tail is much longer than the atomic ion's. TRIM.SP produces few low-energy ejected atoms, while, like QDYN, QDRIM's energy distribution closely approximates experimental distributions.[15] Diatomic ions produce an excess of low-energy ejected atoms compared to the atomic-ions' distribution. One concludes that the near-surface particle dynamics differ in MI and BC simulations.

Detailed analyses of high-yield QDRIM trajectories identify two yield-enhancing *cooperative effects,* which cannot exist in either a linear transport theory or a BC simulation. The first, shown in Fig. 3(a), deals with the behavior of a small *bunch* of near-neighbor atoms. The bunch in Fig. 3(a) consists of nine atoms, initially resident in the first and second layers of the target. Each atom is labeled by a number which is the time in femtoseconds at which it was "ejected." In QDRIM/QDYN that means that the atom has moved up to a position more than half a lattice unit above the surface with sufficient outward velocity to overcome its binding forces.

Atom 376, which is ejected later in the cascade (well after its near neighbors), illustrates the mechanism. The net force binding it *to the bulk material* is greatly reduced from the undamaged crystal value. Atom 376 in effect is

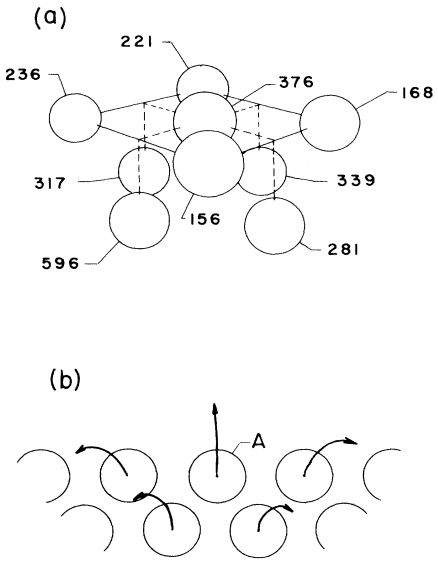

FIG. 3. (a) A *bunch* of atoms, each labeled with its ejection time (in femtoseconds), which includes the atom labeled 376. Atom 376 is ejected by the first mechanism (see text) because the forces binding it to the surface have been reduced by the prior ejection of the rest of the bunch at the indicated times. The previously ejected atoms help to pull it away from the surface. (b) In this qualitative sketch of a bunch, the central atom, *A*, is ejected by the second mechanism (see text). The moving atoms surrounding *A* fall back; so each helps *A* to escape by increasing the relative velocity between *A* and each of its neighbors.

dragged away from the surface by its departing neighbors! For Au, the nominal surface binding energy is 3.81 eV, but atom 376 never has more than 1.5 eV of *kinetic energy*. Similar events have been reported previously.[18]

Figure 3(b) sketches another cooperative ejection mechanism, which occurs in a bunch of near-surface atoms. In this case, the members of the bunch all have some kinetic energy, and are moving away from the surface together, but *none has sufficient energy to escape from the undamaged surface.* In a BC model no ejections occur. In an MI model, where the "surface binding energy" is the sum of pairwise interactions, the relative kinetic energy of each pair of atoms is important. If one member of the pair reverses its motion, now falling back toward the surface, its acceleration increases the *relative velocity* between it and the other member of the pair. As a result, the relative kinetic energy may exceed the attractive potential energy between that pair, and they can separate. Using this mechanism, in an MI model, an atom in the bunch may obtain sufficient relative kinetic energy to overcome the *sum of all pair-wise forces between it and the members of the bunch;* so it escapes as the other atoms fall back to the surface. In this case, the members of the bunch promote one of their number into ejection, even though the majority do not escape.

This mechanism is particularly important for high-yield trajectories. Early in such cascades most atoms in the surface layers convert "transverse" recoil momentum into outwardly directed velocity components, causing an initial "lift" of the surface layer(s) until the motion is reversed by attractive forces from the bulk of the target. During this collective phase some atoms are ejected by the second mechanism, because the remaining forces are unable to reverse their outward velocity. In an actual cascade these mechanisms are not so well separated as Figs. 3(a) and 3(b) would seem to indicate. They tend to occur simultaneously, with emphasis toward one or the other. Their contribution to the total yield could only be quantified by assuming arbitrary thresholds leading to artificial categories. These mechanisms do not require high bombardment energies, or large energy densities. The examples shown in Fig. 3 were calculated at 3.0-keV bombardment energy with atomic ions.

Both mechanisms increase the relatively late ejection of low-energy atoms. Together, they help to explain the differences in the ejected-atom distributions of the two simulations. Both are examples of collective mechanisms which reduce the "effective" forces binding *individual atoms* to the surface. To this extent they are consistent with the "reduced surface binding energy model," but more specific.

The bunch is a region of excess energy, but atom motions are correlated and the mechanisms are cooperative, not local high-temperature effects. Despite some similarities, one cannot say that the yield excess is produced by enhanced evaporation, as from a thermal spike. In some sense, the lifted surface region has shock-wave-like characteristics, but it is quite different because in that model of sputtering the excess yield is caused by a thermally induced pressure.

The supplemental theoretical models are qualitatively similar to the mechanisms described here, but in simulations they would be temporally distinguishable from the identified mechanisms because they are presumed to occur later in the cascade.

To the extent that "thermal spikes" and "shock waves" can be viewed as many-body cooperative phenomena, MI simulations provide comparable effects. Both proposed mechanisms provide some qualitative insight, but neither is identifiable within the description of the cascade provided by simulation. The mechanisms seen in the simulations are true dynamical many-body effects. Despite the similarities, past experience has shown that individual MI trajectories do not develop quantitatively identifiable shock waves or thermal spikes.[15] The dendritic structure of a collision cascade precludes the development of a coherent front in an individual cascade,[19] and in MI simulations, long-lasting high-temperature regions make only minimal contributions to the total ejected-atom yield.[19]

In summary, cooperative mechanisms analogous to pre-

viously proposed supplemental atom-ejection models have been observed in MI simulations of atom ejection, and many-body processes similar to those reported here undoubtedly occur throughout a collision cascade. MI simulations of atomic-collision cascades produce a dynamical picture consisting of a dendritic structure containing cooperative effects produced by correlated particle motion. The cooperative atom-ejection mechanisms depend on more sophisticated many-body processes than individual collisions between two moving particles. These mechanisms enhance the ejected-atom yield over that predicted by linear cascade theory, but the enhancement is not sufficient to explain large molecular-ion effects.

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