Simulation of Cluster Impacts on Metallic Surfaces

M. H. Shapiro
Physics Department, California State University, Fullerton, California 92634

T. A. Tombrello
Division of Physics, Mathematics and Astronomy, California Institute of Technology, Pasadena, California 91125
(Received 17 January 1990)

Energy, number-density, and confinement-time properties of 1-keV/atom Al_{32} and Al_{63} cluster impacts on Al and Au targets have been investigated by molecular-dynamics simulation. Inertial confinement of the incoming clusters leads to number-density increases of ~ 2 for very short time periods (< 20 fs) in the primary impact zone. Simultaneous and near-simultaneous multiple collisions occasionally increase the potential energy of a particle to more than twice that allowed in isolated two-body collisions. Up to 12% of the kinetic energy of the incoming cluster is transferred to atoms which eject early in the collision cascade.

PACS numbers: 79.20.Nc, 02.70.+d, 79.20.Rf

Cluster bombardment of solid surfaces produces very high transient particle and energy densities in very small regions of the target.[1,2] This results from the fact that large clusters (up to a few hundred atoms) with moderate energies (up to a few hundred keV) lose most of their energy through elastic nuclear collisions, rather than electronic processes.[2] Thus, a cluster impact can produce far more localized damage than an individual ion impacting with comparable energy. This Letter reports the results of simulations carried out with a modified version of the molecular-dynamics (MD) sputtering code[3] SPUT2 on model cluster-target systems in an attempt to understand better the behavior of the early stages of energetic cluster impacts on metallic targets. In particular, we have investigated the enhanced erosive effects of such collisions, and the energy and density properties of the early stages of the collision cascade for systems where the interatomic potentials are well understood.[4] To our knowledge, this is the first full three-dimensional MD simulation of energetic cluster impacts on solid surfaces.

Recently, Beuhler, Friedlander, and Friedman (BFF) have used accelerated cluster ions in an attempt to initiate D-D fusion reactions.[5] In their experiments singly charged $(D_2O)_n$ clusters with $25 \leq n \leq 1300$ and energies up to 325 keV impacted deuterium-loaded Ti targets. They state that they have observed D-D fusion at extraordinarily high rates in these experiments ($\sim 10^{-11}$ per incident cluster). Since such a high fusion rate would require an unrealistically high fusion cross section for an uncompressed system, BFF have speculated that the incoming clusters create shock waves that result in significant compression of the cluster and target material.[5] While it was not our purpose to investigate in detail the nuclear physics of cluster-impact fusion, our results on the purely collisional aspects of cluster impacts allow us to comment on the possibility that enhanced density or energy effects are responsible for the high fusion rates seen by BFF.

Our calculations were carried out for 63-atom Al clusters impacting normally on Al and Au targets, and for 32-atom Al clusters impacting normally on Au targets. In each case the incident clusters were given an energy of 1 keV per cluster atom (i.e., either 63 or 32 keV total energy). In most of the calculations six-layer, fcc single-crystal targets containing 1875 atoms were oriented with their (100) faces perpendicular to the incoming clusters. A few trajectories were computed with similar twelve-layer targets containing 3750 atoms. For computational simplicity, the clusters themselves were chosen to be aluminum crystallite cubes oriented with their (100) faces perpendicular to the direction of incidence.

In the SPUT2 code, interactions between atoms are represented by a linear superposition of two-body potentials. Repulsive Moliére potentials[6] joined smoothly to attractive Morse wells[7] by cubic splines were used to describe the interactions between target atoms and between cluster atoms. The parameters for the two cases (Al-Al and Au-Au) were the same as those used for previous molecular-dynamics simulations.[4,8] The interactions between cluster atoms and target atoms were described by Al-Au and Al-Al Moliére potentials cut off at 2.6 Å.

In addition to the usual information about sputtered atoms (which will be reported in detail elsewhere), the modified program computed the total potential energy of each atom in the cluster-target system at each time step. Whenever the total potential energy for an atom exceeded that which could be achieved in an isolated two-body collision with the target atom at rest (500 eV for Al-Al collisions and 774 eV for Al-Au collisions), the information was recorded in a file. The modified program also produced a count of the number of particles in a small cubical region of the target, which had its front face centered on the primary impact zone of the target at each

TABLE I. Energy loss to ejected atoms.

Cutoff time / System	100×10^{-15} s		500×10^{-15} s	
	Number of impacts	Energy loss per impact (keV)	Number of impacts	Energy loss per impact (keV)
Al_{32}-Au	300	3.97	100	5.11
Al_{63}-Au	200	6.90	100	9.18
Al_{63}-Al	300	0.30	100	1.09

time step. The size of this box was chosen to be 2.5 lattice constants on a side (10.2 Å for the Au targets and 10.1 Å for the Al targets).

For the (100) targets used in these simulations, the primary impact zone is a square region retaining the basic symmetry of the target. This region is divided into 100 small squares which are scanned uniformly. Within each small square the impact coordinates are chosen randomly. For each of the cases studied, trajectories for 100 to 300 impacts were computed with cutoff times of 100 and 500 fs (see Table I for details). The time required to compute an individual trajectory on a two-processor Apollo 10000 workstation varied from approximately 10 min to 1 h depending on the number of atoms in the cluster and the cutoff time.

Number-density and confinement-time information for the three systems that were studied are given in Fig. 1 for several typical impacts. Sharp transient increases in number density in the primary impact region generally peak within the first 20 fs. For both the Al_{63}-Al and the Al_{32}-Au cases peak number densities are close to the maximum allowable (1.22×10^{29} and 8.95×10^{28} m^{-3}, respectively). For the Al_{63}-Au case peak number densities typically are about 85% of the maximum allowable value (1.19×10^{29} m^{-3}). Confinement times, estimated from the full width at half maximum (measured relative to the *initial* density of the target) of these number-density versus time curves, typically are about 20 fs for the Au targets and about 15 fs for the Al targets. Following the initial compression of the primary impact zone, material is expelled rapidly. Within 100 fs number densities drop to less than 25% of the initial target density for impacts with the 63-atom clusters. The 32-atom clusters are only slightly less effective in removing material from the primary impact zone. Almost no difference in number-density or containment-time values were seen when the same impact points were used on twelve-layer targets instead of six-layer targets.

The behavior of the potential energy for individual atoms in the collision cascades is shown in Fig. 2, where the number of time steps for which a given potential energy per atom is achieved are plotted. (For the Al_{32}-Au and Al_{63}-Au systems only the results for Al atoms are shown.) Because several time steps may be required to complete a collision, these plots overestimate the number of *collisions* for which a given potential energy is achieved. However, for the higher-energy events the overestimate is small. For aluminum clusters striking gold targets potential energies up to 1628 eV were observed, while for aluminum clusters striking aluminum targets potential energies up to 722 eV were seen in the simulations. The first value is slightly more than twice the maximum potential energy that can be achieved in an isolated two-body collision between a moving aluminum atom and a stationary gold atom.

A significant fraction (up to 12%) of the kinetic energy of the incident clusters ultimately is transferred to atoms that eject from the surface relatively early in the

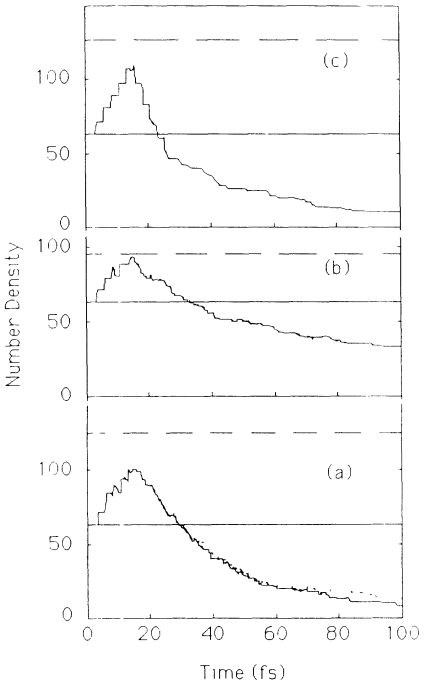

FIG. 1. Number density vs time for typical cluster impacts for the (a) Al_{63}-Au, (b) Al_{32}-Au, and (c) Al_{63}-Al systems. The dashed curve in (a) was computed with a twelve-layer target. The solid horizontal lines represent the initial number density of the target, and the horizontal dashed lines represent the number density that would be reached if all cluster atoms were confined with no loss of target atoms. The vertical scales in (a) and (b) should be multiplied by 9.38×10^{26} cm^{-3}, and that in (c) should be multiplied by 9.68×10^{26} cm^{-3} to obtain absolute number densities.

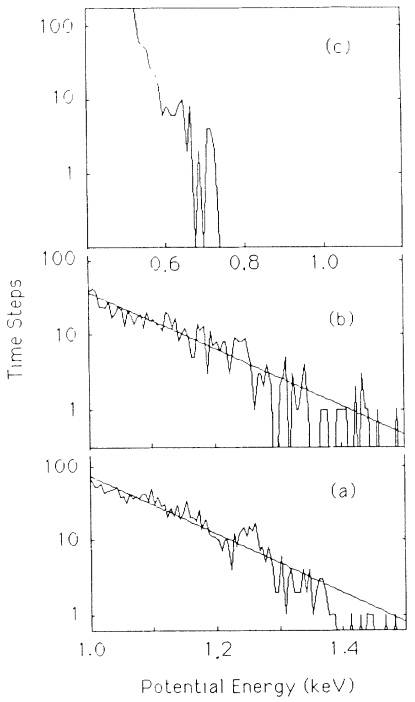

FIG. 2. Number of time steps at which given potential energies per atom were reached for the (a) Al_{63}-Au, (b) Al_{32}-Au, and (c) Al_{63}-Al systems. Only results for aluminum atoms are shown in (a) and (b). The vertical scale in (a) should be divided by 200 to obtain time steps per impact. Similarly, those in (b) and (c) should be divided by 300. Note the different energy scale in (c). The solid lines represent least-squares fits of decaying exponentials to the simulation results, the parameters of which are given in the text.

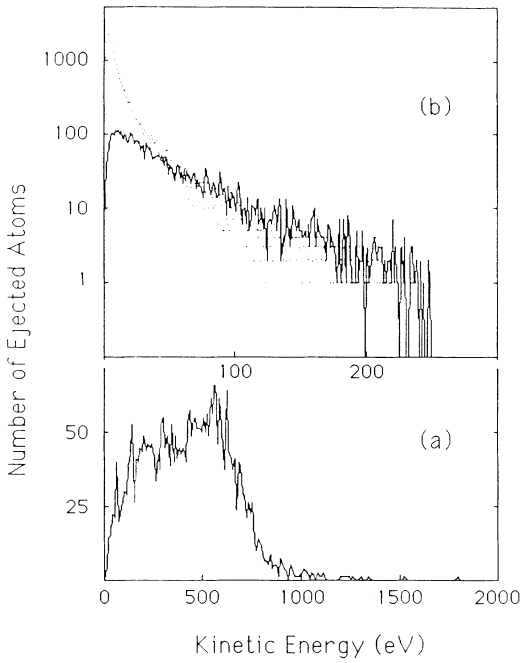

FIG. 3. Energy spectra for (a) aluminum and (b) gold atoms ejected during the first 100 fs in Al_{63} impacts on gold targets (solid lines). The energy spectrum for gold atoms ejected during the first 500 fs is shown dotted. Note the different energy scales.

collision cascade (by 100 fs) for aluminum-cluster impacts on gold, but not for aluminum-cluster impacts on aluminum targets (see Table I). To a large extent this is a result of a significant loss of atoms with high kinetic energies from the aluminum clusters as they impact the gold target. Energy spectra for both aluminum and gold atoms that eject during the first 100 fs of the collision cascade are shown in Fig. 3 for the Al_{63}-Au case. A small number of the aluminum atoms eject from the cluster with kinetic energies above 1 keV, while most eject with energies of several hundred eV. The energy spectrum of the sputtered gold atoms also is exceptionally hard, which reflects the highly nonlinear nature of the collision cascade even in its early stage.

We can draw the following conclusions from the simulation results presented in Figs. 1–3 and Table I:

(1) Cluster impacts in this size and energy range (30–60 atoms and $\sim$1 keV/atom) effectively remove material from the primary impact zone through collisional processes, creating microcraters in the target surface.

(2) Transient compression of material in and near the point of impact is significant; however, the compressed material remains confined for an extremely short period of time. Confinement parameters ($n\tau$) computed from the information in Fig. 1 typically were found to be about 3×10^{14} m^{-3}s for the Al_{63}-Au case and less for the other cases. This is about 8 orders of magnitude less than that needed for the initiation of D-D fusion by self-heating of compressed material.[9]

(3) Nonlinear collision processes between moving atoms occasionally increase the energy of an individual atom to as much as twice the incident energy per atoms; however, the probability for such events decreases rapidly with increasing energy. The Al_{63}-Au and Al_{32}-Au results in Fig. 2 are reasonably well fitted with decaying exponential functions. For the first case the number of events per incident cluster is approximately $N(E)=3.12\times10^{3}\exp(-9.04E)$; for the second case it is $N(E)=8.02\times10^{2}\exp(-8.77E)$, where E is in keV. Thus, events which lead to atoms with potential energies as high as 4 keV would be very rare—approximately 1 in 10^{12} impacts.

(4) When the mass of the incident cluster atoms is much less than the mass of the target atoms, there is significant deformation and ejection of energetic material from the cluster at impact. This, together with the sputtering of target atoms, removes a significant amount of energy from the impact region, thereby making it even more difficult to maintain high transient densities for long containment times.

Our simulations highlight the importance of nonlinear effects in collision cascades induced by cluster impacts on solid surfaces. However, these purely collisional mechanisms do not appear to be sufficient to explain the high rate of fusion observed by BFF;[5] therefore, one must necessarily look carefully into other mechanisms that might be responsible for their observations.

T.A.T. acknowledges partial support from the NSF (DMR 86-15641).

[1]R. Beuhler and L. Friedman, Chem. Rev. **86**, 521 (1986).

[2]P. Sigmund, J. Phys. C **2**, 175 (1989).

[3]M. H. Shapiro, T. A. Tombrello, and D. E. Harrison, Jr., Nucl. Instrum. Methods Phys. Res., Sect. B **30**, 152 (1988).

[4]D. E. Harrison, Jr., Crit. Rev. Solid State Mater. Sci. **14**, S1 (1988).

[5]R. J. Beuhler, G. Friedlander, and L. Friedman, Phys. Rev. Lett. **63**, 1292 (1989).

[6]G. Moliére, Z. Naturforsch. A **2**, 133 (1947).

[7]P. M. Morse, Phys. Rev. **34**, 57 (1929).

[8]M. H. Shapiro and J. Fine, Nucl. Instrum Methods Phys. Res., Sect. B **44**, 43 (1989).

[9]H. Enge, *Introduction to Nuclear Physics* (Addison-Wesley, Reading, MA, 1966), p. 456.