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Section B

Molecular dynamics simulations of low-energy cluster deposition on metallic targets *

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A modified version of the multiple interaction code SPUT2 was used to simulate impacts of 63-atom Al and Au clusters on 7-layer Au targets. For 1, 5, and 10 eV/atom Al and Au clusters, 50 impacts each were calculated up to a cutoff time of 2 ps. For each case studied, we found that the final shape and penetration depth of the incoming cluster was almost independent of the initial cluster position relative to the target. The 1 and 5 eV/atom Al clusters were flattened to less than 40% of their initial thickness and exhibited registration with the substrate at 2 ps. The 10 eV/atom Al clusters formed a poorly registered monolayer on the Au surface. In these higher-energy collisions a significant number of Al atoms were reflected from the Au surface. The 1 eV/atom Au clusters were flattened to approximately 60% of their initial thickness and also exhibited clear registration with the substrate at 2 ps. Higher-energy Au clusters penetrated deeply into the targets, causing substantial damage and crater formation.

1. Introduction

Low-energy deposition of clusters may become an important new technique for producing epitaxial films at low temperature [1]. Tombrello [2] has pointed out the importance of molecular dynamics (MD) simulations to improve our understanding of this process, and to help guide future experimental work in this field. Only a few previous attempts have been made to simulate low-energy cluster deposition. Müller [3] and Biswas, Grest, and Soukoulis [4] attempted MD simulations of film formation with very low energy clusters. Hsieh and Averback [5], using an embedded-atom MD code, simulated one event each for 13-atom Cu clusters with energies of 3.54 and 25 eV/atom impacting a Cu target, and one event each for 92-atom Cu clusters with energies of 1 and 3.54 eV/atom impacting the same material. They found that the 25 eV/atom, 13-atom cluster produced a small crater, while the clusters with energies/atom less than or equal to the Cu binding energy (3.54 eV) produced epitaxial layers on the surface without creating point defects. Yamamura [6] used a Monte Carlo code to simulate the deposition of silver clusters on carbon, and found significant differences between results from linear and nonlinear versions of the program.

In order to understand more fully the processes of defect production, atomic mixing, implantation, and epitaxy during low-energy cluster bombardment, we have simulated a large number of impacts for both 63-atom Al and 63-atom Au clusters on Au(100) targets. For both species simulations were carried out at cluster energies of 1, 5, and 10 eV/atom. The first energy is well below the binding energy for both Al and Au (3.4 and 3.8 eV, respectively), the second is just above the binding energies, and the third is more than a factor of 2 above the binding energies.

2. Simulation model

A modified version of the multiple interaction code SPUT2 was used for the simulation results reported in this paper. The algorithms for improved efficiency in the modified code were the same as reported previously [7]. Semi-empirical two-body potentials consisting of Morse wells joined smoothly to Molière cores were used to describe the interactions between atoms. The parameters for the Al–Al and Au–Au potentials as well as the parameters for the Al–Au Molière core were the same as those used by Shapiro and Tombrello [8]. The geometric means of the parameters for the Al–Al and Au–Au Morse wells were used to describe the Morse well for the Al–Au potential used in this work. This Morse well was joined smoothly to the Molière core by a cubic spline extending from 1.8 to 2.2 Å.

The primary modification made to the basic SPUT2 program for this work was the inclusion of a damped restoring force of the form $-m\omega^2 r - m\gamma v$ for a single

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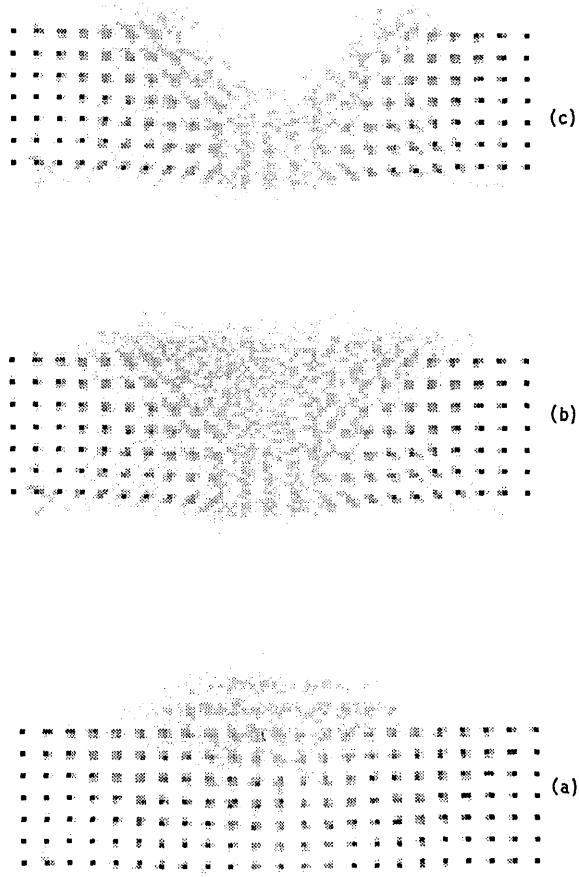

Fig. 1. (a) Cross-sectional density plot at 2 ps for fifty 5 eV/atom Au cluster impacts on Au targets; (b) cross-sectional density plot for fifty 10 eV/atom Au cluster impacts on Au targets; (c) same as (b) but only target atoms are shown. See text for more details.

layer of atoms at the sides and back of the target crystallite. The restoring force was similar to that used in ref. [5]. This force was included to simulate the flow of energy from the target crystallite to a semi-infinite surrounding medium. The restoring force on each border atom varied linearly with distance from the equilibrium point of that atom in the undisturbed (100) lattice, and was directed toward the equilibrium point. The coefficient ω was obtained from a quadratic approximation to the bottom of the Au–Au Morse well. The damping coefficient γ was chosen to be twice as strong as that needed for critical damping. Preliminary simulations showed that this choice provided the most efficient removal of energy from the target crystallite

when the collision cascade reached the borders of the cell.

Calculations were carried out for 63-atom Al and Au clusters impacting normally on Au targets with 1, 5, and 10 eV/atom. The targets were 7-layer, fcc single crystals containing 1915 atoms. They were oriented with their (1, 0, 0) faces perpendicular to the direction of incidence. The clusters were simulated by small fcc crystallite cubes. (Our choice of potentials for interactions between cluster atoms ensures that the incoming clusters are stable entities.) The primary impact zone for the incoming clusters was chosen to be a square region at the center of the target, which retained the basic symmetry of the target. This square region was divided into 100 small squares. Fifty of these squares, evenly spaced, were chosen for the individual impacts. Within each small square the exact impact coordinates were chosen randomly. In order to reduce the time needed for computation, calculations were cut off at 2 ps, which is roughly the end of the "collisional" phase of the impact [5,6].

3. Simulation results

The average cluster center-of-mass (CM) locations and changes in shape at the simulation cutoff time (2 ps) for each case simulated are given in table 1. Changes in shape have been measured by comparing the RMS deviations of the x-, y-, and z-positions of cluster atoms at the end of the simulation to those at the start of the simulation. The uncertainties quoted are the standard deviations of the various quantities.

For all cases studied there is not more than a ± 0.2 atomic layer spread in final CM locations. This indicates that the initial impact location has relatively little effect on the final penetration depth of the cluster, and that channelling is unimportant for cluster impacts at these energies. The insensitivity of our results to initial impact location also suggests that changes in the orientation of the incoming clusters would have little effect on the results. The 0.5 atomic layer CM penetration depth for the 10 eV/atom Au clusters is an indication that significant target damage and cratering occur for these impacts. In contrast, the 10 eV/atom Al clusters do not appear to cause significant cratering, but tend to compress the top layer of the target.

The results presented in table 1 also show that for all impact energies the Al clusters flatten out much more than the Au clusters. The 1 and 5 eV/atom Al clusters flatten out to little more than one atomic layer in thickness by 2 ps, and exhibit relatively good registration with the substrate (see below). Volume is not conserved as well for the 5 and 10 eV/atom Al and Au clusters as it is for the 1 eV/atom clusters. This is an indication of greater mixing of substrate and cluster atoms for these cases.

Fig. 1 shows side views of the target and Au clusters (5 and 10 eV/atom) at 2 ps, while fig. 2 shows a similar view of the target and 5 eV/atom Al clusters. These figures were obtained in each case by superposing results from 50 impacts, and taking a two atomic layer wide cross-section that included the CM of the cluster. The cross-sections were divided into squares 0.4 Å on a side. The density of atoms in each square is represented by a gray-scale, with black representing the highest density of atoms.

It is clear from the results in table 1 and from fig. 1a that the 1 and 5 eV/atom Au clusters flatten to about three layers on the surface, with at most moderate damage to the first layer of the substrate. On the other hand, the 10 eV/atom Au clusters bury themselves almost entirely in the substrate (fig. 1b), and cause significant cratering of the target material (fig. 1c). The 5 eV/atom Al clusters (fig. 2) flatten to form slightly more than a monolayer on the surface, with relatively little damage to the substrate.

Density plots of the first epitaxial layer are shown in figs. 3a and 3b for 1 eV/atom Au clusters and 5 eV/atom Al clusters, respectively. Again, 50 impacts have been superposed for each case, and the grid size is 0.4 Å on a side. For these low-energy impacts there is very good registration of the first epitaxial layer with the substrate lattice at 2 ps. The registry for both Al and Au clusters improves as the cluster energy decreases. Both the 5 and 10 eV/atom Au clusters create

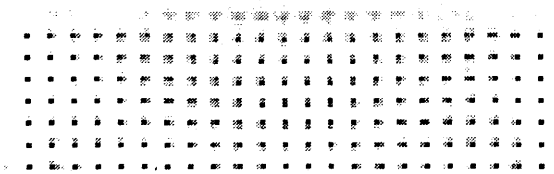

Fig. 2. Cross-sectional density plot at 2 ps for fifty 5 eV/atom Al cluster impacts on Au targets.

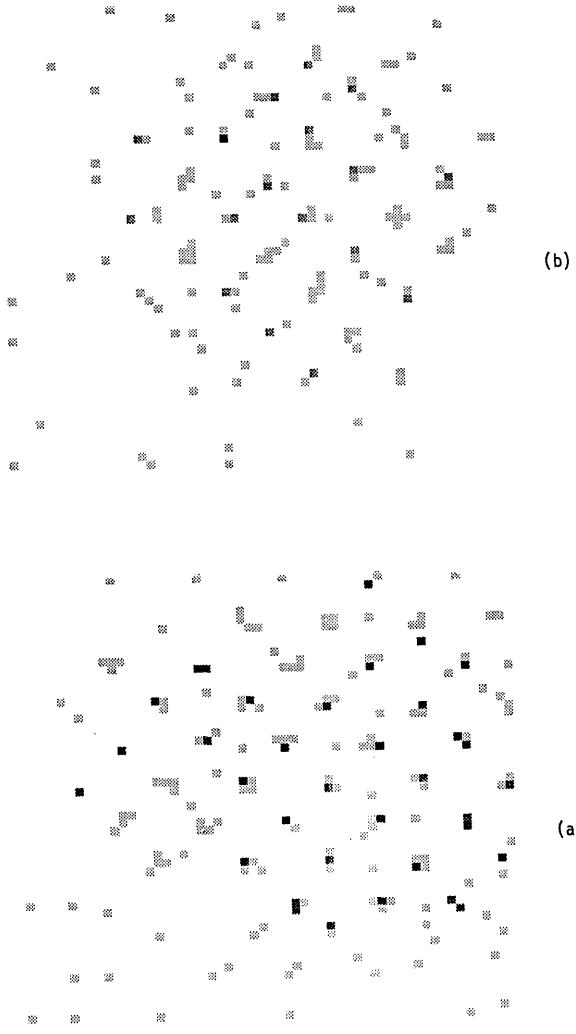

Fig. 3. Density plots of the first epitaxial layer at 2 ps for (a) fifty 1 eV/atom Au cluster impacts, and (b) fifty 5 eV/atom Al cluster impacts on Au targets. See text for more details.

Table 1
Cluster penetration depth and change in shape at 2 ps

Cluster type	Incident energy [eV/atom]	Cluster CM penetration [a] [atomic layers]	Ratio of final-to-initial rms deviations of cluster atom positions [b]		
			x	y	z
Au	1	-1.70 ± 0.10	1.38 ± 0.09	0.61 ± 0.05	1.38 ± 0.08
	5	-0.90 ± 0.17	1.41 ± 0.10	0.64 ± 0.07	1.43 ± 0.13
	10	0.50 ± 0.20	1.41 ± 0.08	0.79 ± 0.06	1.39 ± 0.08
Al	1	-1.11 ± 0.07	1.79 ± 0.15	0.38 ± 0.07	1.81 ± 0.18
	5	-0.64 ± 0.05	2.77 ± 0.26	0.25 ± 0.04	2.71 ± 0.27
	10	-0.30 ± 0.06	3.30 ± 0.24	0.30 ± 0.04	3.27 ± 0.25

[a] Zero corresponds to the first layer of the target. Negative values indicate that the cluster CM lies above the target.
[b] Clusters impact the target along the y-direction.

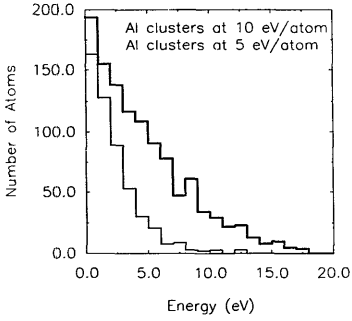

Fig. 4. Energy spectra of sputtered Al atoms following 5 eV/atom (light line) and 10 eV/atom (heavy line) Al cluster impacts on Au targets.

too much target damage and atomic mixing to permit much registration by 2 ps. The 10 eV/atom Al clusters flatten to essentially a monolayer by 2 ps, but exhibit little registration with the substrate. In constrast, both the 1 and 5 eV/atom Al clusters (fig. 3) exhibit good registration by 2 ps.

A significant fraction of the atoms in the 5 and 10 eV/atom Al clusters, 27% and 59%, were ejected upon impact with the Au target. The energy spectra for these ejected atoms (fig. 4) are quite hard, extending to more than twice the incident energy per atom. No sputtering was observed for any of the Au impacts, or for 1 eV/atom Al impacts.

4. Discussion

These simulations suggest that low-energy cluster beams should be an effective means for the deposition of very smooth epitaxial metal films. They also suggest that damage to the substrate will be minimal if the energy of individual atoms in the incoming clusters is kept below the binding energy. For 1 and 5 eV/atom Al and for 1 eV/atom Au cluster impacts on gold substrates recovery of the crystalline structure was evident by 2 ps. This is a much faster recovery than was observed by Hsieh and Averback in their Cu–Cu simulations [5], and probably is the result of the rapid and extensive spreading of the incoming Al clusters on the substrate surface. Our results also suggest that full 3D MD simulations of film formation for cluster atom energies up to 5 eV/atom are feasible.

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