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Molecular-dynamics investigation of cluster-beam deposition

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The interaction of energetic clusters of atoms with solid surfaces has been investigated by molecular-dynamics computer simulation with use of embedded-atom-method potentials for Cu. Clusters containing 13 or 92 atoms impinging on Cu(100) surfaces with energies ranging between 13 and 326 eV were investigated with respect to defect production, atomic mixing, and implantation. It is found that a 13-atom cluster with 326 eV completely embeds itself within the substrate while creating a small crater of six vacancies. A 92-atom cluster of the same energy, on the other hand, forms two epitaxial layers on the surface without creating point defects, but with some penetration. In both events, the penetration of the cluster is accommodated by plastic deformation of the substrate. By following the time evolution of these events, a clear picture cluster-solid interaction is obtained.

Recently, the problem of an energetic cluster of atoms impinging on a solid surface has become of interest as a potentially important method for growing high-quality films at low substrate temperatures[1] and possibly also for fusion reactions.[2,3] A unique feature of ionized-cluster-beam deposition (ICBD) is that a large quantity of energy can be delivered to a locality while each atom in the cluster carries only a small fraction of that energy. This differentiates ICBD from other beam-assisted methods where individually accelerated atoms contain sufficient energy to create radiation damage as well as to become implanted. Although these rudimentary ideas about cluster-beam deposition are intuitively clear, no general understanding of how cluster-solid interactions differ from atom-solid interactions has developed. Moreover, the more generic problem of energetic-particle interactions with surfaces is still not well understood, particularly in the nonlinear cascade regime where all atoms in a local region are set into motion. In this work we have used fully dynamical computer simulations to elucidate the physical mechanisms of such interactions. In addition, we have examined these events for defect production in the film, intermixing of cluster and substrate atoms, and the morphology of the cluster on the surface, quantities important for cluster-beam deposition.

The details of the molecular-dynamics (MD) code employed here are described elsewhere.[4] The substrate in the present work was a orthorhombus with dimensions $11a_0 \times 11a_0 \times 5a_0$ (a_0 is the lattice parameter). Two damped layers were employed inside fixed boundaries on five of the faces, with damping coefficients adjusted to mimic the flow of vibrational energy from the region of the computational cell to a semi-infinite surrounding medium. No constraints or damping were placed at the free surface. In this study, we have employed an embedded-atom-method (EAM) potential for Cu (Ref. 5) for both the cluster and the substrate. The lattice parameter is 3.61 Å and the cohesive energy $E_{\rm coh}$ is 3.54 eV. Although the EAM potentials are derived by fitting properties near equilibrium, that for Cu has been shown adequate for describing atomic collisions at energies as high as ≈ 25 eV.[6] The incident clusters, which contained 92 or 13 atoms, were approximately spherical but were relaxed before initiating the event; although the surface energy was, in all events, less than $\approx 10\%$ of the initial kinetic energy. Each event was followed until 99% of the kinetic energy was dissipated, approximately 15 ps. Four events are described in the present work. Events "*A*" and "*B*" represent, respectively, clusters of 92 or 13 atoms with initial kinetic energies of 326 eV (this yields 1.0 $E_{\rm coh}$/atom in the 92-atom cluster); these events are described in detail. Events "*C*" and "*D*" describe a 13-atom cluster with 1 $E_{\rm coh}$/atom initial kinetic energy and a 92-atom cluster with 1 eV/atom, respectively. As starting conditions for each event, the crystallographic axes of the cluster was aligned with those of the substrate, but its lattice sites were translated 0.5 Å in the ⟨110⟩ direction and its velocity was directed 9° from the surface normal, [001], with the transverse velocity in the ⟨110⟩ direction.

A clear picture of the interaction of an energetic cluster of atoms with a solid obtained from snapshots of the atom positions at various instants of time during an event as il-

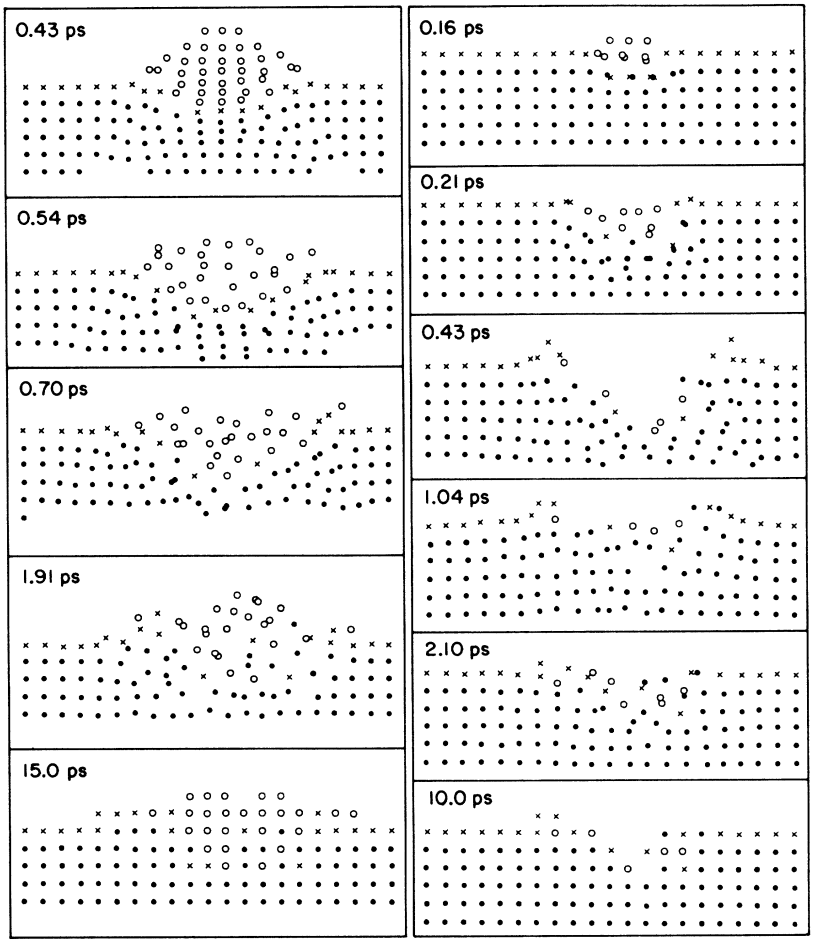

FIG. 1. Snapshots of events A (left column) and B (right column). Atoms within a cross-sectional slab of thickness a_0 (two atomic planes) normal to [100] are shown.

lustrated in Fig. 1 for events A and B. Atoms within a cross-sectional slab of thickness a_0 are shown (two atomic planes): cluster atoms as (○), original surface atoms by (×), and other substrate atoms by (●). It is observed that the 92-atom cluster initially depresses the substrate surface without penetrating it. A line of demarcation clearly separates the cluster from the substrate. The crystalline structure of the cluster is lost at ≈ 0.54 ps (≈ 0.3 ps after contacting the surface) and is not regained until much later in the event ≈ 15 ps; the substrate, on the other hand, is heavily deformed but without loss of crystalline structure or indication of melting. Maximum compression of the substrate occurs at ≈ 0.70 ps, after which the impacted region rebounds upward. During this expansion phase, when the kinetic energies are high and the densities low, some intermixing between substrate and cluster atoms occurs. It is observed that plastic deformation accommodates the penetration of cluster atoms into the substrate as original surface atoms (13 in all) flow to the first epitaxial layer at the cluster perimeter (bottom left in Fig. 1). The larger number of surface atoms flowing to the left in the figure is due to the component of cluster momentum initially in that direction. Event B is quite similar to event A; however, its smaller projection area creates much larger stress on the substrate and, consequently, greater deformation. This cluster completely embeds itself in the matrix, leaving a "crater" of vacancies above it, as shown at 0.43 ps. It is worth noting the small amount of interpenetration between cluster and substrates atoms. Like the larger cluster, substrate atoms are forced onto the surface at the periphery of the cluster, again the asymmetric flow reflects the initial cluster momentum. At 1.04 ps the substrate rebounds, but due to the limited atomic mobility on the surface, the crater above the cluster is left unfilled. These snapshots show little evidence for the phenomenon of local melting that has been shown to characterize energetic displacement cascades.[7] This we believe is a consequence of the relatively low total cluster energy so that only a small volume gains sufficient kinetic energy for melting. Consequently, the energy is dissipated rapidly into the substrate as will be discussed below.

Depth distributions of the cluster atoms for the four events are plotted in Fig. 2. A clear trend of deeper penetration with increasing energy per atom is observed, with all cluster atoms in event B (25 eV/atom) being embedded in the substrate while only 10% are embedded in

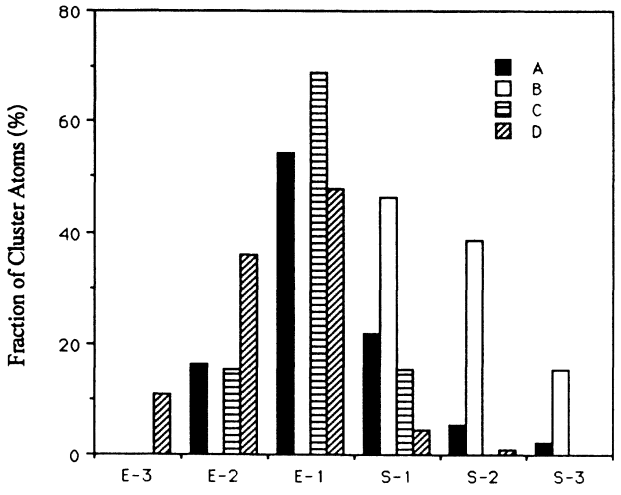

FIG. 2. Distributions of cluster atoms among different layers for events A (92-atom cluster with 3.54 eV/atom), B (13-atom cluster with 25.08 eV/atom), C (13-atom cluster with 3.54 eV/atom), and D (92-atom cluster with 1 eV/atom) at the end of the event. S-1 is the surface layer of the substrate, S-2 is the substrate layer beneath S-1, S-3 is the substrate layer beneath S-2, E-1 is the epitaxial layer on S-1, E-2 is the epitaxial layer on E-1, and E-3 is the epitaxial layer on E-2.

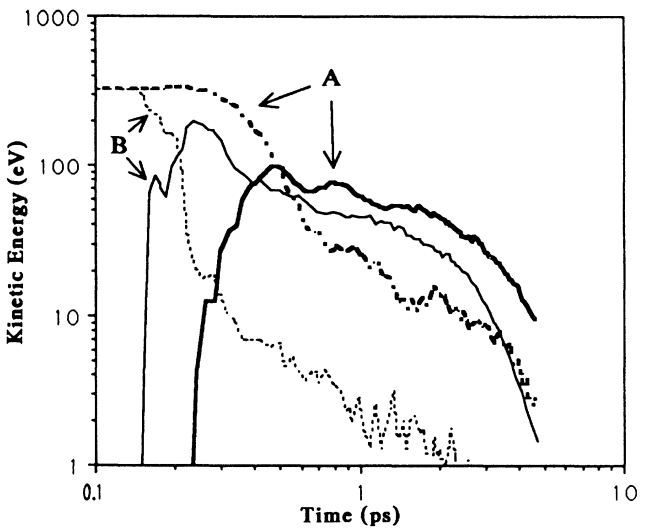

FIG. 3. Changes of kinetic energy following the time for events A and B. Dashed lines are total kinetic energy of the cluster atoms. Continuous lines are total kinetic energy of the substrate atoms.

event D (1 eV/atom). Event A (3.54 eV/atom), on the other hand, shows both a higher fraction of embedded atoms and deeper penetration than event C (also 3.54 eV/atom). This appears to be due to the greater impulsive stress from the larger cluster and hence greater deformation of the substrate, although some mixing in the thermal spike of event A, which has a greater total energy than event C, may also contribute. The morphologies of the clusters have been considered by determining the ratios of the second moments of the cluster-atom distributions in the normal (z) and transverse (ρ) directions, i.e.,

$$R=\sum_i (z_i^2/\rho_i^2)\,, \tag{1}$$

where z and ρ are measured relative to the center of mass of the cluster. The sum is over all cluster atoms. The ratios have the values 0.045, 0.078, 0.111, and 0.049 for events A, B, C, and D, respectively; a sphere has a ratio of 0.5. In all events, substantial flattening of the cluster is observed. The ratio is smallest for the larger clusters, events A and D, since their transverse deformation is unconstrained on the surface.

Plotted in Fig. 3 are the kinetic energies of the cluster and substrate atoms as a function of time. In both events the kinetic energy decreases rapidly on first impact with the surface, but then decreases much more slowly as a quasilocal equilibrium is established between cluster and substrate atoms. The kinetic energies of the cluster atoms, therefore, provide a rough estimate of the local temperature at the cluster-substrate interface. It can be seen that as the cluster and substrate reach local equilibrium, at $\sim$0.6 and 0.3 ps for events A and B, respectively (impact of the cluster on the surface occurs at 0.21 and 0.12 ps for these two events, respectively) the temperature T (assuming $\frac{3}{2}$ kT equals kinetic energy/atom, where k is Boltzmann's constant) is above the melting temperature (0.175 eV), but only for a rather brief period, <1 ps. It can be noticed in Fig. 1 that during this period, large fluctuations of local density occur. Thus, local melting does not seem to be a useful description for these events. It is interesting, however, that the initial kinetic energy in events A and B are sufficient to melt a volume containing ≈ 1000 substrate atoms, but Fig. 1 shows little evidence for melting of the substrate. Apparently, kinetic-energy dissipation is too rapid. Consequently, large amounts of interdiffusion between cluster and substrate atoms by a liquid diffusion process is not possible. At higher energies, however, this condition is likely to change as was found for energetic displacement cascades in Cu when the cascade energy was increased from a few hundred eV to a few keV.

In conclusion, we find that the interaction between an energetic cluster and a substrate is far different than that of single-particle–solid interactions since the correlated collisions of the cluster play an important role. Unlike ion implantation, the cluster penetrates the solid by forcing atomic flow of the substrate around the cluster onto the surface. This results in a ring of original substrate atoms on the surface around the cluster. For the smaller cluster, event B, the deformation also produces a crater above the embedded cluster. Only limited effects of a "thermal spike" are apparent in the simulations, which is probably due to the relatively low total energy of the clusters. Other important results are that no point defects are created in either the substrate or epilayers for any of the events other than the crater in event B and that no atoms evaporate from the surface. We also do not find any atoms breaking away from the cluster and migrating long distances over the substrate in these 0-K simulations.

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