Molecular dynamics simulation of formation of accelerated neutral atom and cluster beams

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Molecular dynamics simulation of formation of accelerated neutral atom and cluster beams

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Abstract. A molecular dynamics simulation of accelerated argon cluster ion collisions with residual gas atoms was carried out. Cluster ions having an icosahedral structure were considered. The dependence of the impact fragmentation results of cluster ions on their initial sizes and on the value of the accelerating potential was studied. The simulation demonstrated that in the case of impact breaking of cluster ions along with the forming of a “cloud” of individual atoms, the formation of heated fragments and their complete or partial melting are observed. Further intensive evaporative cooling of these fragments occurred, which caused the appearance and/or growth of an ordered (icosahedral or crystal) structure in them. The probability of charge loss of the fragments was estimated. It was demonstrated that certain of the fragments can to be a part of a mixed accelerated neutral atom and cluster beams.

1. Introduction
The present development level of various industries (for instance, microelectronics, optics, biomedicine, etc.) requires technologies for very precise treatment of material surface. These requirements are satisfied by the new challenging technology which modifies surfaces on an angstrom level via the so-called Accelerated Neutral Atom Beam (ANAB) [1-3]. ANAB is a product of collisions gas cluster ion beam (GCIB) [4] with non-ionized residual gas atoms, which are present along the beam path. The separation of ANAB from charged species remaining after the collisions is accomplished by an electrostatic deflector.

Experimental studies of some aspects of GCIB interaction with single gas atoms were performed in [1,5]. Kirkpatrick et al. [1] stated that each destroyed cluster ion turns into an aggregate of electrically neutral atoms still traveling in close proximity together. These aggregates of atoms have a necessary impact on target surfaces. However, our previous molecular dynamics (MD) simulation demonstrated that the atoms emitted from the cluster ion as a consequence of collision undergo strong scattering [6]. Thus, it can be declared that the available volume of reliable information on the features of ANAB formation, and, hence, on their composition, is not sufficient to interpret adequately the results of various material surface processing with these beams.

The work is concerned with a detailed molecular dynamics (MD) analysis of evolution of argon cluster ions after the collision with residual gas atoms. This investigation is aimed to fill the existing gap in theoretical knowledge about the ANAB nature.
Figure 1. Snapshots (sectional view) of 30 keV Ar_{923} at different time points after the collision. \( n_h \) – the total number of atoms. The number of atoms in the fcc structure (dark gray), the hcp structure (light gray), and the ih structure (black) recognized by the CNA method are also indicated.

2. Computational details

Cluster ions having an icosahedral structure were considered [7]. Their initial temperature was assumed to be equal to 35 K [8]. An evolution of cluster ions after the collision was observed for 1.5 \( \mu \)s. The dependence of the impact fragmentation results of cluster ions on their initial sizes and on the value of the accelerating potential was studied. The number of atoms in cluster ions varied from 309 to 1415. The accelerating potential value varied in the range from 10 to 100 keV.

A Lennard-Jones pair potential was used to describe the interaction of argon atoms. The simulation was carried out in the coordinate system, associated with the initial position of the cluster center. In the chosen reference system, a single atom was incident on a stationary cluster ion with a velocity. The positive direction of the \( z \)-axis was established, coinciding with the direction of the velocity vector of the single atom.

The simulation area had a form of a rectangular parallelepiped with dimensions \((L_{xy}, L_{xy}) \times (-L_z, L_z)\), where \( L_{xy} = R_c + 30 \text{ nm} \), \( L_z = R_c + 20 \text{ nm} \), \( L_z^+ = R_c + 50 \text{ nm} \), \( R_c \) is radius of the sphere, entirely containing the cluster ion. If in the simulation process atoms crossed the borders of this area, then they were excluded from further consideration.

The MD simulations were performed using the Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) code developed at Sandia National Laboratories [9].

The local atomic structure of the cluster fragments was identified using the Common Neighbor Analysis (CNA) method [10] implemented in the Open Visualization Tool (OVITO) [11].

3. Results and discussion

The simulation demonstrated that in the case of impact breaking of cluster ions along with the forming of a “cloud” of individual atoms, the formation of heated fragments and their complete or partial melting are observed (figures 1(a), 1(b) and figure 2(a)). The sizes of the fragments and their phase state are determined by the ratio of two parameters indicated above (figure 3(a)).

The significant gap between the graphs for Ar_{561} and Ar_{742} observed in figure 3(a) for large accelerating potentials is explained by the multiple effect of the initial cluster size on the collision results, all other things being equal. First of all, the collision energy increases as it decreases. In addition, the number of atoms is reduced between which this energy is redistributed. Finally, the number of atoms being able to congregate after collision into a stable large fragment is reduced. As the accelerating potential increases, the activity of these factors rises, which leads to the marked effect.

It was also found in the simulation that the processes with weak or moderate damage of cluster ions, occurring at the initial stage of the fragment formation (several dozen picoseconds), have a
oscillatory nature (figure 2(a)). The frequency of these oscillations in the considered variants was in the range of 220-280 GHz.

![Figure 2.](image)

**Figure 2.** The time evolution of 30 keV Ar$_{923}$ after the collision: (a) over the course of first 15 ps, (b) on the time interval between the 1st and 10th nanoseconds. $n_f$ – the number of atoms in the cluster fragment, $T_f$ – the temperature of the cluster fragment. The bottom figures present the number of atoms in the fcc structure (f), the hcp structure (h), and the ih structure (i) recognized by CNA method.

Further intensive evaporative cooling of the fragments occurs, which causes the appearance and/or growth of an ordered (icosahedral or crystal) structure in them (figure 2(b) and figure 1(c)). By the end of considered time interval (1.5 μs) the decrease of fragment sizes and their temperature caused by evaporation slows down significantly (figure 3(b)).

![Figure 3.](image)

**Figure 3.** The relative number of $n_f/N_c$ atoms in the cluster fragments: (a) at 100 ps after the collision depending on the initial cluster size $N_c$ and the accelerating potential $U_a$, (b) at 1.5 μs after the collision depending on the initial cluster size $N_c$. 

Figure 4. Velocity distributions of atoms scattered from 30 keV Ar$_{923}$. (a) Distribution of atoms leaving the simulation area by the absolute value of their velocity ($|v|$). (b) Distribution of atoms leaving the simulation area by the value of their velocity component perpendicular to the collision line ($v_r$). $V_c$ is the cluster velocity. The number of atoms that left the simulation area: $n$ is the total number; $n_{xy}$ is the number of atoms flying through its lateral borders; $n_{z}$ is the number of atoms flying through its lower border; $n_{+z}$ is the number of atoms flying through its upper border. $\delta E$ is the part of the collision energy taken away by the atoms that have flown out of the simulation area.

The fragments will require several tens of microseconds to overcome the distance from the zone in which GCIB interacts with the residual gas to the target (provided that they do not contain ionized atoms and therefore will not be removed by the electrostatic deflector). Consequently, the results of this work do not allow accurately describing the characteristics of the fragments as they approach the target. But the results from ref. [12] evidently suggest that after the time interval used for the simulation the changes in fragment sizes and temperature are not significant.

It seems that the distribution of sizes and energies of the fragments is determined by three factors: the mass-spectrum of the initial GCIB; the charge state distribution of cluster ions of each size represented in the mass-spectrum; distribution of fragment sizes which form from cluster ions of a certain size at a given accelerating potential.

The interaction of these factors determines the probability that some formed fragments will become part of ANAB, i.e. will become electrically neutral. The probability of charge loss of the formed fragments in the case of singly charged cluster ions can be estimated as $1 - P$, where $P = n_f/N_c$. The data in figure 3(b) show that this probability increases with the decrease in the initial size of the cluster ions and at 30 kV accelerating potential begins to exceed $\frac{1}{2}$ at $N_c$ of about 500. It can be assumed that at this accelerating potential the maximal number of large fragments (100-300 atoms) may occur in ANAB if in initial GCIB cluster ions of 400-600 atoms predominate. Thus, the energy-per-atom of these fragments will be in the range of 50-75 eV/atom.

MD simulation shown that atoms, both knocked out from the cluster ions at the collision, and evaporated from the cluster ions fragments, do not remain in close vicinity to each other, but experience rather strong scattering. Although, a significant part of them does not fly out from the narrow cone which has the apex angle of 3-5° (figure 4) that agrees well with the data in [1]. Thus, the flow of the individual atoms in ANAB has a rather low density.

4. Conclusion
The molecular dynamics simulation of accelerated argon cluster ion collisions with residual gas atoms was carried out. The evolution of cluster ions after the collision was observed for 1.5 $\mu$s. The dependence of the impact fragmentation results of cluster ions on their initial sizes and on the value of the accelerating potential was studied. The simulation demonstrated that the impact breaking of cluster ions causes, along with the forming of a “cloud” of scattered atoms, the formation of a quantity of neutral fragments of the cluster ions. These fragments become a part of ANAB which would be more correctly named as mixed Accelerated Neutral Atom and Cluster Beams.
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