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Molecular Dynamic Analysis of Collision-Induced Fragmentation of Accelerated Argon Cluster Ions

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Abstract

Aiming at investigation into generation of accelerated neutral atom beam (ANAB), we performed molecular dynamic simulation of collisions between accelerated argon cluster ions and residual gas atoms. The influence of cluster size and accelerating voltage on impact fragmentation has been analyzed. It has been demonstrated that ANAB is probably comprised of cluster fragments remained after collisions. Herewith, the atoms emitted from clusters as a consequence of collision undergo strong scattering. In addition, it has been established that in the case of single collisions and upon accelerating voltages of about 30 kV, generation of ANAB having acceptable intensity and comprised of cluster fragments with initial size of at least 100 argon atoms is impossible.

Keywords: Gas cluster ion beam, accelerated neutral atom beam, collision, molecular dynamics simulation

INTRODUCTION

An efficient method of surface nanostructuring of numerous materials based on application of gas cluster ion beams (GCIB) was developed in the 1990-s in Kyoto university [1]. However, the current development level of numerous industries (for instance, microelectronics, optics, etc.) requires for technologies for even finer, more uniform and less destructive processing of material surface. These requirements are satisfied by the recently proposed technology which modifies surfaces by means of the so called accelerated neutral atom beams (ANAB) [2, 3]. These beams are generated as a consequence of collisions of accelerated cluster ions with atoms of remaining gas and subsequent electrostatic separation of generated fragments. Experimental studies of various aspects of GCIB interaction with single gas atoms were performed in [2, 4-6]. However, the available reliable data on features of the processes upon the mentioned collisions seem to be insufficient for adequate interpretation of the obtained experimental data which can prevent successful development of technologies based on ANAB. It is possible to fill the gap in the theoretical knowledge of ANAB generation by means of computer simulation.

This work describes simulation results of collisions between cluster ions moving at high speed and stationary atoms. The simulation was based on molecular dynamics (MD) (for instance, see [7]). It should be mentioned that MD simulation played significant role in development of GCIB technology (see [8] and appropriate references).

MODEL DESCRIPTION

In a standard assembly for GCIB generation gas under pressure is injected via small nozzle into vacuum where it is adiabatically expanded. Gas is condensed with generation of clusters which can contain several thousands of atoms [1]. After skimming of major portion of non-clustered gas, clusters are ionized by electron impact and then accelerated. In the modified assembly described in [2], ANAB is generated upon collision of accelerated cluster ions with non-ionized gas atoms which exist along the beam path. Herewith, all charged particles remained after cluster fragmentation are separated from ANAB by electrostatic deflector.

Upon MD simulation illustrated in Fig. 1(a), the clusters are considered which are comprised of argon atoms. It is assumed that the clusters have face-centered cubic lattice (lattice constant H = 5.256 Å). Cluster building is carried out by filling argon atoms into sphere with the radius $R_c = C_c H$, where C_c is certain preset number. The number of atoms (N) required for this operation for the sake of brevity will be referred to as cluster size.

In order to describe interaction of argon atoms, the Lennard-Jones pair potential is used (for instance, see [7]) with the parameters ε = 10.32 meV, σ = 3.405 Å. In calculations of energy obtained upon acceleration of cluster ions, they are assumed to be singly charged (that is, at accelerating voltage U = 30 kV a cluster obtain energy equaling to 30 eV).

MD simulation of cluster collision with single atom is comprised of two stages. At preliminary stage after cluster generation, it is thermolyzed to T_0 = 10 K in 10 ps. Then collision and subsequent relaxation of the considered system are simulated. This stage is subdivided into two time intervals: duration of the first one is 20 ps with integration step of 0.1 fs, and duration of the second one is 200 ps with integration step of 1 fs. The simulation is performed in coordinates correlated by initial position of cluster mass center. In the selected reference system, the atom clusters are stationary, and the velocity obtained upon acceleration is assigned to a single atom. The simulation region is rectangular parallelepiped with the following sizes: $(-D_c, D_c)\times(-D_c, D_c)\times(-D_c, D_c+200\ \text{Å})$, where $D_c = R_c+100$ Å. Herewith, positive direction of the axis z coincides with the velocity vector of incident atom. If upon simulation atoms intersect boundaries of this region, they are excluded from subsequent consideration.

The described algorithm of MD simulation was implemented by LAMMPS software package (Large-scale Atomic/Molecular Massively Parallel Simulator) [9].

RESULTS AND DISCUSSION

Figures 1(b)-(d) illustrate the results of MD simulation obtained at various accelerating voltages for Ar_{381} cluster ion. It is seen that if at 10 kV the cluster retains major portion of its mass, then at 60 kV it is nearly completely destroyed after collision. Cluster fragments remained after collision and system relaxation are sufficiently steady formations since most atoms in them are located at the distance not exceeding $d_w = 2^{1/6}\sigma$ (see Fig. 2(a)). This distance corresponds to the bottom of potential well in the Lennard-Jones potential. Another important fact determined upon simulation was strong scattering of atoms escaping from cluster after collision. As follows from Fig. 2(a), nearly all atoms in 220 ps after collision leave the boundaries of the simulation region.

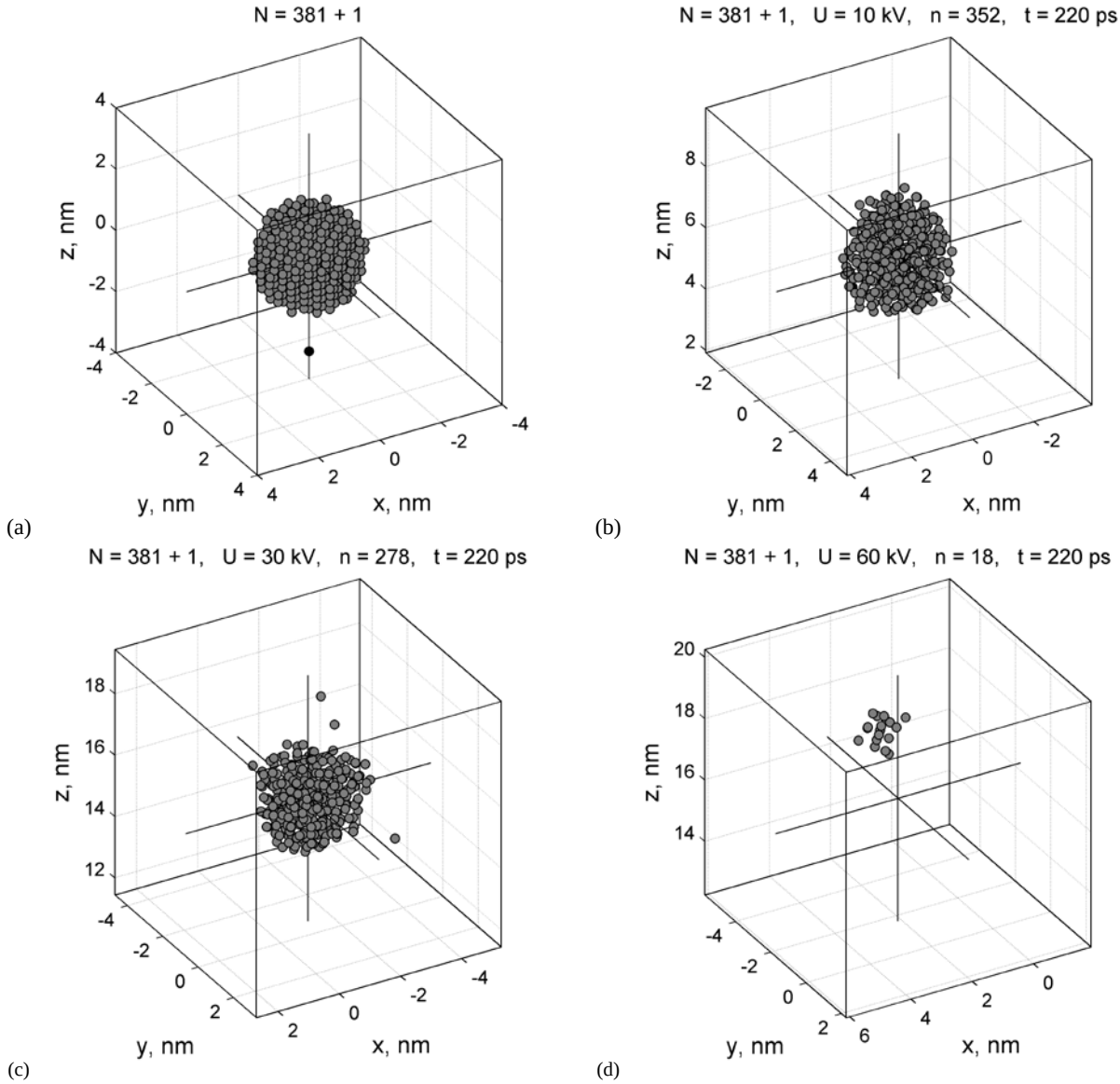


Fig. 1. (a) Initial position of single Ar atom and cluster ion comprised of 381 Ar atoms. (b)-(d) MD simulation results obtained at various accelerating voltages *U*. *n* – number of atoms in the image; *t* – time after collision. Intersection point of three lines parallel to axis corresponds to current position of mass center of the simulated system.

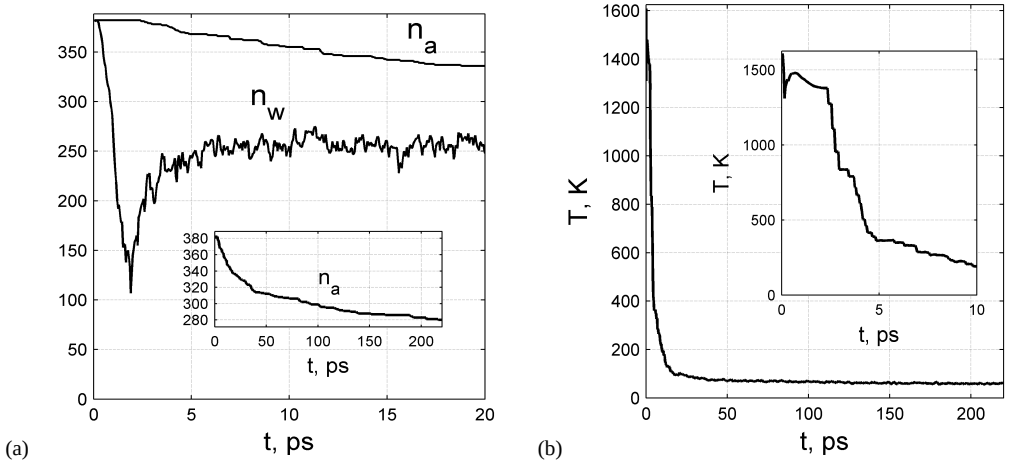
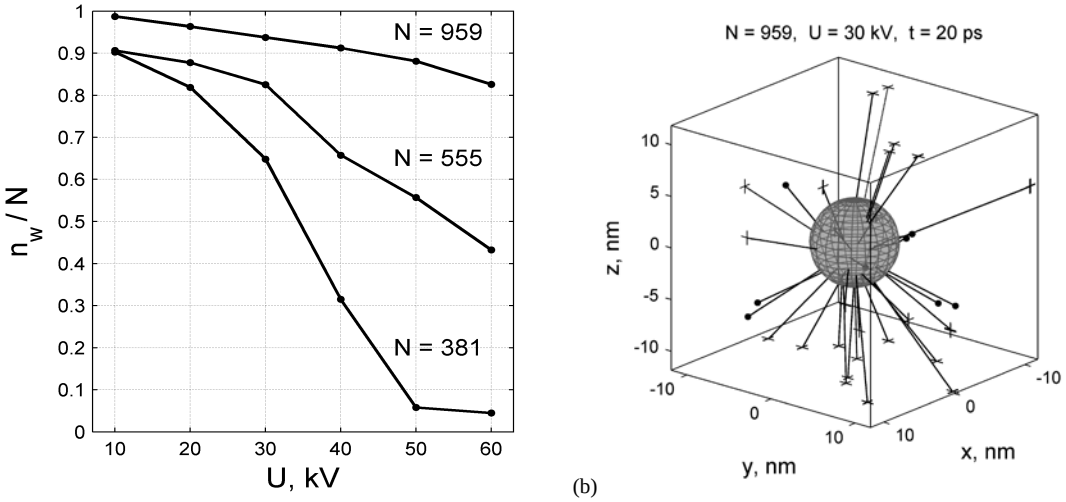


Fig. 2. MD simulation results obtained for Ar_{381} cluster ion at accelerating voltage *U* = 30 kV. (a) Evolution of the considered system after collision. n_a – total number of atoms remained in simulation region. n_w – cluster fragment size when interatomic distance is not higher than d_w. (b) Temperature of the simulated system is considered as a function of time.



(a) (b)

Fig. 3. (a) Number of atoms n_w in cluster fragments remained after collision as a function of initial cluster size N and accelerating voltage U. The values of n_w are given at the time t = 220 ps. (b) Trajectories of atoms escaped from Ar_{959} cluster ion after collision at accelerating voltage U = 30 kV. Radius of semitransparent sphere is $2R_c$.

Temperature of the simulated system as a function of time (Fig.2(b)) demonstrates that major portion of kinetic energy of incident atom is carried out by scattered atoms and only its minor portion is consumed for heating of remained fragments. Herewith, the temperature of fragments is remarkably lower than the melting and boiling points of argon (83.8 K and 87.3 K, respectively), which evidences their sufficient stability in time as a comprehensive whole.

Figure 3(a) demonstrates variation of number of atoms in steady cluster fragments remained after collision as a function of initial cluster size and accelerating voltage. It can be seen that the fraction of atoms escaping from Ar_{959} cluster ion after collision at 30 kV is lower than 10%. In addition, it follows from Fig. 3(b) that the areas, from which these atoms are emitted, are distributed highly non-uniform over cluster surface. It evidences that in this case probability of escaping of the ionized atom from the cluster is very low. Most probably, such cluster cannot participate in generation of ANAB and will be removed by electrostatic deflector.

CONCLUSION

Aiming at theoretical investigation into generation of ANAB, we performed MD simulation of collisions of accelerated argon clusters after collisions with residual gas atoms. It has been demonstrated that ANAB is probably comprised of cluster fragments remained after collisions. Herewith, the atoms emitted from clusters as a consequence of collision undergo strong scattering. In addition, it has been established that in the case of single collisions and upon accelerating voltages of about 30 kV, generation of ANAB having acceptable intensity and comprised of cluster fragments with initial size of at least 100 argon atoms is impossible.

The obtained results are radically different from the conclusions arrived at in [2]. In this model, in particular, it is assumed that ANAB is based on the atoms escaped from cluster. In addition, it is assumed that these atoms do not scatter but continue motion to target in close vicinity to each other.

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