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Influence of hydrodynamic behavior of laser ablation plume on cluster formation

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Abstract

We conducted fast-photography of the ablation plume and mass analysis of generated clusters. Time-resolved observation of the ablation plumes showed that the behavior of the plume drastically changes depending on the background helium gas pressure. Particularly when the pressure was more than 100 Pa, a strong light emission from the front edge of the plume was observed, indicating that the strong compression of the plume vapor occurs and the kinetic energy of the drifting plume may be converted into the thermal energy of the compressed vapor. We found that this plume compression process could be enhanced by introducing an ellipsoidal cavity, which is due probably to converging waves in the background gas towards the focal point of the ellipsoid. A TOF mass spectroscopy indicated that clusters having a mass corresponding to Al, AlO, Al_2, and Al_2O were generated in the laser ablation cavity and transported downstream. However, large aluminum clusters consisting of more than a few tens of atoms were not observed in the present experimental condition.

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Keywords: Laser ablation, cluster generation, TOF measurement

1. Introduction

Recently high-energy cluster ions have been attracting renewed attention as a powerful tool to generate extremely high-energy-density states in solid material since the acceleration of cluster ions to energies beyond ~100 keV/u

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turned to be realistic after the invention of an induction synchrotron by Takayama *et al*.[1]. The induction synchrotron can accelerate ion species having any specific charge (*q/m*) in principle because of its large bandwidth of revolution frequency. Based on this novel technology, a circular induction accelerator called induction microtron, which is specialized in accelerating large cluster ions having small *q/m*, was recently proposed [2], and its design work is under way at KEK.

The acceleration of cluster ions itself has been demonstrated over the last two decades. The first pioneering work was done by Dammak *et al*., where fullerene (C_{60} and C_{70}) ions were accelerated up to several tens of MeV using a tandem electrostatic accelerator [3]. The velocity of the accelerated fullerene ions was, however, limited to several tens of keV/u, so they could interact only with the surface layer of solid material. The deep interaction of cluster ions with solid material has been achieved using smaller clusters consisting of a few atoms. Tomita *et al*. experimentally showed that the mutual coupling effects among neighboring constituents of a cluster, namely vicinity effects, can affect the stopping power and the secondary electron yield of the target material [4]. Energetic large cluster ions (> ~100 keV/u) composed of more than ten atoms are considered to induce more remarkable vicinity effects in the target, but it has not been explored because almost all previous experiments for cluster acceleration were carried out by electrostatic accelerators, where the achievable kinetic energies of cluster ions were restricted by the maximum terminal voltage of the accelerator (≤ ~20 MV).

In contrast to the electrostatic linear accelerators, circular accelerators can achieve much higher beam energy owing to the principle of multiple acceleration. Thus, the induction microtron is expected to make a breakthrough in the field of cluster acceleration. On the other hand, an extremely high vacuum system must be employed for the circular cluster accelerator because beam loss due to collisions with residual gas atoms can be a serious problem. The beam loss issue also requires the ion source to supply a large amount of metal- or covalent-binding cluster ions. There has been, however, only a few works on the development of high-flux cluster ion sources.

Laser ablation of solid material is often employed to generate metal- or covalent-binding clusters. In conventional laser-ablation cluster sources, ablated vapor is rapidly cooled down in a helium gas flow, which causes the condensation of atoms into clusters. Iwata *et al*. have been developed a new type of laser-ablation cluster source called spatiotemporal confined cluster source (SCCS), which adopts an ellipsoidal cavity to confine the ablation plume and control the condensation process [5]. They reported that silicon cluster super-lattice structure was formed on a substrate [6], showing that their cluster source is excel in size uniformity. The hydrodynamic and thermodynamic behaviors of the vapor plume in the ellipsoidal cavity strongly affect the properties of generated clusters (mean size, size uniformity, and yield). The purpose of this study is to investigate the correlation between vapor plume behavior and cluster properties in the laser-ablation cluster source. This paper presents results of the analysis of cluster size distribution using a time-of-flight mass spectrometer (TOFMS) and the temporally-resolved observation of the vapor plume with fast photography.

2. Experimental setup

A schematic of the experimental apparatus is shown in Fig. 1. An aluminum disk target was set for laser ablation in a small cavity, which was filled with helium gas continuously fed from a gas cylinder. The typical pressure of the cavity ranged from 1 to 350 Pa. The target was mounted on a rotational feedthrough so that the target could be rotated manually without breaking vacuum. A frequency-doubled Nd:YAG laser (~55 mJ, 5 ns) irradiated the target and generated a dense aluminum vapor plume. The laser spot on the target had an elliptical shape (~$1.1\times0.8\pi$ mm^2) and the typical laser intensity was 4 W/cm^2. To ensure the reproducibility of laser ablation, the target surface was refreshed every ~ten thousand shots by rotating the target. Cooling of the vapor plume by surrounding helium gas promoted cluster generation, and then the clusters were extracted through a long nozzle (4 mm in diameter, 80 mm long) into vacuum (≤ ~0.1 Pa) with the help of

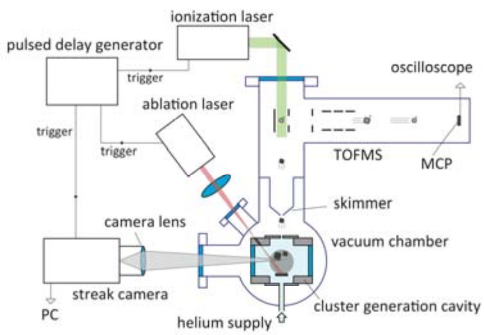

Fig. 1. Experimental setup of fast-photographing and TOFMS.