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A Model of Large Droplet Behavior in Ablation Plasma Produced by Intense Pulsed Light Ion Beam

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Spherical objects of ~100 μm in size were observed on an Al target which was irradiated by intense pulsed ion-beam evaporation. It was estimated that the spherical objects flied toward the substrate as large droplets, and the large droplets were fragmented into small droplets with < 18 μm in diameter during the flight into ablation plasma. Thin films of Al were produced by ion-beam evaporation at target-substrate distance (d_{TS}) = 40, 70 and 130 mm. The relationships between the droplets diameter, the number density of droplets on the Al thin films and d_{TS} were investigated quantitatively. In consequence, the number density of large droplets with > 54 μm in diameter decreased with increasing d_{TS}. On the other hand, the number density of droplets < 18 μm in diameter increased with increasing d_{TS}. The following model was proposed to explain the obtained results. The spherical objects flied toward a substrate, and the spherical objects were fragmented into small droplets by colliding with Al ions during the flight into ablation plasma.
Key words: ablation plasma, ion-beam evaporation, thin film, droplet, target-substrate distance

1. INTRODUCTION

When an intense pulsed light-ion beam (LIB) is irradiated onto a solid target, a high-density ablation plasma is generated. By utilizing the plasma, thin films can be deposited onto a substrate which faces the target. This method has been termed pulsed ion-beam evaporation (IBE) [1-7]. Various thin films including $B_{12+x}C_{3-x}$ [4, 5], $SrAl_2O_4$: Eu [6] and TiFe [7] were successfully prepared by IBE.

However, many droplets existed on some of thin films prepared by IBE. The droplet generation becomes a serious problem in other physical vapor deposition methods, such as laser ablation methods [8] or ion plating methods, as well as the IBE method.

Empirically, it was known that the number of large droplets on thin films decreased with increasing the target-substrate distance in IBE[9]. If the above-mentioned empirical rule is correct, the large droplets may be fragmented into small pieces or evaporated when the large droplets fly in the ablation plasma. However, the relationships between the number density of large droplets on the deposited thin film, the droplets diameter distribution and the target-substrate distance were not fully understood.

The purpose of this study is to quantitatively clarify the relation between the number density of droplets on the deposited thin films and target-substrate distance. Moreover, the other purpose of this study is to propose a droplet behavior model between the target and the substrate from experimental results.

2. EXPERIMENTAL DETAILS

The experiments were carried out using an intense, pulsed, light-ion-beam generator "ETIGO-II" [10] in Nagaoka University of Technology. Figure 1 shows the schematic of the experimental setup for thin film preparation by IBE. The right half of the figure is a diode chamber for generating an ion beam. A magnetically insulated ion diode was used for the experiment. A polyethylene sheet (flashboard) was attached on an Al anode as the ion source. A stainless-steel cathode had slits over the area which faced the anode. A high-voltage pulse with 1 MV and 70 ns from "ETIGO-II" was applied to

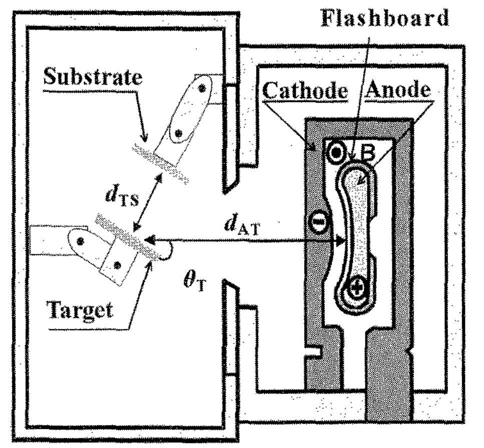

Fig. 1. Schematic of ion-beam diode and thin-film preparation chambers.

the magnetically insulated diode, and plasma was generated on the anode surface. The ions in the plasma were accelerated to cathode by high voltage, and an ion beam was generated.

The left half of the Fig. 1 is a chamber for thin film preparation. Table I summarizes the experimental conditions. An Al plate with 40x40x1 mm^3 in size and Si wafers with 20x40x0.5 mm^3 in size were used as a target and substrates, respectively. The Al target was tilted at 30 degrees to the beam axis. In this experiment, anode-target distance (d_{AT}) was 200 mm. The target-substrate distance (d_{TS}) was adjustable from 40 mm to 130 mm. The thin-film preparation chamber was evacuated to a pressure of $\sim 10^{-4}$ Torr.

For paying attention to the increase in surface roughness of the target by the increase in the number of LIB irradiation, LIB was irradiated onto the Al target until the target roughness (R_a) became about 18 μm [11]. The thin films of Al were prepared by irradiating LIB onto the above-mentioned target at d_{TS} = 40, 70 and 130 mm. The ablation plasma was observed by using a high-speed camera (ULTLA-NAC, NAC Inc.). Surface morphology of the Al target and the thin film was observed by using an optical microscope. From these micrographs, the number and the size of droplets with diameters larger than 1 μm were counted. For statistical accuracy, more than 100 droplets were observed on each thin film. Moreover, the relation between the number density of droplets with different size and d_{TS} was clarified.

3. RESULTS

Figure 2 shows the high-speed photograph of the Al ablation plasma taken at (a) 3 μs and (b) 5 μs after the LIB irradiation. From these photograph, Al ablation plasma had divergence angle about 3 degrees when the plasma was flying toward the substrate. Thus, geometrically the plasma density should decrease by about 28 % with the increasing in d_{TS} from 40 to 130 mm. In addition, from Fig. 2 (b), it was observed that the plasma plume was directly reached the substrate at least until d_{TS} = 100 mm.

Table I Experimental conditions.

Accelerating voltage (peak)	1 MV
Pulse width (FWHM)	70 ns
Energy density of ion beam	30 J/cm^2
Ion species	H^+ (75%)
Number of shots	1～10
Pressure	2×10^{-4} Torr
Targets	Al
Angle of target (θ_T)	30 deg.
Substarate	Si (100)
Substarate temperature	R.T.
d_{AT}	200 mm
d_{TS}	40,70,100,130mm

Figure 3 shows the optical micrograph of the Al target surface after LIB irradiations. From this optical micrograph, there were some spherical objects with diameter of about 100 μm on the Al target [12].

Figure 4 shows the optical micrographs of the Al thin films (a) and (b) prepared at d_{TS} = 40 mm, (c) prepared at d_{TS} = 70 mm, (d) prepared at d_{TS} = 130 mm.

From Fig. 4 (a) and (b), on an Al thin film prepared at d_{TS} = 40 mm, many large droplets with diameters larger than 50 μm were observed. Most of the droplets were discoid, however, at the center of Fig. 4 (b), a spherical droplet was observed. The diameter of the spherical droplet was 93 μm. From Fig. 4 (c), on an Al thin film set at d_{TS} = 70 mm, some large droplets with diameters larger than 10 μm were observed. From Fig. 4 (d), on an Al thin film prepared at d_{TS} = 130 mm, the number of large droplets with diameters beyond 10 μm decreased. However, many small droplets with diameter of ~10 μm were observed.

The number density of the large droplets was measured using the above-mentioned optical micrographs, and the droplets with diameters larger than 1 μm were counted.

Figure 5 shows the number density of the droplets as a function of d_{TS}. It is found that the number density of the droplets increases with increasing d_{TS}.

Figure 6 shows the droplet size histogram. For all d_{TS}, the number density of droplets with diameters 6 to 12 μm was the largest.

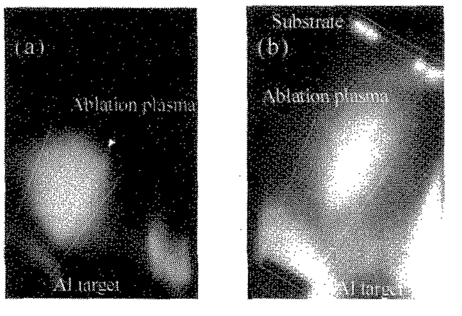

Fig. 2. High-speed photograph of Al ablation plasma formed by ion beam irradiation at the substrate delay time of (a) 3 μs and (b) 5 μs was located at d_{TS} = 100 mm.

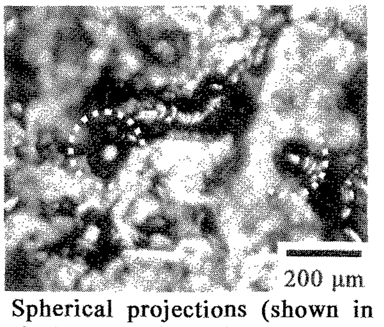

Fig. 3. Spherical projections (shown in white dotted circle) on the Al target after LIB irradiations.

For clarification of d_{TS} dependence of the droplet size, the number density of droplets with diameter smaller than 6 μm (a), between 6 and 18 μm (b), larger than 18 μm (c) and larger than 54 μm (d) plotted against d_{TS} in Fig. 7. From Figs. 7 (a) and (b), the number density of droplets with diameters smaller than 6 μm and 6-18 μm increased with increasing d_{TS}. On the other hand, from Figs. 6 (c) and (d), the number density of droplets with diameters larger than 18 μm decreased with increasing d_{TS}. It is thought that the droplet collection efficiency varies depending on the droplet's state such as solid or liquid. Moreover, the smaller the droplet size is, the easier the solidification is. However, from the experimental results, many small droplets existed on the Al thin film prepared at d_{TS} = 130 mm. Therefore, it is considered that the droplets collecting method in this research is appropriate.

4. DISCUSSION

Experimental results with the present study can be summarized as follows.

(1) Many spherical objects with the diameter of ~100 μm were observed on the Al target after LIB-irradiation.

(2) A spherical droplet with diameter 93 μm was observed on the Al thin film prepared at d_{TS} = 40 mm.

(3) The number of large droplets with diameters larger than 18 μm decreased with increasing d_{TS}. The large droplets were not observed on the Al thin film prepared at d_{TS} = 130 mm.

(4) The number density of droplets with diameters smaller than 18 μm increased with increasing d_{TS}.

From the results (1) and (2), the diameters of spherical objects on the Al target almost corresponded to the diameter of spherical droplets on the Al thin film prepared at d_{TS} = 40 mm. Accordingly, it is thought that the spherical objects are the origin of spherical droplets on the thin films.

From the results (3) and (4), the number of large droplets with diameters larger than 18 μm decreased with increasing d_{TS}. On the other hand, the number density of droplets with diameters smaller than 18 μm increased with increasing d_{TS}. In addition, the plasma plume was formed between the target and substrate.

From these results, it is considered that the large droplets are fragmented into small droplets during the flight into ablation plasma by colliding with Al ions.

Another hypothesis to decrease the number density of large droplets by increasing d_{TS} is divergence of large droplets in the plasma. It was

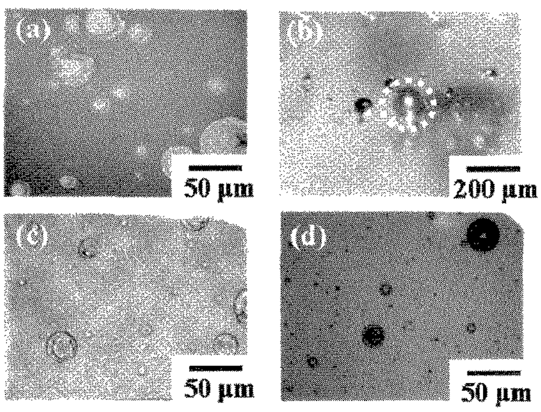

Fig. 4. Optical micrographs of the surface of Al thin films deposited at d_{TS} = 40 (a) and (b), d_{TS} = 70 (c) and d_{TS} = 130 mm (d).

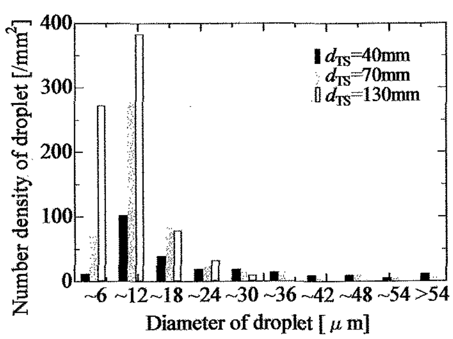

Fig. 6. Size histogram of droplets on the Al thin films.

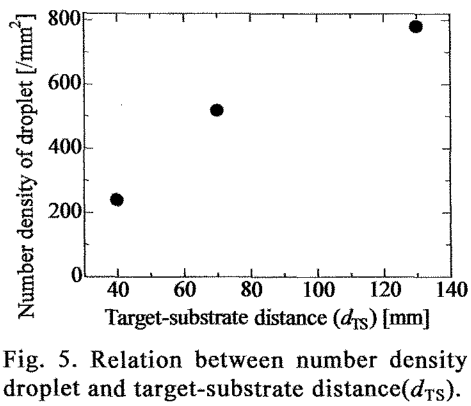

Fig. 5. Relation between number density of droplet and target-substrate distance(d_{TS}).

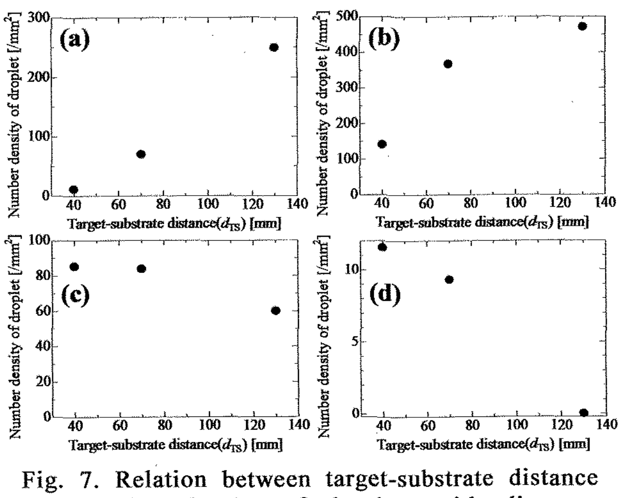

Fig. 7. Relation between target-substrate distance and number density of droplets with diameter smaller than 6 μm (a), between 6 and 18 μm (b), larger than 18 μm (c) and larger than 54 μm (d).

assumed that the divergence angle of droplets is almost equal to the divergence angle of ablation plasma. As mentioned before, the decrease in the plasma density by the increase in d_{TS} from 40 to 130 mm was about 28 %, which should be identical to the decrease in number density of droplets. On the other hand, the measured decrease in large droplets were 100 % by the decrease in d_{TS} from 40 to 130 mm (Fig. 7 (d)). Consequently, it is considered that the cause of the decrease in number density of large droplets is not due to the divergence of the droplets.

In Fig. 7 (a), the number density of small droplets increased with the increase in d_{TS}. This phenomenon can be explained if we assume formation of the small droplets by aggregation of the plasma. From this assumption, the aggregation must take place at/below its boiling point. In Fig. 2 (b), the thermal equilibrium plasma plume caused by IBE [3] reached at the substrate at d_{TS} = 100 mm, which might indicate the high-temperature plasma still existed on the substrate. However, the number density of droplets increased until d_{TS} = 100 mm, though the plasma plume directly reached the substrate. Therefore, formation of the droplets by the aggregation of the plasma is not likely.

From the above-mentioned results, the large droplet behavior between target and substrate is explained. The proposed model is schematically drawn in Fig. 8. When ion-beam irradiates onto a solid target, the spherical objects are formed on the target surface. The spherical objects depart from the target, and become large droplets. The large droplet is generated during ablation plasma generation. The large droplets are fragmented into small droplets. As a result, the number of large droplets decreases with increasing flight distance. In addition, the thin films with smooth surface can be fabricated with increasing d_{TS}[9].

5. CONCLUSIONS

By characterizing an LIB-irradiated Al target and deposited Al thin films, the relationships between the number density of droplets, the size distribution of droplets and target-substrate distance were quantitatively investigated. From these results, the following conclusions were obtained.

1) The spherical objects with diameter of ~100 μm were observed on the Al target after LIB-irradiations. On the thin film prepared at d_{TS} = 40 mm, a spherical droplet with diameter of 93 μm was observed. From these results, the spherical objects were formed by irradiating LIB, and was considered to be the origin of the droplets.
2) The number density of large droplets with diameters larger than 54 μm decreased with increasing d_{TS}. On the other hand, the number density of droplets with diameters smaller than 18 μm increased with increasing d_{TS}. It was considered that the large droplets were fragmented into small droplets by colliding with Al ions during the flight into ablation plasma.

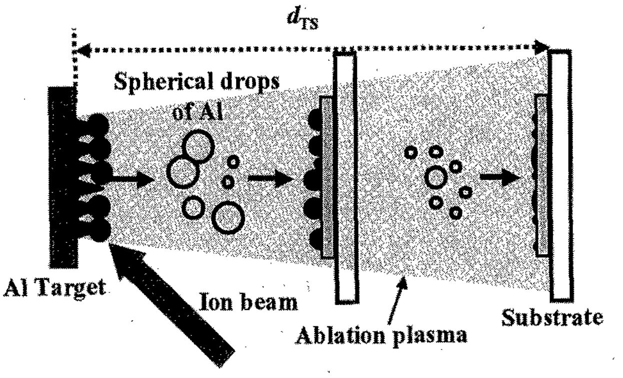

Fig. 8. Schematic of large-droplet generation and behavior model

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