Dynamics of bubbles induced by laser ablation in liquid nitrogen

液体窒素中でのレーザーアブレーションによって誘起された気泡の ダイナミクス

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We examined the dynamics of bubbles induced by laser ablation in liquid nitrogen. The bubble dynamics in liquid nitrogen at the boiling point did not show the shrinkage and the collapse, and in other words, the bubble induced in liquid nitrogen at the boiling point was not a cavitation bubble. On the other hand, when we used liquid nitrogen at a lower temperature than the boiling point, we observed the expansion, the shrinkage, the collapse, and the reformation of a second bubble. This bubble dynamics was similar to the dynamics of a cavitation bubble induced by laser ablation in water at room temperature.

## 1. Introduction

In recent years, many studies on liquid-phase laser ablation have been reported, which attracts much attention as a new method for synthesizing functional nanoparticles. Almost all the works employ water or alcohol based liquids as the medium of laser ablation, but using liquid nitrogen is another possibility, especially for synthesizing nitride nanoparticles. In several years ago, we tried to synthesize TiN nanoparticles by laser ablation in liquid nitrogen [1].

On the other hand, we have reported that a cavitation bubble, which is induced in front of the target by the laser irradiation, plays an essential role in the synthesis of nanoparticles in liquid-phase laser ablation [2]. In this work, we examined the behavior of cavitation bubbles induced by laser ablation in liquid nitrogen. In general, the bubble dynamics is significantly affected by the liquid temperature. An originality of this work is to use liquid nitrogen which has a lower temperature than the boiling point.

## 2. Experiment

Figure 1 shows the experimental apparatus. In order to realize laser ablation in liquid nitrogen at a lower temperature than the boiling point, we constructed a cylindrically symmetric chamber which consisted of four shells. The shells were named the vacuum chamber, the saturation chamber, the pressurized chamber, and a shell for collecting nanoparticles, from the outside.

The vacuum chamber was evacuated using a turbomolecular pump for the thermal isolation. The saturation chamber was filled with liquid nitrogen.



Fig. 1 Experimental apparatus for laser ablation in liquid nitrogen at an arbitrary temperature between the freezing and boiling points.

The saturation chamber was evacuated using an oil rotary pump, so that liquid nitrogen in the saturation chamber was at the boiling point at the reduced pressure. This temperature was lower than the boiling point of liquid nitrogen at an atmospheric pressure. The pressurized chamber and the collection shell were also filled with liquid nitrogen, which was cooled down by low-temperature liquid nitrogen in the saturation chamber. The pressurized chamber and the collection shell were able to be pressurized up to two atmospheres. In this way, we realized liquid nitrogen at an arbitrary temperature between the freezing and boiling points.



Fig. 2 Pictures of cavitation bubbles observed in liquid nitrogen at temperatures of 77 and 66 K. The pressure was 0.1 MPa.

We employed the second harmonics of a Nd:YAG laser for ablation. A titanium target installed in the collection shell was irradiated by the YAG laser beam from the normal direction via four quartz windows and a focusing lens which was placed in the saturation chamber. The cavitation bubble induced in front of the target was visualized by shadowgraph imaging. The light source in the shadowgraph imaging was a white lamp, and we used a high-speed camera for capturing the transmitted lamp light when we needed a movie of the cavitation bubble. When we needed a finer picture of the cavitation bubble, the high-speed camera was replaced with an ICCD camera.

## 3. Results and discussion

Figure 2 shows shadowgraph images of bubbles induced in liquid nitrogen at temperatures of 77 and 66 K. The pictures at a temperature were picked up from a high-speed movie (200 kf/s), and the times indicated below the pictures show the delay time from the irradiation of the YAG laser pulse. The pressure in the pressurized chamber was 0.1 MPa, and the fluence of the laser pulse on the target surface was estimated to be approximately 15 J/cm<sup>2</sup>. We observed the formation and the expansion of a bubble at both the temperatures. At a temperature of 77 K, which was the boiling temperature of liquid nitrogen at 0.1 MPa, the expansion of the bubble was not followed by the shrinkage. We observed the change in the bubble shape from a hemisphere to distorted one, and after that, we observed that the bubble was shredded into small bubbles at delay times longer than 1 ms. In contrast, we observed the shrinkage of the cavitation bubble at a temperature



Fig.3 Temporal variation of the bubble size observed at different temperatures and pressures.

of 66 K. The shrinkage was followed by the collapse and the reformation of the secondary bubble. This behavior is more or less similar to the behavior of a cavitation bubble induced by laser ablation in water at room temperature. It is considered that a liquid temperature lower than the boiling point is necessary to induce a cavitation bubble.

Figure 3 shows the temporal variation of the bubble size. The definition of the bubble size here is the distance between the target surface and the top (the leading edge) of the bubble. As shown in the figure, the shrinkage, the collapse, and the reformation of the bubble were observed at 66 K in the ambient pressure of 0.1 MPa. When the pressure of the pressurized chamber and the collection shell was 0.2 MPa, the maximum bubble size became smaller and the collapse of the cavitation bubble became earlier. In addition, we observed the formation of the third cavitation bubble at 0.2 MPa and 66 K, which may be due to the fact that the boiling point at 0.2 MPa is higher than that at 0.1 MPa. It is suggested that the temperature in comparison with the boiling point is essentially important for the dynamics of a cavitation bubble.

## References

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