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Influence of changing frequency of superposed ultrasonic wave on reaction fields induced by laser ablation in water
重畳超音波の周波数変化が及ぼす水中レーザーアブレーション反応場への影響

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1. Introduction

Recently, liquid-phase laser ablation attracts much attention of many researchers as a novel effective technique for synthesizing crystalline nanoparticles.[1–3] Since this technique requires no vacuum equipment, the replacement of a target and the collection of nanoparticles are easier in comparison with gas-phase laser ablation. By irradiating an intense laser pulse onto a solid-state target immersed in liquid, a dense plasma with a high temperature and a high pressure is induced at the vicinity of the target because particles ejected from the target are confined tightly by ambient liquid. After the disappearance of the ablation plasma, a cavitation bubble which has the dynamics of expansion, shrinkage, and collapse is formed at the ablation point on the target surface. It is well known that the cavitation bubble at the collapse also has a high pressure and a high temperature.[4] This special state may play an important role as the reaction field for the synthesis of nanoparticles.[5] For controlling the dynamics of the reaction fields, we have proposed adding ultrasonic sound pressure to ambient liquid.[6] To date, the influence of ultrasonic wave on chemical reactions in water has been investigated in the field of sonochemistry.[7] It is well known that acoustic cavitation in water induces the formation of active hydrogen and hydroxy radicals, and enhances chemical reactions.[8–10] Therefore, we expect that the characteristics of nanoparticles syhthesized in liquid-phase laser ablation is controlled by ultrasonic wave. In our fundamental examinations, we have found that the irradiation of a laser pulse at the negative phase of the sound pressure results in an ablation plasma with a stronger optical emission. In addition, the repetitive formations and collapses of cavitation bubbles have been driven by the ultrasonic wave.[6] In the above works, we used an ultrasonic trasducer with a frequency of 32 kHz. In this paper, we examined the influence of the frequency of the ultrasonic wave on the ablation plasma and the cavitation bubble.

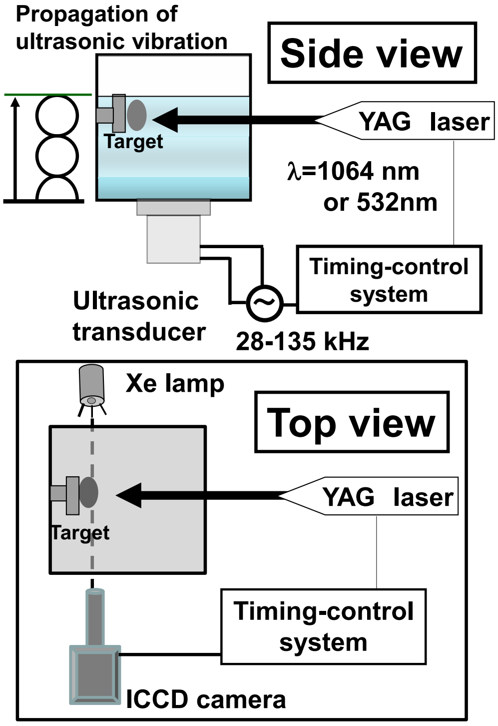

Fig. 1 Schematic drawing of experimental setup.

2. Experimental

The experimental apparatus is schematically shown in Fig. 1. A rectangular container was filled with distilled water. Ti or Au target was immersed in water, and was irradiated by YAG laser pulses at a wavelength of 1064 or 532 nm from the normal direction. The YAG laser beam was focused using a lens, and the range of the laser energy was 80-360 μJ/pulse. The container had an excitation system of ultrasonic wave at the bottom. The ultrasonic wave propagated toward the top, and was reflected back at the boundary between water and air. The depth of water was adjusted to obtain a standing-wave structure of the ultrasonic wave. The irradiation position of the YAG laser beam corresponded to the maximum amplitude of the standing ultrasonic wave. The frequencies of the ultrasonic wave were 28, 32, 81 and 135 kHz. A low ultrasonic power of 30 W was used. At this power, we never observed bubbles induced by the ultrasonic wave. The YAG laser was operated in the single-shot mode, and the

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phase of the ultrasonic wave at the YAG laser irradiation was controlled using a digital delay system. The dynamics of the cavitation bubble was observed by shadowgraph imaging using a flash lamp and a charge-coupled device camera with a gated image intensifier (ICCD camera). The same ICCD camera was also used for observing the optical emission image of the plasma.

3. Results and discussion

Figure 2(a) shows the temporal variation of the radius of the cavitation bubble when the laser pulse irradiated the target at the zero phase of an ultrasonic wave at 135 kHz, while Fig. 2(b) shows the same result when the frequency and the irradiation phase were 32 kHz and 300 degrees, respectively. For comparison, the bubble radii observed without ultrasonic wave are also plotted. In addition, the waveforms of the sound pressure at the irradiation point are illustrated in the figure. In the case of 135 kHz, as shown in Fig. 2(a), the temporal variation of the bubble radius was almost not affected by the sound pressure of the ultrasonic wave. Formations and collapses of cavitation bubbles were observed for some times, and the bubble radius decreased with time. A similar result was observed when the laser energy was 360 μJ. This result may be attributed that the expansion and the shrinkage of the bubble may not be able to follow the fast change in the ultrasonic pressure. On the other hand, we observed a remarkable change in the dynamics of the cavitation bubble when the frequency of the ultrasonic wave was 32 kHz, as shown in Fig. 2(b). We observed the repetitive formation and collapse of the cavitation bubble in the case with the ultrasonic wave at 32 kHz. This means the transition from laser-induced bubbles to ultrasonic-driven ones, since the repetition frequency of the formation and the collapse was almost the same as the frequency of the ultrasonic wave. The repetition may induce additional etching of the target and more significant crystallization of nanoparticles. Regarding the ablation plasma, the enhancement of the optical emission intensity was observed by the irradiation of the laser pulse at the negative phase of the sound of pressure at all the ultrasonic frequencies between 28 and 135 kHz. The enhancement of the optical emission intensity was more significant at a lower ultrasonic frequency.

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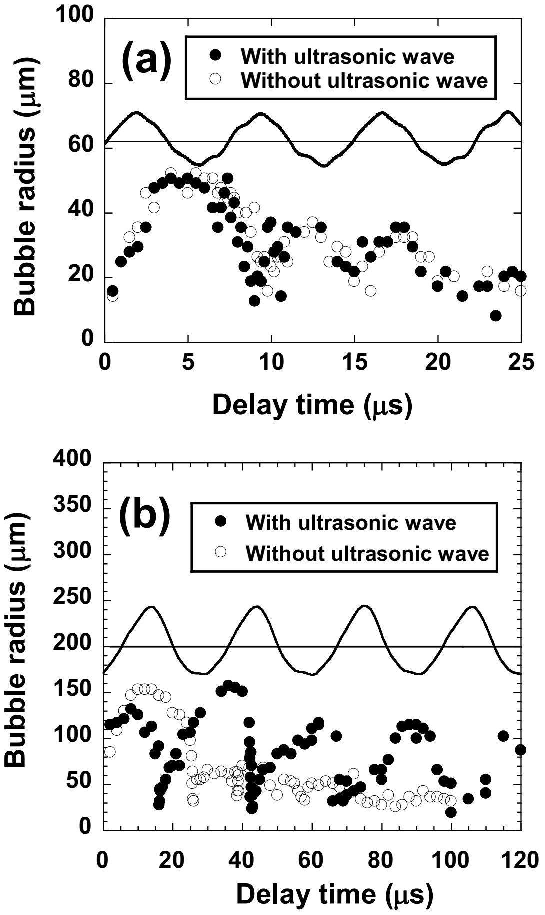

Fig. 2 Temporal variations of the bubble radii after laser ablation in the cases with and without the ultrasonic wave. The laser energy was 80 μJ, and fluence was about (a) 1 and (b) 1.5 J/cm^2. The YAG laser pulse irradiated the target at phases of (a) 0 and (b) 300 degrees.

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