New Method of Liquid Atomization Based on Pulse Laser Ablation of Liquid Surface

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・Abstract

We propose a new method of liquid atomization by pulse laser ablation. When a pulse laser is incident from the water onto the air, the liquid jet occurred on the water surface and atomization proceeds at the tip of liquid jet. It is shown that the produced liquid jet has a very complicated structure, and the duration of the phenomenon is about several tens of μs. In order to observe the liquid jet process, the high-resolution and high-speed imaging of the phenomenon is necessary. In this study, we succeeded in imaging the details of the phenomenon by introducing the combination of a pulse laser shadowgraphy with a high-resolution film.

In this study, we observed liquid jet formation process and investigated the dependence of parameters such as laser energy, beam pattern, and laser fluence (J/cm^2) at the liquid surface. The liquid jet is formed by driving liquid surface by ablation-induced plasma plume. Many slender liquid ligaments are seen extending from water surface like milk crown. Only a part of the phenomena characteristics can find some resemblance to crowning phenomena, and others not. In the liquid jet at an early delay time from the ablation, atomization is seen at the tip of the ligaments. It is also shown that air shock wave precedes the ligaments. Jet tips are moving at supersonic velocity but decelerated very rapidly. After several tens of μs from laser ablation, the jet direction at the water surface gradually approaches to almost vertical to the surface and the jet collapses afterward. In addition, observation showed that the behavior of the liquid jet and atomization at the tip depends on liquid material.

1.Introduction

Studies of fuel spray and atomization processes have been extensively made for the purpose of gasoline engine developments. In such studies, major atomization method is apparently by using nozzles, and few other methods have been pursued.

We are proposing an entirely new way of atomization by adopting pulse laser ablation of liquid-air interface. The principle of the present method is based on the enhancement of laser energy density on the liquid surface, which was proved to be the case by laser reflection at the material interface when the incident laser beam is irradiated from the material of higher refractive index to that of lower one [(1)-(3)]. Key idea of this method lies in our previous study of the enhancement of laser energy absorption by intentionally roughened surface of transparent materials [(4)-(7)]. In that case, laser fluence of ablation threshold does depend on the direction of laser irradiation, i.e., ablation threshold is found to be much lower for laser irradiation through the transparent medium than for that directly focusing the beam on their surface. These behaviors of laser ablation of materials have quite different characteristics extensively compiled so far[(8)-(10)].

Based on these previous studies, we have adopted various liquids and air as the combination of materials with different refractive index instead of using solid transparent materials. As described later, we have found the similar effect for liquid-air combination. In our case, not only ablation but also liquid jetting and atomization have been observed. In the present case, however, the areas of application of the present method may not contain the use for automobile engines. One of the possible areas of application is to use it to design a new propellant system such as micro-satellite for space applications[(11)-(13)]. This phenomenon can also be used to medical applications for micro-surgery and/or drug delivery[(14)].

In this paper, we limit our aim of our research effort to obtain precise information on the fundamental characteristics of laser ablation of the liquid surface. The phenomena have been observed by pulse laser shadowgraphy with varying laser parameters as well as liquid materials in order to understand the main features of the proposed method.

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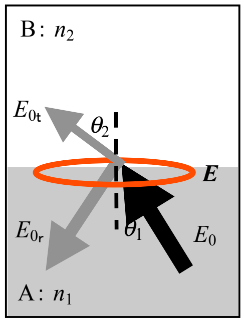
2.Physical principle of the present method and experimental method

Importance of the difference in refractive indices by pulse laser interaction with transparent media was first pointed out by Greenaway et al as an analysis of damage threshold of optical fibers at input and output surface[8]. They explained the higher damage probability of output surface by Snell's law with difference in refractive indices [8],[9]. Figure 1 shows a schematic illustration of the physical principles of the present method. This figure qualitatively explains the reduction of laser ablation threshold at liquid-air interface. When the pulse laser beam is incident from the material with higher refractive index onto the interface of the material with lower refractive index, the reflected beam to the material of laser incidence and the transmitted beam occur as shown in Fig. 1.



E_0 : Incident beam electric field
E_{0t} : Transmitted beam electric field
E_{0r} : Reflected beam electric field
n_1 : Refractive index of material A
n_2 : Refractive index of material B
($n_1 > n_2$)
θ_1 : Incident angle
θ_2 : Transmitted angle

Figure 1. Schematic illustration of the physical principle of field intensity increase.

Where the polarization of the incident beam is assumed to be perpendicular to the scattering plane, the electric field of the incident and reflected beam can be given by the simple constructive superposition of them. Electric field intensity at the interface is, then, given by the following Fresnel formula as shown in Eq. (1).

$$E = E_0 + E_{0r} = E_0 + \left(\frac{n_1 \cos\theta_1 - n_2 \cos\theta_2}{n_1 \cos\theta_1 + n_2 \cos\theta_2} \right) E_0 = \left(\frac{2n_1 \cos\theta_1}{n_1 \cos\theta_1 + n_2 \cos\theta_2} \right) E_0 \qquad (1)$$

For the case of total internal reflection, the electric field intensity at the interface doubles the incident value due to Eq. (1). Electromagnetic density at the surface may also increase due to the increase in electric field intensity. One may be careful about this explanation in the sense that Snell's law cannot explain the ablation phenomena, since it does contain neither absorption of laser energy by materials nor the nonlinearity of strong pulse laser beam. Snell's law can be used for the qualitative description of the phenomena.

We have tried to demonstrate the situation described by the principle experimentally in the case of two fluid combination of liquid and air. Figure 2 shows one of typical pulse laser shadowgraph optics to observe the phenomena. We have used an Nd:YAG laser of 10 ns duration with SHG crystal as a light source of the pulse laser shadowgraphy. In order to record the phenomenon with high resolution, we have used the Minicopy Film from FUJIFILM Co., which has very high spatial resolution of 850 pixels/mm and has the sensitivity of ISO 32. We also have two kinds of Nd:YAG lasers of 10 ns duration for the energy source of pulse laser ablation at the liquid surface. They have Gaussian or top-hat beam profile.

We start the experiment for water-air combination. Schematic illustration of experimental assembly we have used in the present experiment is shown in Fig. 3(a). In this assembly, pulse laser beam is focused through the water vessel onto the water-air interface. In the ablation phenomena, the energy density per unit area, the laser fluence is one of the most important parameter to determine the phenomena. In this experiment, the laser fluence (J/cm^2) at the water-air interface is adjusted to the value which cannot cause breakdown induced cavitation bubbles. Since incident angle to the interface is set to the value of critical angle of total internal reflection of 49 degrees, laser beam irradiated from the water region is reflected at the water-air interface. Superposition of incident and reflected light causes to increase the energy density at the water surface, and this eventually leads to water ablation. At the beam waist of the reflected laser beam, breakdown occurs and sometimes several cavitation bubbles are produced. It is noteworthy that the picture shown in Fig. 3(b) contains emissions from two cavitation bubbles and also a stronger emission from the ablation at the interface. This is the evidence of the pulse laser ablation at the interface due to laser reflection at the interface. The picture of Fig. 3(c) was taken as a shadowgraph with the aperture shown in Fig. 2 and contains various wave fronts in water as well as an air shock wave driven by the produced ablation plasma plume. Estimated laser fluence which leads to ablation is found to be 12-16 J/cm^2 calculated by using the laser energy and the focused cross section at the water surface. While in the case of laser irradiation from air onto water, similar ablation like behavior can be realized by much higher laser fluence of 190 J/cm^2. In this case, the phenomena can be interpreted as the ablation at the interface or the air breakdown. This observation is an example of ablation threshold reduction by the laser irradiation from the material with high refractive index.

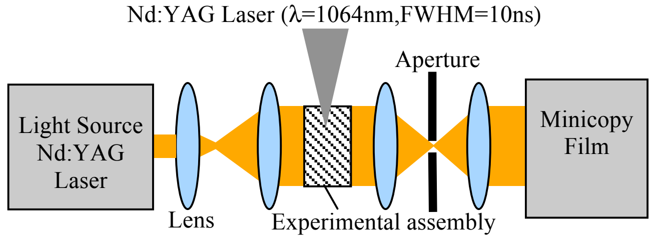


Figure 2. Schematic of optical set up for laser shadowgraphy.

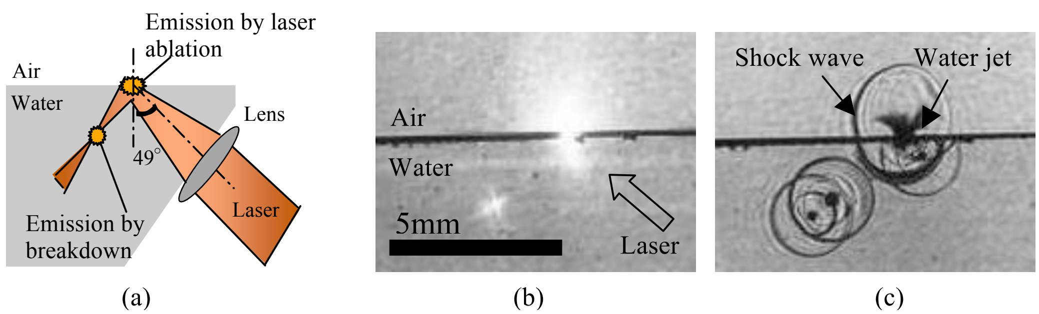


Figure 3. Reproduction of typical pulse laser shadowgraph pictures. (a) Schematic of experimental assembly, (b) laser shadowgraph without the aperture in front of the objective lens, and (c) the same with aperture to eliminate the emission from the event. Delay time of both records is 0.6 μs. Laser energy is 37mJ and laser fluence is 24J/cm^2.

These results show that the phenomena depend on the laser irradiation direction of which material has higher refractive index.
We have planned to study the dynamics of laser ablation of water surface, and also those of other liquid surface. As shown later, the phenomena followed by the surface ablation is quite complicated and have detailed structures, it is inevitable to observe the phenomena by the optical method for instantaneous recording with very high spatial resolution.

3. Results and Discussion

A series of experiments on the water surface ablation has been performed by varying the laser energy. Several other liquid samples such as ethanol have been used to study the dependence of material properties on the phenomena. As shown later, we adopted different sample assembly for water and ethanol.

3.1 Water ablation experiments

Figure 4 shows the reproduction of laser ablation shadowgraphs with relatively large laser energy (a) and with relatively small laser energy (b). In the experiments (a), we used an Nd:YAG laser whose output laser pattern is nearly Gaussian beam profile. On the other hand, in the experiment (b), the laser we used has the beam pattern something like donut shaped. We can see the influences of output laser pattern between Fig. 4(a) and (b) as a liquid jet profile.

As seen in Fig. 4(a), liquid jet produced by the laser ablation of water surface is found to consist of plenty of slender liquid ligaments. Laser energy of these shots are 300mJ and the laser fluence is 20J/cm^2 at the water surface. Observation with further delay time reveals that ligaments extend and atomization takes place at the tips of the ligaments. It is shown that the form of the liquid jet itself changes with time. Ligaments expanding like a fan shown in

Fig. 4(a) are produced by the high pressure and high temperature environments induced instantaneously by laser ablation of liquid surface layer. High speed liquid jets are found to be produced by ablation whose tip velocity is estimated to be about 1 km/s. This means that air shock wave and a pressure wave in water both have the nature of a blast wave. Shadowgraphs with later time show that ligament tends to emanate normal to the water surface. Fig. 4(b) shows a laser shadowgraph in smaller laser energy experiments (Laser energy : 62 mJ, laser fluence : 25 J/cm^2)compared with those of Fig. 4(a) (Laser energy : 300 mJ, laser fluence : 25 J/cm^2). In order to realize an appropriately large fluence at the water surface with smaller laser energy, focused area cross section must be adjusted to a very small value to ensure ablation but not very close to beam waist. This is not an easy task, and furthermore, the phenomenon itself occurs in relatively small scale. This is also not an easy object for close photography. Whole processes of jet formation and ligaments collapse are seen common and no laser energy dependence.

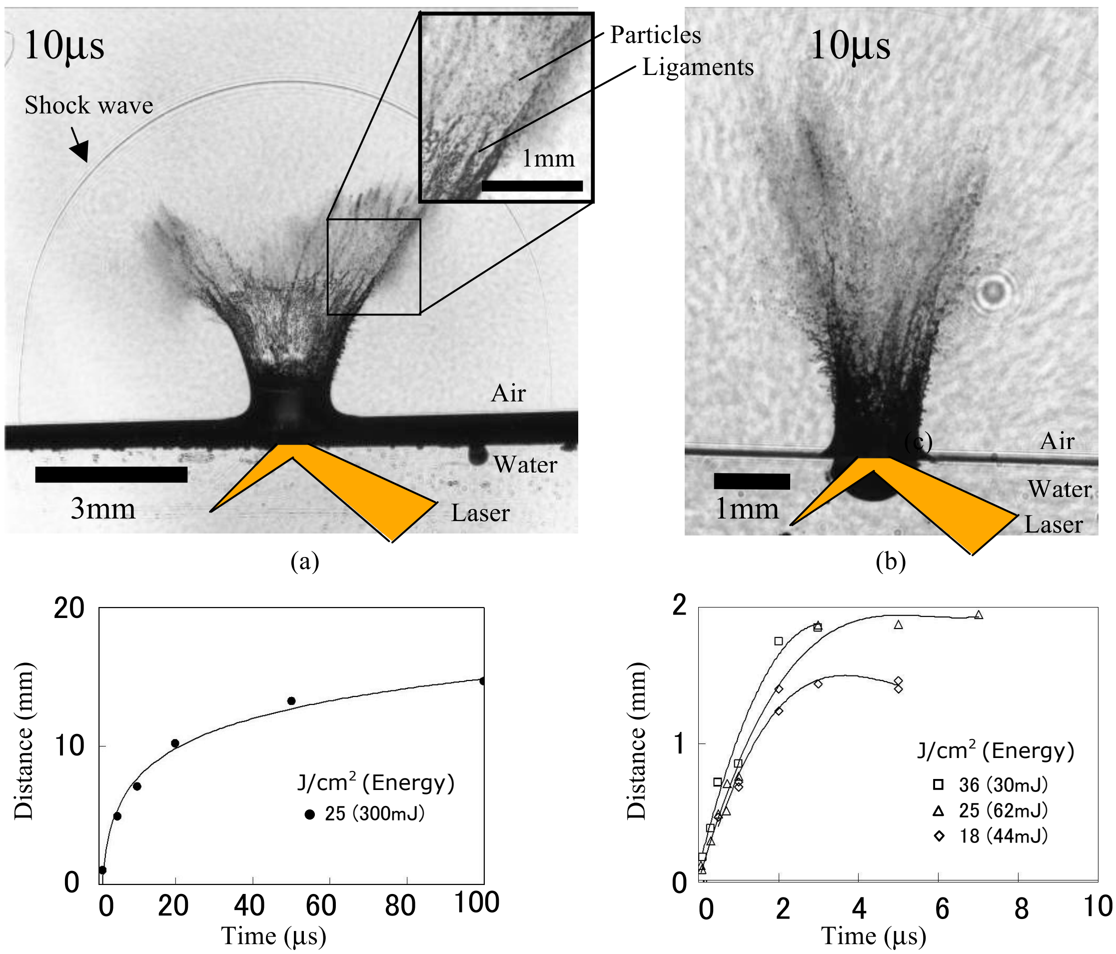


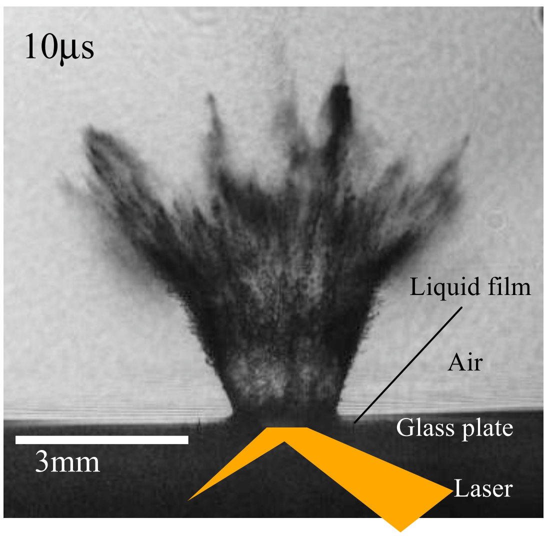
Figure 4. Laser shadowgraph reproduction of the laser ablation of water surface and relationship of jet tip distance from water surface against delay-time from laser ablation. Delay time is shown in each picture, respectively.

Group of ligaments gradually tend to collapse and after several hundred μs, they collapse to join to the central axis of the surface normal. Many ligaments and atomization is seen especially in the early stage of the phenomena.

Relationship of delay-time from laser ablation and jet tip distance from water surface is shown Fig.4(c),(d). As seen in both plots, accelerated liquid jet is decelerated very drastically. Compared with these two plots, jet deceleration is faster in case of small laser energy. This shows that the time scale of the ligament behavior and atomization at the early stage of the process clearly depends on the input laser energy.

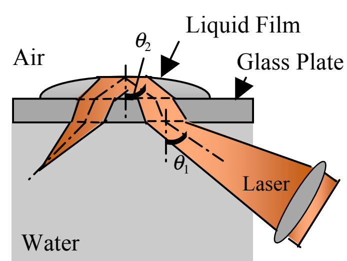
Some of the features in the every stage of the liquid jet formation, ligaments extension, atomization at the tip of them, and ligament collapse have some resemblance to the phenomena of milk crown by liquid drop impact on the shallow liquid layer. It is emphasized here, however, that the present phenomena cannot be explained as the simple crowning phenomena by the liquid drop impact.

Detailed inspection of the shadowgraphs taken in this study, we can find many droplets in focus in various areas of the pictures. Most of the droplets in the pictures are, however, an inline hologram from which we cannot deduce diameter information due to the insufficient image quality and inappropriate far-field number. At this moment, we can estimate diameter of liquid particles at the tip of ligament almost 10 – 20 μm from images in sharp focus. Since x-t plots in Fig. 4 suggests very rapid atomization at the ligament tip, we need to observe droplets more precisely by either taking clear inline hologram or other particle diagnostics.



(a)

n_{air} : Refractive index of Air
n_{water} : Refractive index of Water
n_{sample} : Refractive index of Sample
θ_1 : Incident angle (water-glass)
θ_2 : Transmitted angle (air-sample)
θ_{cr} : Critical angle (air-sample)



(b)

Figure 5. Laser shadowgraph reproduction of the laser ablation of ethanol surface. (a) Delay time of the record is shown in the photograph. Laser energy is 300 mJ and laser fluence is 20J/cm^2. (b) Schematic of experimental assembly. By the use of the Fresnell's formula to predict the necessary condition of total internal reflection of the laser beam at the liquid sample-air interface, i.e., $\theta_2 > \theta_{cr}$, one must choose the incident angle θ_1 to be lager than the critical angle of water-air interface, such that $\theta_1 > \sin^{-1}(n_{air}/n_{water})$.

3.2 Ethanol ablation experiments

Experiments using ethanol was conducted to investigate the effects of material properties on the present phenomena. In this experiment, a different sample assembly shown in Fig. 5(b) is adopted. Assembly consists of water vessel, a glass plate on water surface and a sample liquid drop on the glass plate. Sample liquid used is a drop of about 50 mg and is placed on a glass plate to be a liquid film. A vessel used for the water experiments cannot be a convenient choice for ethanol, since the sample has a relatively high vapor pressure, and the liquid surface changes with time by constant evaporation. Due to the importance of the value of laser fluence for the experimental condition, liquid surface height must be fixed to ensure keeping the focused position of the laser beam. These are the reason why we have to use a different geometry of assembly from that of water experiment. Figure 5(a) show typical laser shadowgraphs of liquid jet produced by the ablation of ethanol liquid film. As for the control of laser irradiation direction, we found a very convenient relationship between critical angle and incident angle. Please refer to the figure caption in Fig. 5(b).

Ethanol jet does not look like clear slender ligaments and seems to atomize faster than in the case of water and looks like clouds. Faster atomization process of ethanol is attributed to the relatively higher vapor pressure of ethanol. It is not clear only from this picture whether the cloud like jet consists of the atomized particles or of the vapor during the expansion of jet in Fig. 5(a). Ligament disintegration and atomization, anyhow, depend strongly on the material properties of liquids.

Summary

This study is motivated by our previous works which showed that when the irradiation of pulse laser beam from the material of higher refractive index to that of lower one, laser energy density increases at the reflected surface area which can be used to atomize liquids by using the liquid materials as the material of higher refractive index. In this process, the most important material property is optical property and not the mechanical properties like compressibility and/or viscosity. Phenomena produced by ablation is very detailed in nature, including plasma plume production, evaporation, ligaments extension, and atomization.

Summary of the experimental results is as follows:

(1) Only in the case of laser irradiation from the material of higher refractive index to that of lower one, laser ablation takes place at relatively low fluence. Ablation threshold in reverse laser incidence is much higher.

(2) Liquid jet is formed by extension of plasma plume produced by laser ablation on the liquid surface. In the early stage of the process, plenty of ligaments are produced and this phenomenon is similar to milk crown. Ligament extension is observed in the time interval of several to several tens of μs, and the tips of ligaments atomize very rapidly. Velocity of the ligament and also of the atomized particles decreases very fast probably due to air resistance.

(3) Comparison of water and ethanol experiments elucidates the difference of phenomena by the difference in liquid properties.

It is necessary to study further to clarify the transition process from ligament to atomized particles, influences for jet shape, particle size, atomization process and so on by varying various experimental conditions for various liquid samples. In order to understand the particle size and spatial distribution of the atomized particles, an inline holographic observation will be planned.

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