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Spatial confinement effects of bubbles produced by laser ablation in liquids

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ABSTRACT
The paper deals with the influence of the spatial confinement on the evolution of laser bubbles and shock waves by means of the shadowgraphy technique. Due to the constraint of walls, the bubble center shrinks faster than the bubble edge and collapses before the edge of the bubble, splitting into left and right small bubbles that continue to shrink down. The result validates that the Bjerknes force has little effect on bubble evolution at the expansion stage but a great influence on it at the collapse stage. We study the evolution of laser bubbles with different Al-plate intervals for displaying a gradual transition from constrained conditions to unconstrained conditions. In addition, we describe the dynamics of the first bubble at the expansion stage using the Rayleigh-Plesset equation. The pressure and temperature inside laser bubbles are calculated in the meanwhile.

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I. INTRODUCTION
Bubble dynamics has found important applications in many fields, such as shipbuilding engineering,1–3 mechanical engineering,4–6 ocean engineering,7–9 chemical engineering,10–12 nanomaterial engineering,13–15 nuclear engineering,16–18 medical science,19–21 and so on. In the field of shipbuilding engineering, a shock wave generated by a charge explosion under water will first cause several damages to surrounding ship structures.22 As for marine prospecting, high pressure air gun bubbles are widely used in the petroleum exploration, geological survey, and environment evaluation.23 In the field of nuclear engineering, the presence of vapor bubbles in nuclear reactors affects the fuel burnup of the nuclear reactor core as well as the steady state and transient response and the inception of flow instability of many industrial plants using boiling heat transfer.24 In the field of medical diagnosis, microbubbles are most commonly used in clinical diagnostic ultrasound imaging because they can be injected into the blood stream as a contrast agent for ultrasound imaging.25 The bubble also plays an important role in the nanoparticle’s formation when the material is ablated by pulsed laser in liquids because the ablated materials present in the bubble interact with each other in a process of nucleation and growth before dispersing in liquid.26–28

The abovementioned bubble dynamics and motion under water are multiphase problems29 involving very complex physical processes, such as heat and mass transfer effects, phase changes, hydrodynamic effects, and bubble evolution deformation.30 These complex phenomena and evolution processes bring great challenges to theoretical analysis, numerical simulation, and experimental research of bubble dynamics.31 For theoretical analysis, most research studies on bubble dynamics are based on the Rayleigh-Plesset equation which is very suitable to describe the motion of cavitation bubbles in an infinite fluid. Liang et al. found that the solutions of the RP equation fit well with the experimental data of bubbles generated by a deep underwater explosion.32 Soliman et al. modified the conventional Rayleigh-Plesset theory to solve the problem that the theoretical model did not agree with the experimental results on the temporal variations of the sizes of cavitation bubbles produced by laser ablation in water.33 After that, Lam et al. further gave an insight into the chemical composition and the thermodynamic properties within the bubble by using the Rayleigh-Plesset equation, quantitatively investigating the
mass fraction of the ablated matter with respect to the solvent vapor.34

Depending on the mechanism of formation, bubbles can usually be divided into the following categories: underwater explosion bubbles,32,35 high pressure air gun bubbles,36 spark bubbles,37 laser bubbles,38,39 rising bubbles,40 and so on. Among them, the laser bubble is an important branch in the field of bubble dynamics research, which belongs to a high pressure pulsating bubble, and its theoretical study is similar to that of the underwater explosion bubble and air gun bubble. Although the evolution and motion of bubbles in different environments and boundary conditions have been studied extensively by many researchers,41–44 only a few reports can be found for laser bubbles under a constrained condition. Under the effects of spatial constraints in bulk water, the dynamic evolution process of shock waves, bubbles, and plasma becomes more complicated. Therefore, it is necessary to study the evolution of space-constrained bubble under water, which provides a reference for the dynamics of bubbles close to a wall.

In this paper, we report on a study of the effect of spatial confinement on laser bubble dynamics, which was carried out using a pair of aluminum plate walls. First, we discuss the evolution characteristics of a laser bubble during its expansion and contraction time in a space confinement condition. Next, we explored the influences of the interval of the two walls on the evolution of bubbles and shock waves which were investigated by shadowgraphy images. Finally, we describe the dynamics of the first bubble at the expansion stage using the Rayleigh-Plesset equation. The pressure and temperature inside the bubble are calculated in the meanwhile.

II. EXPERIMENTAL SETUP

The schematic diagram of the experimental setup is shown in Fig. 1. The laser ablation process is performed using a Q-switched Nd:YAG pulse laser source (SGR-60, <3 J, Beamtech Optronics Co., Ltd.) with 1064 nm wavelength and 8 ns full width at half maximum (FWHM). A plano-convex (L1) with a focal length of 32 mm in air is placed in a glass container filled with pure de-ionized water. The focal length of this plano-convex in water will be approximately four times of that in air. The laser passes through the glass wall of the container and then is focused on the surface of the aluminum target that is fixed on a sample holder under water, as shown in the inset of Fig. 1. A pair of parallel aluminum plate walls (W) is used to confine the shock waves and laser bubbles. The sample holder with the target is connected to a three-axis stage, and we focus the laser beam in the middle of the two walls by adjusting its displacement in the z direction. Time-resolved shadowgraphy is performed to track the shock waves and bubbles. The shadowgraphy is imaged to a CCD camera (Canon, 700D) coupled with a probe laser beam generated by a picosecond Nd: YAG laser (PL2251C-10-SH-FH-PreT, EKSPLA) with a wavelength of 532 nm and a FWHM of 30 ps. Also, a “4-f” optical imaging system (L2-L3) is used for imaging the probe beams to the CCDs with the focal length of 250 mm.

The timing of the Nd:YAG laser and probe laser is controlled by a digital delay pulse generator (DG535, Stanford Research Systems). By changing the delay, we can observe the evolution of shock waves, reflected waves, and laser bubbles through the time-resolved shadowgraphy. In addition, a high-speed visible photon detector...
(DET10A, Thorlabs) is used to monitor the action of laser and record the time stamp. It is found that the transmission attenuation of 1064 nm laser in water is serious due to the strong absorption of water. In this experiment, the energy of laser reaching the surface of the aluminum target is about 10 mJ with the volume attenuation coefficient of water of about 14.4 m$^{-1}$.

**III. RESULT AND DISCUSSION**

**A. Evolution of bubbles in spatial confinement**

The detailed process after ablation laser irradiation is shown in Fig. 2. The images in Fig. 2(a) are captured in no space confinement condition. First, we can see that the shock wave caused by the laser-induced plasma emerges, and then it expands as a hemisphere. There are several lines parallel to the target surface and numerous circles that are the reflection waves of the plasma shockwave from the target. Also, the small circles are due to laser pulse ablation of nanoparticles in pure de-ionized water with a few nanoparticles which are produced when we focus repeatedly on the target to take images at different delays. From the figure, we can see that the bubble starts to appear at the moment of 1 μs, and it expands in a hemispherical shape until 210 μs when the bubble reaches the maximum radius. After that, since the pressure inside the bubble starts to be smaller than the external pressure, the bubble begins to shrink down. At the end of bubble shrinkage shown by the image at 410 μs, the bubble collapses and generates a shock wave, releasing energy outward. At the time of 430 μs, we can observe the second cavitation bubble generated, which then begins to expand but not as a standard hemisphere shape.

The images in Fig. 2(b) are in a constrained condition with an interval distance of 5 mm of the parallel Al-plate walls. We can observe that the early shock waves are reflected by two Al-plate walls and then propagate beyond space restricted range as the images shown at 20 μs. The shock waves and reflected waves propagate and reflect in confined narrow space, which causes the water between the walls to oscillate violently. There are obvious differences between the process of the first bubble oscillation in a constrained condition with an interval distance of 5 mm and that in no space confinement condition. On the one hand, the first bubble appears at the time of

![Fig. 2](image-url)
about 3 μs, which is significantly later than the unconstrained contrast experiment at the time of 1 μs. On the other hand, the collapse time of the first bubble is also later than that in the case of unconstrained conditions, which is 465 μs in the former and 410 μs in the latter.

Moreover, as shown in Fig. 2(b), although the evolution behavior of the first period bubbles in the expansion process is similar to that in an unconstrained condition, the shrinkage process is completely different from that in an unconstrained condition. In the process of the shrinkage of the first bubble, due to the constraint of the walls, the bubble front falls rapidly and the bubble begins to become flat from a hemisphere. Eventually, the center of the bubble collapses before the edge of the bubble, splitting into left and right small bubbles that continued to shrink down. Based on the subsequent repeated experiments, we observe a weak shock wave generated and propagated outward when the center of the bubble touches the target surface in this process, which is shown by Fig. 2(b) at 459 μs. After the collapse of the left and right bubbles, two new shock waves are generated with the lower left corner and the lower right corner as the center of the circle at which time energy is released, respectively, as shown in images at 465 μs. Due to the stability and repeatability problems of laser ablation in water, the two small bubbles separated by different repeated pulses had different volumes, different collapse time, and different shock waves. After the first bubble completely collapsed, the second bubble begins to form from the lower left corner and the lower right corner and tends to converge in the middle as shown in images at 520 μs.

B. Evolution of bubbles with different intervals

To show a gradual transition from constrained conditions to unconstrained conditions, we study the evolution of bubbles with different interval distances at 3 mm and 8 mm, shown in Fig. 3. The dashed lines represent the position of the target, as shown in Fig. 3(a). Similar to the case with an interval distance of 5 mm, the first bubble still shows a hemispherical expansion in the expansion stage with the distance of 3 mm and 8 mm. As can be seen, the longer the interval distance, the shorter the first bubble collapse time (520 μs of 3 mm-interval, 458 μs of 5 mm-interval, and 415 μs of 8 mm-interval). As the interval distance of a constrained condition decreases, the tendency of collapse in the center of the bubble before the edge of the bubble is more obvious. Also, it is hard to find the splitting of bubbles when the interval distance is greater than 8 mm, which behaves more like an unconstrained case.

Based on the experimental results of previous shadow images, we measure the shockwave front positions at each moment under different constrained conditions for three times and calculated their mean and standard deviation. We can plot the average propagation distance of the shockwave front as a function of time in an unconstrained condition and constrained conditions, as shown in Fig. 4. Colored shapes are the experimental results shown in the shadowgraphy images, and the line of the corresponding color is the fitting result. From the slope of the black fitted curve corresponding to an unconstrained condition, the velocity of the shockwave in the case can be estimated to be about 1536 m/s. This result is comparable to the velocity of the shockwave under constrained conditions. The velocity of the shockwave with the interval distance at 3 mm, 5 mm, and 8 mm is calculated at 1543 m/s, 1544 m/s, and 1528 m/s, respectively. The velocities of these shockwaves are not significantly different from those under unconstrained conditions, and they are all roughly equivalent to the propagation velocity of sound in water within the allowable range of error. So, there are no differences regarding the shockwave velocity between the different constrained conditions and geometrical configurations.

Figure 5 displays the ratio of center height to width of the first and second bubbles as a function of delay. Here, the bubble center height refers to the distance between the boundary surface at the bubble center position and the target surface and bubble width refers to the distance between the leftmost boundary and the rightmost boundary of the whole bubble, as shown in Fig. 2(b). The reason why the ratio of center height to width of 3 mm-interval (the red line) is significantly higher than other cases is that the bubble had hit two Al-plate walls in the expansion process under such a small constrained interval, causing the top of the bubble to rise higher. The ratio of center height to width of the bubble remains around 0.5 with Al-plate interval at 5 mm (the blue line) and 8 mm (the orange line) before the delay of 240 μs, which means the bubbles appear to have

![FIG. 3. Evolution of bubbles in laser ablation in liquids by time-resolved shadowgraphy in a constrained condition with the Al-plate interval at (a) 3 mm and (b) 8 mm. The dashed lines represents the position of the target.](image-url)
hemispherical expansion early in evolution. After 240 μs, the ratio of center height to width of bubbles decreases rapidly under three constrained conditions. This further confirms that the first bubble is affected by strong spatial constraints during the shrinkage process. The ratio of center height to width of the bubble with 5 mm-interval and 8 mm-interval has a period of time when it drops to zero, which indicated that the middle of the first bubble first touched the target surface and caused the bubble to split. We cannot see the orange line to intersect the axes for the reason that the first bubble collapses similar to the process of bubble collapse under unconstrained conditions. In addition, the shape of the second bubble with the interval distance at 8 mm is more regular than 5 mm and 3 mm. All these results can be verified from the above time-resolved shadowgraphy images.

We speculated that this special phenomenon is related to the effect of the Bjerknes force on bubble motion characteristics. When pulsating objects in a fluid oscillate in the same phase, they attract each other, and when they oscillate in the opposite direction, they repel each other. Later, this interaction between pulsating objects in a fluid is called the Bjerknes force. This force is also reflected in the effect of the rigid wall surface on the bubble. It can be assumed that the fluid around the bubble is incompressible in its pulsation. When there are other boundaries around the bubble, such as structural surface or free surface, the Bjerknes effect at the boundary causes the fluid around the bubble to move asymmetrically. This may explain why the first bubble exhibits completely different evolutionary characteristics under a constrained condition. The space confinement has little effect on the first bubble in its expansion process because the edge of the bubble is away from the walls in this stage. However, when the bubble oscillates near the structure surface, it will be slightly repelled by the structure surface at the expansion stage and strongly attracted by the structure surface at the collapse stage. This is like the two Al-plate walls creating an attraction to the edge of the bubble, preventing the edge from shrinking toward the middle. As a result, the interaction between the two walls on the bubble edge leads to the bubble center shrinking faster than the bubble edge during the bubble contraction process, causing the formation of left and right small bubbles. The greater the distance between the two walls, the greater the effect of the Bjerknes force on the bubbles. In order to further explore the influence of fluid mechanics on bubble evolution dynamics under a constrained condition, more research is needed to elucidate this phenomenon.

C. Dynamics of laser-induced bubble

Although the evolution of bubbles was affected by spatial confinement, it satisfied by the following hypothesis that (i) inertial forces drive the bubble dynamics, (ii) vapor evolution is adiabatic, (iii) the bubble is mainly composed of evaporated solvent, and (iv) there is no significant mass exchange during the first bubble oscillation. Based on the above analysis, we found that the first bubble in the expansion stage presented hemispherical expansion, similar to the unconstrained conditions. This indicates that it is less affected by the spatially constrained effect. So, we tried to use the Raleigh-Plesset equation to describe the bubble dynamics in the expansion stage of the first period.

In fluid mechanics, the Rayleigh-Plesset equation is an ordinary differential equation that describes analytically the bubble dynamics. It can be expressed by the following formula:

\[
RR + \frac{3}{2}R^2 + \frac{1}{\rho} \left[ P_B(t) - P_l - \frac{2\sigma}{R} - \frac{4\eta R}{R} \right].
\]

where \( R, P_B, P_l, \sigma, \rho, \) and \( \eta \) denote the bubble radius, the internal pressure, the surrounding liquid pressure, the fluid surface tension, the liquid mass density, and the dynamic viscosity of the liquid, respectively. Lam et al. proved that the viscous term and the surface tension term in the equation above can be neglected by parameter calculation. Moreover, considering adiabatic evolution and neglecting mass transfer from and to the bubble, the bubble pressure is found,

\[
P_B(t) = (1 - \gamma) \left\{ P_l + \frac{3\rho}{2} \left( \frac{R^2}{C_l} \right) \right\},
\]
where $\gamma$ is the heat capacity ratio and $C_1$ is the integration constant. The integration constant $C_1$ is then determined at the bubble maximum, where $R = 0$. The instantaneous temperature $T_B(t)$ can be deduced from the isentropic relation for an ideal gas,

$$ T_B(t) = T_0 \left( \frac{P}{P_0} \right)^{\frac{\gamma-1}{\gamma}}, $$

where $P_0$ and $T_0$ used in the RP calculation correspond to the pressure and temperature of the first snapshot, respectively.

In this paper, the interpolation of $R$ is obtained when $R$ is extracted numerically by fitting $R^2$ with a 5th order polynomial regression. $R$ and $\dot{R}$ are calculated by mathematical differentiation, whereas $R'$ is obtained from a direct fit. $P_l$ was defined at 2 bars, which can be attributed to a localized gradient of liquid density as reported by Lombared et al. $\sigma$ was defined at $100 \times 10^{-3}$ N/m at standard temperature and pressure. $\rho$ was defined at $10^3$ kg/m$^3$ and $\eta = 10^{-3}$ Pa s for liquid water. The heat capacity ratio $\gamma$ was defined at 1.3, according to the previous reports.\textsuperscript{22}

The lines in Fig. 6(a) are the fitting curves of bubble center height in the expansion stage. The collapse duration of the first bubble is significantly longer than its expansion duration, as shown in Fig. 6(a), which indicates that the evolution of the first bubble is asymmetric. We can find that the bubble center height and bubble life increase with increasing interval distance. A good fitting effect of the first bubble at the expansion stage with the Al-plate interval at 5 mm and 8 mm is obtained by RP calculation. The result further validates that the Bjerknes force has little effect on bubble evolution at the expansion stage but a great influence on it at the collapse stage. Due to the bubble deformation caused by strong spatial constraints under a short interval distance, the bubble center height of the interval at 3 mm is not well fitted compared to 5 mm and 8 mm. Figures 6(b) and 6(c) show the evolution of the pressure and rescaled temperature inside the first bubble at the expansion stage obtained with the interval at 3 mm, 5 mm, and 8 mm. From the result of calculation, the pressure and temperature within the bubble show a similar evolution at different interval distances.

Since the Bjerknes force has a significant effect on bubble morphology evolution in the collapse stage, the effect of bubble surface tension must be considered. For laser bubbles in water, the pressure on the bubble surface must meet the continuous condition, that is, the pressure at the point on the bubble surface is equal to the sum of the pressure of the gas in the bubble and the surface tension component. So, the bubble will be subjected to an additional pressure pointing to the inside of the bubble in the process of expansion and contraction under the action of interfacial tension.\textsuperscript{46} This pressure will favor the bubble contraction while consequently suppressing its expansion. Therefore, the existence of interfacial tension may affect the bubble expansion speed and oscillation period.

FIG. 6. (a) The graph displays the temporal bubble center height evolution with the Al-plate interval at 3 mm, 5 mm, and 8 mm. The lines in the figure are the fitting curves of bubble center height at the expansion stage. (b) Bubble pressure at the expansion stage obtained with the interval at 3 mm, 5 mm, and 8 mm. (c) Rescaled temperature $T_B(t)/T_0$ at the expansion stage obtained with the interval at 3 mm, 5 mm, and 8 mm.
The combined action of the Bjerknes effect and the bubble surface tension causes the asymmetry of bubble evolution. When the bubble reaches the maximum radius, its pressure is reduced to a saturated vapor pressure of the liquid. At this time, the inner pressure inside the bubble is much less than the pressure of the surrounding liquid, and the bubble starts to shrink. However, due to the Bjerknes effect on the two side walls, the bubble is attracted by the two side walls in the contraction stage, and the top of the bubble shrinks faster than the edge of the bubble. As a result, the ratio of center height to width of bubbles decreases rapidly after 240 µs, leading to a smaller curvature of the vapor-liquid interface, which can be verified in Fig. 5. So, the smaller the curvature, the smaller the tension. As the tension at the vapor-liquid interface at the top of the bubble decreases, its promoting effect on bubble contraction also decreases, and thus, the bubble contraction takes longer. This could explain the phenomenon that the bubble lifetime and asymmetry increase with increasing interval distance. There are still many phenomena to be studied because of the complex interaction between bubbles and structures. It can be considered to use the numerical simulation method to study the motion and evolution of laser bubbles in constrained conditions in the future.

IV. CONCLUSIONS

In this paper, the evolution of bubble and shock waves in space confinement has been studied under water by the time-resolved shadowgraphy images. We have discovered a new phenomenon in the process of the shrinkage of laser bubbles. Due to the constraint of the Al-plate walls, the bubble center shrinks faster than the bubble edge and collapses before the edge of the bubble, splitting into left and right small bubbles that continue to shrink down. After the first bubble completely collapses, the second bubble begins to form from the lower left corner and the lower right corner and tends to converge in the middle. We attribute this special phenomenon to the effect of the Bjerknes force on bubble motion characteristics. When bubbles oscillate near the two Al-plate walls’ surface, they will be slightly repelled by the structure surface at the expansion stage and strongly attracted by the structure surface at the collapse stage. In order to show a gradual transition from constrained conditions to unconstrained conditions, we study the evolution of bubbles with different Al-plate intervals from 3 mm to 8 mm. A good fitting effect of the first bubble at the expansion stage with the Al-plate interval at 5 mm and 8 mm is obtained by the Raleigh-Plesset equation compared to 3 mm. We can find that the bubble center height and bubble life increase with increasing interval distance. The result further validates that the Bjerknes force has little effect on bubble evolution at the expansion stage but a great influence on it at the collapse stage. Although previous papers have reported the influence of laser ablation in liquids on a finite size target, we provide more detailed analysis, including shockwave propagation and the pressure and temperature inside the bubble with different interval distances. This work is expected to be a useful reference for the study of bubble dynamics in different environments and boundary conditions.

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