Brightened single-bubble sonoluminescence by phase-adjusted high-frequency acoustic pulse

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This paper experimentally and numerically studies the effect of a high-frequency acoustic pulse on brightening single-bubble sonoluminescence (SBSL). A polyvinylidene fluoride point-focusing transducer was driven by a 700-W pulse generator to superimpose the acoustic pulse on the sonoluminescing bubble. The center frequency of the pulse was 10 MHz and the duration was 0.15 μ s. The pulse was triggered every 100 cycles of the low-frequency standing wave used to make SBSL. The intensity of SBSL was measured as a function of time lag of superimposed pulse. Only the pulse that arrived at the bubble at the early growing stage could increase the brightness. This trend was confirmed with a numerical calculation based on the Rayleigh-Plesset equation. The increased brightness reached 300% of those of the classical SBSL flashes when the time lag was correctly adjusted.

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I. INTRODUCTION

An acoustic standing wave traps a single bubble at the antinode spot in water, which repeats growth and collapse, and emits flashes of ultraviolet light with picosecond duration at every collapse. This phenomena, single-bubble sonoluminescence (SBSL) [1-3], has received extensive study by many researchers for the purpose of clarifying its physical background. Nowadays, much attention is being paid to seek applications of SBSL, including purification of water [4]. A demand always exists for giving them realities, that is, intensification of the SBSL flashes. Theoretical and numerical studies [5-8] indicate that the brightness of the SBSL flashes can be increased by increasing the bubble-wall velocity when the bubble collapses. A classical way to achieve this is to increase the standing-wave amplitude so as to raise the maximum bubble radius and to cause a faster convergence of the bubble toward collapse. However, there is a limit of the standing-wave amplitude, beyond which the bubble becomes unstable and fails to emit the flashes, corresponding to the brightness limit.

To overcome this difficulty, two approaches appear: (1) use of multiple or harmonic standing waves [9–12], and (2) use of an acoustic impulse to hit the bubble [13–15]. The former simultaneously introduces a low-frequency and high-amplitude standing wave for giving rise to SBSL, and higher-frequency and lower-amplitude standing waves for stabilizing the bubble even for the acoustic amplitude beyond the classical limit. Holzfuss, Rüggeberg, and Mettin [9] reported a 300% enhancement of the SBSL intensity. Krefting, Mettin, and Lauterborn [12] achieved a 250% gain. But, Seeley [10] did not find any enhancement with the same type of experiment as Holzfuss's work. Thus, this method is still open to questions as to the effectiveness.

The latter approach uses the high-frequency acoustic impulse on the oscillating bubble. Moss *et al.* [8] and Matsuda, Ogi, and Hirao [13] suggested that either a short acoustic pulse introduced at the bubble collapse or a long negative acoustic pulse at bubble's growing stage would cause a higher velocity of the bubble collapse and then a brighter SBSL. Hargreaves and Matula [14] applied an acoustic pulse by a focused immersion transducer with 1.88 MHz center frequency from the top of the sonoluminescing bubble and observed the doubled brightness. Matsuda, Ogi, and Hirao [13] also achieved the SBSL intensification in a similar magnitude by applying a 1-MHz acoustic pulse to the bubble before the collapse. Thomas, Forterre, and Fink [15] used a spherical grass cavity with two piezoelectric transducers for driving SBSL and eight piezoelectric transducers with center frequency of 0.7 MHz for focusing the acoustic pulses on the sonoluminescing bubble at the center. They obtained a 90% gain of the light intensity.

Thus, these previous studies actually achieved enhancement of the SBSL intensity by adding an acoustic pulse. However, frequencies of the added pulses were so low that the major sound fields would be changed for several cycles after the excitation of the pulse. Indeed, Matsuda, Ogi, and Hirao [13] observed such a modification of the fundamental standing-wave amplitude after adding the pulse. In this case, it is unclear that the bubble is actually brighter than that for the classical case. Therefore, a short and high-frequency pulse will be ideal to research the effect of the addition of the pulse. Furthermore, there is no systematic study as for the phase relation between the standing-wave and the superimposed pulse. This is the key to increase the maximum bubble radius, the velocity at the collapse, and then the SBSL brightness. Here, we explore the effect of the pulse's time lag on the brightness gain using an acoustic pulse with 10-MHz frequency driven by a 700-W pulse generator. The result is favorably supported by a numerical computation. We obtained a 300% gain for the brightness by applying the pulse at the best suitable time lag.

II. MEASUREMENT

Figure 1 shows our measurement setup. We glued two hollow-cylinder ceramic transducers on the opposite sides of the rectangular epoxy cell $(60 \times 60 \times 80 \text{ mm}^3)$ and filled it with distilled water containing 25% glycerin. The frequency synthesizer has two outputs synchronized each other: one output was a sinusoidal wave with frequency *f* for driving SBSL after amplification and the other was a rectangular



FIG. 1. Setup for measuring the intensity of SBSL flashes by applying the acoustic pulse. f denotes the frequency used to make SBSL.

signal with frequency f/100 for triggering the acoustic pulse described below. Driving the transducers with the sinusoidal wave of the resonance frequency (~40 kHz) caused the standing wave in water. As the driving pressure increased, a single bubble appeared at the center antinode and emitted visible flashes, which were detected by the photomultiplier located outside the cell. We increased the standing-wave amplitude close to the threshold, beyond which the bubble became unstable and failed to emit flashes. All the measurements were done, keeping this upper limit of the standingwave amplitude.

We applied a short and high-amplitude impulse to the sonoluminescing bubble, using a point-focusing polyvinylidene fluoride (PVDF) polymer transducer with 30 mm focal length and 10 MHz center frequency. The PVDF transducer was set near the top of the cell to locate the focal point on the bubble. A square pulse of 700 W power from the pulse generator drove the PVDF transducer. Figure 2 shows the acoustic pulse generated by the PVDF transducer, reflected at the bottom surface of the epoxy cell, and received by the Same transducer. The duration of the pulse was 0.15 μ s. The FFT spectrum showed a peak near 10 MHz being much higher than the standing-wave frequency. From the driving voltage and piezoelectric coefficient of the PVDF polymer, we roughly estimated the acoustic pressure of the pulse at the bubble to be more than five times larger than the pressure of



FIG. 2. Wave form of the acoustic pulse generated and received by the PVDF transducer.

the standing wave. Because of a limited repetition rate of the pulse generator, we applied the acoustic pulse every 100 cycles of the standing wave, using the triggering signal from the second output of the synthesizer. Since the two outputs are synchronous to each other, we could control the arrival time of the acoustic pulse at the bubble by changing the phase between them.

III. NUMERICAL CALCULATION

It is widely accepted that the bubble dynamics obeys the Rayleigh-Plesset equation [5-7]. Here, we adopt the Rayleigh-Plesset equation including the liquid compressibility [6] for predicting the effects of the acoustic impulse on the maximum bubble radius, which is highly related to the velocity at the collapse and the SBSL brightness. The governing equation takes the form

$$\left(1 - \frac{\dot{R}}{c_{\infty}}\right) R\ddot{R} + \frac{3}{2} \dot{R}^{2} \left(1 - \frac{\dot{R}}{3c_{\infty}}\right)$$

$$= \frac{1}{\rho_{\infty}} \left(1 + \frac{\dot{R}}{c_{\infty}}\right) \left\{p_{g}(R, t) - \frac{4\eta \dot{R}}{R} - \frac{2\sigma}{R} + p_{A} \left(t + \frac{R}{c_{\infty}}\right) - p_{\infty}\right\} + \frac{R}{\rho_{\infty}c_{\infty}} \frac{dp_{b}(t)}{dt}.$$

$$(1)$$

Here, t denotes time, R the bubble radius, ρ_{∞} the ambient liquid density, p_{∞} the ambient pressure, and c_{∞} the sound speed in the liquid at ambient pressure. $p_A(t)$ is the driving acoustic pressure and expressed by $p_A(t) = P_A \sin \omega t$ with angular frequency ω and amplitude P_A . η denotes the dynamic viscosity of the liquid and σ the surface tension. $p_g(t)$ represents the pressure on the gas side of the bubble wall, for which we assume $p_g = P_0 (R/R_0)^{-3\gamma}$ from the adiabatic conservation equation with the radius R_0 and pressure P_0 inside the bubble at equilibrium, and the specific-heat ratio γ . This simple expression for p_g prevents us from predicting precisely the radius change at the bubble collapse because the van der Waals effect appears in such an extremely small volume [5]. However, the expression remains applicable for the aim of investigating the effect of adding the acoustic impulse on the maximum bubble radius. In Eq. (1), we ignored the vapor pressure and the viscosity of the gas because of their small contributions.

We modeled the short acoustic pulse $p_P(t)$ with a singlecycle acoustic wave and included it in $p_A(t)$:

$$p_A(t) = P_A \sin \omega t + p_P(t), \qquad (2)$$

$$p_P(t) = \begin{cases} P_P \sin[\omega_P(t-\tau_P)], & \tau_P < t < \tau_P + 2\pi/\omega_P \\ 0, & \tau_P > t \text{ or } t > \tau_P + 2\pi/\omega_P, \end{cases}$$
(3)

where ω_P and P_P are the angular frequency and amplitude of the acoustic pulse. τ_P is the time lag relative to the standing wave (see Fig. 3).



FIG. 3. Calculated bubble radius change (upper) and introduced acoustic pressures (below).

We numerically solved Eq. (1) for *R* in time using a finitedifference method. The values used were those for water with 25% glycerin [1]; $p_{\infty}=1$ atm, $c_{\infty}=1563$ m/s, $\eta=1.59$ $\times 10^{-3}$ Pa s, $\sigma=6.9\times 10^{-2}$ N/m, $\rho_{\infty}=1059$ kg/m³, P_A = 1.3 atm, and $P_P/P_A=5$. We assumed that $R_0=5 \ \mu$ m.



FIG. 4. Calculation of the increased maximum bubble radius by the acoustic pulse as a function of the time lag, τ_p . The maximum bubble radius is normalized by that of the classical SBSL (=25.76 μ m) without adding the pulse. The bottom figure includes calculations when the cycle number and amplitude of the acoustic pulse were increased.



FIG. 5. SBSL intensity detected by the photomultiplier (single shot) and the standing-wave signal at the bubble position (broken line). The vertical broken line indicates the time when the acoustic pulse arrived at the bubble. The origin of the horizontal axis is arbitrary.

IV. RESULT AND DISCUSSION

Figure 3 exemplifies calculated bubble radius change with and without the acoustic pulse. The high-frequency pulse increases the maximum radius, but it hardly affects the time of collapse, indicating an increase of the velocity at the collapse and then a brightened flash. Figure 4 shows the normalized maximum bubble radius as a function of the time lag of the pulse, τ_P . The time lag that causes the highest gain is insensitive to the frequency. The maximum efficiency occurs at $\tau_P = 2-3 \ \mu$ s. This means that an effective increase of the maximum bubble radius is only possible in the early stages of bubble's growth. The acoustic pulse will encourage the negative pressure of the standing wave to reach a larger maximum of radius.

The gain for the maximum radius decreases as the frequency of the pulse increases, as shown in Fig. 4, because a low-frequency pulse includes longer negative pressure and enhances the negative pressure of the standing wave more effectively. This view suggests that a long negative pulse would cause much larger maximum radius. Actually, we numerically confirmed that this is true. However, such an acoustic pressure of only negative polarity cannot be gener-



FIG. 6. Dependence of the SBSL intensification on the time lag of the pulse. I_p denotes the intensity of the flash just after the pulse arrival and I_0 denotes that before the pulse arrival. Two independent measurements are shown together with the numerical simulation of the maximum radius for the 10-MHz pulse [solid line in Fig. 4(c)].

ated in practice because elasticity of water must cause a positive pressure and then an oscillation, which disturbs the standing wave to a large extent and makes the bubble unstable. Pulses with lower frequencies also vary the major acoustic field caused by the standing wave. Thus, a highfrequency and high-amplitude acoustic pulse is preferable for increasing the bubble radius, keeping the bubble stable without disturbing the major acoustic field.

In the numerical calculation, we assumed a single cycle and the amplitude five times larger than the standing-wave amplitude for the acoustic pulse. However, actual acoustic pulse consists of two cycles rather than single cycle and the amplitude when the pulse hits the bubble is unknown. However, both the cycle number and amplitude of the pulse are insensitive to the time lag to provide the maximum gain, as shown in Fig. 4(c), although they affect the gain of the maximum radius.

Figure 5 shows the measurement of the SBSL flashes when the acoustic pulse was applied by the PVDF transducer. Before the pulse arrival, no acoustic pulse exists and therefore the brightness of the flashes is the same as that of

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the limit of the classical SBSL flashes. After the pulse was added, the brightness was obviously increased. We averaged the outputs of the photomultiplier 500 times and determined the intensification rate by comparing the flashes before and just after the pulse arrival. The results are compared with the numerical calculation for the 10-MHz pulse in Fig. 6. The measurements showed similar trend with the calculation. Especially, the maximum gain occurs at the time of around 3 μ s. We achieved approximately 300% gain with this technique.

V. CONCLUSION

We were able to increase the brightness of SBSL up to 300% by applying a high-frequency and high-amplitude acoustic pulse on the bubble. The pulse frequency was near 10 MHz and the duration was 0.15 μ s. The brightness gain was measured by changing the time lag of the pulse relative to the standing wave. The maximum efficiency occurs by superimposing the pulse at the early growing stage of the bubble. A numerical simulation confirmed this result.

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