## Inhomogeneous pressure field inside a collapsing bubble accelerated by an acoustic pulse

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The temperature inside a collapsing bubble could become very high if a converging shock wave were launched inside the bubble. Even if the collapse speed reaches Mach 4, the recent models and experiments show that this shock wave is not necessary to explain the sonoluminescence. Its hypothetical existence depends strongly on the assumption made in the various numerical models proposed. However, it has been established that its generation depends strongly on the acceleration of the bubble surface. This work presents an experimental parametric study demonstrating that a pressure pulse applied on the bubble with an accurate timing significantly accelerates the bubble collapse. It is shown that the induced brightness gain is very sensitive to the time of arrival of the pulse. Moreover, a numerical simulation of these experiments relates this dependence to the gas dynamic in the bubble.

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Single bubble sonoluminescence (SBSL) [1] is one of the most effective systems to concentrate energy by inertial confinement [2,3]. This remarkable result is obtained by fast compression of the gas contained inside the bubble, which is collapsing with a speed close to Mach 4 [2]. This very fast compression of the gas induces a rise in the gas pressure of several tens of thousands of atmosphere [4]. The gas density is then close to that of the liquid. The estimate of the gas temperature fluctuated by four orders of magnitude according to the assumptions controlling the gas dynamics [5–8]. These fluctuations mainly come from the hypothetical existence of an acoustic shock wave inside the bubble, which would come to still improve the inertial confinement and lead to very high temperature at the bubble core. Several arguments do not support this last assumption. The first of them came from precise measurement of the flash duration by time correlated single photon counting [9]. Values ranging between 50 and 350 ps were thus obtained and are compatible with radiation mechanisms such as bremsstrahlung radiation in a weakly ionized plasma [6,7]. However, radiation by a tiny blackbody is another possiblity [2,3]. The second argument came from recent models, which predict that the vapor ends up saturating the effectiveness of the inertial confinement [8]. This last argument is supported, on the one hand, by the quenching induced by the molecular excitation of the liquid vapor that absorbs 100 times more energy than the optically radiated one [10]. On the other hand, it is supported by the tenfold bubble brightness rise when the temperature of water is lowered close to 0°C<sup>2</sup>. Moreover, in a recent numerical study, homologous bubble interior dynamics has been shown to be compatible with a uniform gas pressure for the SBSL domain of stability [11]. However, water vapor by decreasing the speed of sound in the bubble could promote the occurrence of a shock wave [6,8]. This effect is reinforced by taking into account water vapor diffusion in the gas mixture, but again no shock wave occurs for

argon or helium filled bubbles, whereas for a xenon bubble, a wave disturbance exists only in the last instant of the collapse and evolves actually in a shock wave after a rebound on the bubble wall [12]. This paper addresses the feasibility of modifying the SBSL domain of stability to reach this shock wave dynamic in the bubble interior. This study is conducted by focusing an acoustic pulse on a bubble in the sonoluminescence regime. This manuscript reports a parametric study of the effect of this acoustic pulse by varying the time of arrival of the acoustic pulse and the amplitude of the low monochromatic pressure field driving the sonoluminescing bubble. These experimental results are compared with a numerical simulation, and the validity of a uniform pressure inside the bubble is discussed.

This technique must be distinguished from the methods consisting in increasing and lengthening the phase of expansion of the bubble by the addition of a pulse at the beginning of this stage [13]. In this last case, the bubble gas mixture is probably enriched with water vapor before the collapse. The amplification of the sonoluminescence is obtained here by focusing a positive pressure pulse at the end of the bubble collapse in sonoluminescence regime [14,15]. Moreover, when the experiment is repeated, several seconds of intervals are used to be sure to set out again under the same conditions. The water temperature is 25°C, the measurement of dissolved oxygen after degasing lies between 1.0 and 1.2 mg/L, and the frequency is 27 855 Hz. The experimental setup and the procedure used to focus the pulse wave are described in a previous paper [14]. However, the electric voltage delivered on the eight transducers has been multiplied by 4.

An average on 44 measurements of the Mie scattering with a He-Ne laser is fitted by the radius square resulting from a simulation of the Rayleigh-Plesset equation (RPE), Fig. 1. In this figure, the electric signal from the photomultiplier gives both the time evolution of the radius square and the intensity of the flash. One finds the best agreement for  $R_0$ =6  $\mu$ m and  $P_a$ =1.37 atm, which corresponds well to the high threshold of the stability domain measured by Gaitan and Holt [16]. In this numerical simulation, I used the equa-

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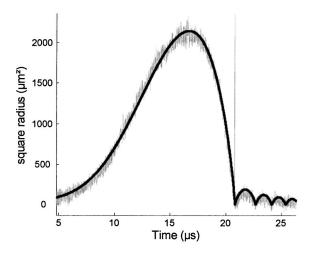


FIG. 1. Gray curve, photomultiplier output; solid line, fit by a  $\mathsf{RPE}.$ 

tion derived by Prosperretti and Lezzi [17], which uses the enthalpy rather than the pressure. This equation was also derived by a nonpertubative analysis for a homologous bubble interior [18]. The polytropic exponent is equal to 1, unless  $R < R_0$ , in which case it is fixed at  $\frac{5}{3}$ . The acoustic pressure is then measured with a needle hydrophone calibrated by this estimate ( $P_a$ =1.37 atm,  $R_0$ =6  $\mu$ m). The threshold for which the bubble becomes unstable and starts to dance is 1.41 atm.

When a focused acoustic pulse is sent with a good timing, a brightness gain of about 500% is obtained (Fig. 2 scale of the left side, gray curve). The Mie scattering measurement shows unambiguously that the bubble dynamic is periodic before the acoustic pulse arrival. The gain is computed as the ratio between the intensity of the amplified flash and an average on the flash intensity of the 29 acoustic cycles preceding the acoustic pulse. Here  $P_a$ =1.37 atm and the pulse impinges on the bubble 0.5  $\mu$ s before the end of its collapse. This behavior was simulated with the RPE previously described (Fig. 2 scale of the left side, black solid line). To

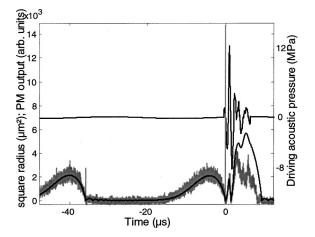


FIG. 2. Scale of right: the gray curve is the photomultiplier output, the superposed solid line is the fit by a RPE. Scale of left: the upper solid line is the applied pressure. Without the acoustic pulse, the flash would occur at t=0.

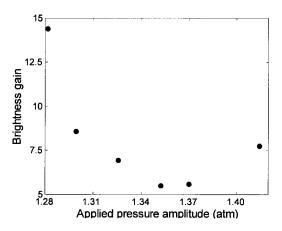


FIG. 3. Brightness gain for different applied pressure amplitudes; the pressure pulse has a constant time shift of  $-0.62\pm0.03~\mu s$ .

simulate the dynamics of the bubble subjected to a pressure pulse, the signal emitted by one of the eight transducers was measured at bubble range, i.e., 3 cm, in a free environment with a calibrated hydrophone. To compute the pressure applied on the bubble, this figure is multiplied by the number of transducers, i.e., eight. Indeed, the adaptive focusing technique [14] ensures that a constructive interference is achieved on the bubble position with an interelement accuracy of T/25, where 1/T=700 kHz is the central frequency of the piezoelectric transducer. This experimentally measured signal is then added to the computed low-frequency applied pressure driving the dynamics of the bubble in the numerical simulation, Fig. 2 (scale of the right-hand side, solid line). Its precise location is determined by the travel time of the pressure pulse from the transducer to the bubble. In order to correct for any jitter of the trigger, the periodic timing of the flashes is used as a time reference. Thus, a first numerical simulation without any acoustic pulse is carried out in order to compute the location of the end of the collapse. This time is synchronized with the experimentally measured time location of the flash. Thereafter, the origin of time, t=0, is determined by the measurement of the flash occurrence when no acoustic pulse is applied, see Fig. 2. Then in a second simulation, the acoustic pulse is added. This applied pressure including the low frequency at 27 855 Hz and the acoustic pulse starting at  $-0.5 \mu s$  is displayed in Fig. 2 (scale of the right-hand side, solid line). Note that only the first rather small lobe of the pulse is used to boost the collapse and that thereafter the acoustic pulse induces a fast inflation of the bubble followed by another collapse. This behavior is very well reproduced by the RPE equation up to the second inflation stage, which last a longer time in the RPE simulation.

The brightness gain varies according to the amplitude of the applied pressure of frequency 27 855 Hz and its evolution is displayed in Fig. 3. For instance, a gain of about 1400% is obtained for an applied pressure amplitude of 1.28 atm. Thereafter, the applied pressure amplitude is fixed at  $P_a$ =1.37 atm just below the threshold of bubble instability measured at 1.41 atm.

To study the influence of this pressure pulse on the dynamics of the bubble, the position of the pulse is shifted from

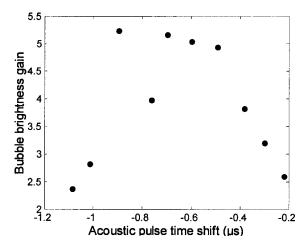


FIG. 4. Brightness gain with acoustic pulse time shift ranging from -0.2 to  $-1.1~\mu s$  before the normal flash location.

 $-0.2 \mu s$  down to  $-1.1 \mu s$ , Fig. 4. Until  $-0.5 \mu s$ , the gain is increasing; then a plateau is observed and finally a decrease starting from  $-0.9 \mu s$ . The interpretation of the first part of this curve is obvious: the more the pulse arrives early and the more the induced acceleration has time to affect the dynamics of the bubble, the faster is the collapse and the better is the inertial confinement. In the second part, this effect is balanced by the braking generated by the change of sign of the acoustic pulse after 0.5  $\mu$ s, i.e., the duration of the first positive half-cycle; see Fig. 2 for the pulse shape. It would not occur if one were able to synthesize monopolar pressure waves. Moreover, this braking may induce an instability destroying the bubble sphericity before the end of collapse. In the third part, this braking is increasingly significant and reduces the gain. Longer time shifts are not presented since, in most cases, the bubble is destroyed. This threshold around  $-1.2 \mu s$  is probably due to bubble shape instability. This point will be discussed below. By the way, another very efficient way to destroy the bubble is to send a negative pressure pulse.

To confirm the increase of the collapse speed, the shift of the flash according to the position of the acoustic pulse was measured (Fig. 5, full circles). Again, a negative time shift signifies that the flash occurs sooner than usual when the

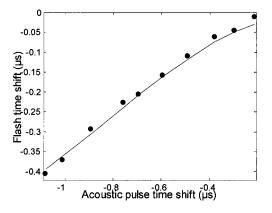


FIG. 5. Flash time shift vs acoustic pulse time shift. Full circles, measurements; solid line, numerical simulation.

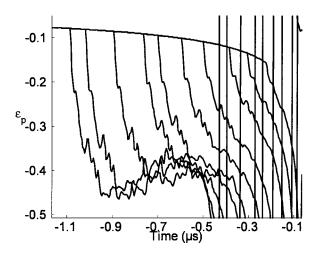


FIG. 6. Computed time evolution of  $\varepsilon_p$  before the first rebound for pulses arriving on the bubble between 1.1 and 0.2  $\mu$ s with a step of 0.1  $\mu$ s before the normal flash location.

acoustic pulse is applied. One observes a quasilinear evolution with a slope of 0.44. This confirms that the flash shift increases less quickly than the pulse shift. The first positive half-cycle of the acoustic pulse is  $0.5~\mu s$ . Thus the applied pressure sign changes before the end of collapse for an acoustic pulse shift of about  $-0.65~\mu s$ , for which the flash shift is  $-0.15~\mu s$ . This behavior has been fitted with the RPE. The measured time shift of the flash is compared with the computed time shift of the end of the collapse when the acoustic pulse shift is varying (Fig. 5, solid line). For this parameter, very good agreement is obtained with the experiment. Thus, this result seems to confirm that, even for these very violent collapses, a RPE closed by a homogeneous polytropic gas is a valid approximation.

To check this last assumption, the results of Lin *et al.* [11] and Elze *et al.* [18] for homologous bubble interior dynamics are used. To this end, the values of  $\varepsilon_p$  and  $\varepsilon_{error}$  are computed, Eqs. 2.9 and 4.12 of Ref. [11]. These two parameters measure the pressure difference between the center and the surface of the bubble and the amplitude of the acoustic wave emitted inside the bubble, respectively. We use here the simpler expression of Lin *et al.* [11],

$$\varepsilon_p = \frac{\rho(t)R(t)\ddot{R}(t)}{\gamma p_c(t)}$$

and simply take the uniform Van de Waals pressure as pressure at the bubble center,  $p_c(t)$ . This crude approximation underestimates the variations of  $\varepsilon_p$  as the pressure at the bubble center should increase more slowly as soon as the pressure becomes heterogeneous;  $\varepsilon_p$  is negative up to the end of the collapse and hence the pressure at the center is smaller than the pressure at the bubble wall. The value of  $\varepsilon_p$  computed by using R(t) from the preceding simulation leads to Fig. 6. Ten curves are plotted corresponding to an acoustic pulse shifted from -0.2 to -1.1  $\mu s$  with a -0.1  $\mu s$  step. One clearly observes a first fast variation of  $\varepsilon_p$  as soon as the pressure pulse hits the bubble and a second one at the end of the collapse. This result shows that the interior of the bubble

can no longer be regarded as uniform as soon as the acoustic pulse reaches the bubble. One can also see the appearance of a plateau for acoustic pulse shifts lying between -0.6 and  $-0.9 \mu s$ . This plateau is replaced by a slight bump for the longest shifts -1.0 and  $-1.1 \mu s$ . Thus, this behavior closely matches the brightness gain variation of Fig. 4. To check the other possibility, namely the shape instability hypothesis, even longer time shifts were simulated. Only for  $-1.3 \mu s$  is the acoustic pulse strong enough and the bubble wall acceleration becomes positive before reaching the hard-core radius. In contrast, for shorter time shift the inertia of the bubble is too high and hence the acceleration keeps decreasing monotonously. This threshold matches the empirically assessed bubble destruction threshold. This is perfectly in agreement with a Rayleigh-Taylor instability. So, the most probable hypothesis is that the brightness gain variation is actually related to the gas interior dynamics and the gas pressure nonuniformity.

In addition, for an acoustic pulse shift of  $-0.6~\mu s$ , the time evolution of  $\varepsilon_{error}$ , which accounts for the fast change in surface acceleration when the pulse arrives, shows that acoustic waves of great amplitude are radiated inside the bubble at this time, Fig. 7. Another fast change is observed at  $-0.2~\mu s$ , which corresponds to the end of the collapse, see Fig. 5.

This simple numerical simulation captures the main characteristics of the brightness gain evolution with the time of arrival of the acoustic pulse. On the one hand, for an acoustic pulse shifted more than  $-1.2~\mu s$ , the bubble wall acceleration becomes positive before the minimum radius is reached. This is the necessary condition to get a Rayleigh-Taylor instability without the stabilizing mechanism of bubble expansion. On the other hand, the appearance of a plateau followed by a decrease of the brightness gain, Fig. 4, is correlated with the departure from uniform pressure inside the bubble. Indeed, after the first fast decrease of  $\varepsilon_p$  induced by the pulse arrival, there is a plateau and even an increase for the longest time shift of the acoustic pulse, Fig. 6. The wave disturbance is generated much sooner (500 ns before the end of the collapse) than in classical SBSL, in the last nanosecond if any,

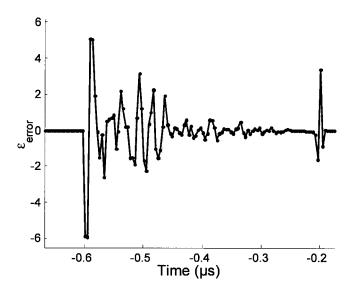


FIG. 7. Computed time evolution of  $\varepsilon_{\rm error}$  before the first rebound for a pressure pulse arriving on the bubble 0.6  $\mu$ s before the normal flash location.

and hence it has a lot of time to build up by spherical convergence. However, if this wave is launched too soon, i.e., the acoustic pulse is too weak, then the pressure tends to become uniform again before the end of the collapse.

Even if this demonstrates that the collapse is much more violent than in classical SBSL, a more complete modeling of the bubble interior dynamic is required to conclude about the existence of a shock wave. Another experimental challenge would be to measure the spectra of the boosted flash to look for a shift of the spectral weight. In any case, if the shock wave is not yet present, nothing seems to limit this method towards stronger acoustic amplitudes. This impulse technique opens a new domain of stability for SBSL, for which the gas interior dynamics has to be taken into account. Reciprocally, the measured bubble brightness variation with the pulse shift could also be used to validate bubble interior dynamics and sonoluminescence models.

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