Ambient Pressure Effect on Single-Bubble Sonoluminescence

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We report the first results on the influence of ambient pressure on single bubble sonoluminescence. The equilibrium radius as well as the maximum radius of the bubble increased as the ambient pressure over the liquid head was decreased. The number of photons emitted during a sonoluminescence pulse increased linearly with a decrease of ambient pressure. These results are in good agreement with the theory of the ambient pressure effect [L. Kondic *et al.*, Phys. Rev. E **57**, R32 (1998)] and with the recent dissociation hypothesis.

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Single bubble sonoluminescence (SBSL) has recently attracted a lot of attention. The reasons for continuing interest in this effect are plentiful: large energy concentration, possibility for some exotic physics, and also a close connection with industrially relevant applications of ultrasound in such diverse fields as sonochemistry, lithotripsy, ultrasonic cleaning, etc.

Recent experimental and theoretical work explained many aspects of sonoluminescence. The main characteristics of SBSL, such as the intriguingly short time scale of the light emission (less than 300 ps) [1,2] and the apparent continuous spectrum of the emitted light [3] have been understood, at least in a qualitative fashion [4,5]. The details of the SBSL emission have been explored in different liquids, with varied liquid temperature, gas content, etc. (see [2] for an extensive review). However, there has been no detailed experimental study of the change of SL emission with variation of ambient pressure.

In this Letter, we present the first complete and well controlled experimental study of the influence of the change of ambient pressure P_0 on bubble dynamics and the resulting SBSL. By combining experimental results with theoretical analysis, we are able to gain further insight into the basic mechanism of the SL emission. The importance of this research is emphasized by the need to better understand other experiments where a variation of P_0 might occur, such as microgravity experiments [6], or closely related experiments performed in a strong magnetic field [7]. Next, our experimental results are relevant to the recently proposed "dissociation hypothesis" [8]. This theory suggests that nitrogen and oxygen from air bubbles dissolve in the liquid, so that only argon is present in a bubble for long times. The dissolution hypothesis received strong experimental support from rather elaborate experiments, which are consistent with the idea of pure argon bubbles [9,10]. Our experimental results provide direct and straightforward verification of this theory.

Figure 1 shows the experimental setup. In the experiments an air bubble was trapped in a 500-ml spherical glass flask which was driven harmonically at about 17.5 kHz by a piezoelectric transducer at the fundamental resonance frequency of the flask system. The liquid used was a 25% by weight degassed water/glycerin mixture, kept at a constant temperature of 21 °C. The air concentration in the liquid was estimated by measuring the concentration of dissolved oxygen by an oximeter (Orion model 810). A small disk shaped pill transducer was mounted on the wall of the flask to calibrate and monitor the acoustic driving pressure in the flask in a noninvasive way.

The ambient pressure over the liquid was controlled and measured by a gas-handling system connected to a vacuum pump fitted with a vacuum gauge (model Ashcroft type 1082) with an accuracy of 2.54 mm of Hg. The time scales involved in the experiments were much shorter than the diffusion time of air in the liquid so that the concentration of the gas in the liquid remained unchanged.

After the acoustic drive amplitude had been set at the resonance a small air bubble was created by poking the liquid surface with a thin wire inserted from the top through a rubber cap (sub-a-seal). At the low acoustic driving level the bubble was trapped at the center of the flask.

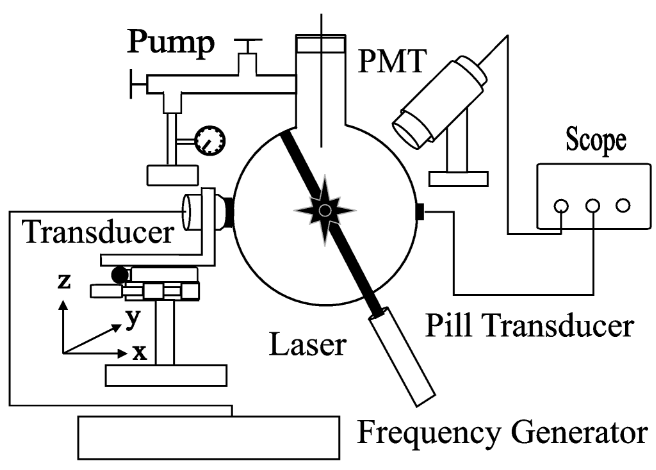

FIG. 1. Experimental setup.

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The driving level was then gradually increased so that the bubble entered the light-emitting regime. The ambient pressure over the liquid head was slowly decreased in controlled steps. However, below a critical value of the ambient pressure the bubble started to move away and finally vanished.

The radius of the bubble as a function of time was measured by the usual Mie scattering technique [11]. The bubble was illuminated by a narrow beam of 30 mW (average power) He-Ne laser (model Melles Griot 05-LPH-991) at 632.8 nm. A photomultiplier (PMT) (model Philips XP2230H) placed at about 80 deg from the forward direction of the incident beam collected the scattered light. The output, $V(t)$, of the PMT, which is proportional to the square of the bubble radius, was fed into a Lecroy (model 9310M) digitizing scope with a sampling rate of 100×10^6 samples/sec (10 ns/point), averaged over 100 runs, and stored into a computer for further processing. The scope was triggered by the sync output of the function generator (Wavetek model 29). The phase of the bubble collapse was measured by putting a needle hydrophone (SPRH-S-1000 from SEA) at the position of the bubble. The phase and amplitude of the output of the pill transducer was measured with respect to those of the needle hydrophone for calibration of the driving pressure amplitudes. The SL signal was measured using the PMT and averaged over 100 runs at constant driving voltage for the PMT. The number of photons has been calculated following Barber *et al.* [12].

Figure 2 shows the square root of $V(t)$, subtracted from the background signal, for different P_0. The acoustic pressure amplitude, P_a, as well as all other experimental parameters are kept constant, and only P_0 is modified. The physical size of the bubble was determined using the Rayleigh-Plesset (RP) equation in the part of the cycle where this equation is known to accurately reproduce the bubble dynamics. While it is possible to measure P_a at the bubble position directly, this procedure is not very accurate. We find that it is advantageous to obtain both P_a and R_0 as fitting parameters directly from the $R(t)$ curve for $P_0 = 1$ atm. For fitting the RP equation to the experimental results, we use (1) the expansion ratio, $R_{\max}/R_0$; (2) the full width at half maximum; and (3) the phase of the first minimum of the $R(t)$ curves. Figure 3a shows both experimental data and the fit for $P_0 = 1.0$ atm. This procedure, which is an extension of a recently proposed suggestion to use $R_0 = R(t)$ at $P_a = 0$ [10], allows us to very precisely determine both P_a and R_0, avoiding the problems related to the scatter of data in the $R(t)$ curves.

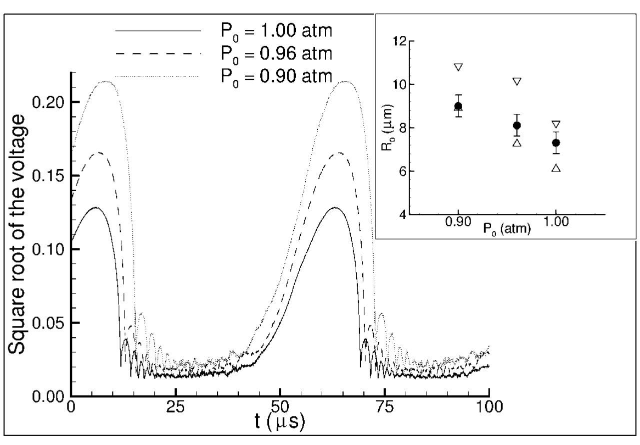

FIG. 2. Experimental $R(t)$ curves as ambient pressure P_0 is varied. The inset shows the results for R_0: experiment (•), and diffusion theory (△ and ▽) obtained assuming polytropic exponent $\gamma = 1.0$ and 1.67, respectively. The estimated combined experimental and fitting uncertainty is also shown.

Once P_a is obtained from the data for $P_0 = 1$ atm, it is kept fixed, so that R_0 is a single fitting parameter for the $R(t)$ curves obtained with different P_0. As can be seen from Fig. 3b, we are able to obtain an almost perfect agreement in the slow expansion part of the cycle where the RP equation is expected to hold. All three quantities (full width, $R_{\max}/R_0$, and the phase of the first minimum) are reproduced very well with a single parameter, R_0. However, the afterbounces are not modeled accurately, implying that there is another damping mechanism that becomes important during the collapse phase, which is not included in the RP equation. The quality of our fitting procedure is not influenced by this discrepancy. More relevant is the heat transfer between the bubble and the liquid, which we model by a polytropic exponent approach. Recent works [13,14] suggest that one should assume perfect heat exchange between the bubble and the liquid; we use this approach and note that the results for our fitting parameters depend only weakly on this assumption. Further, the results are independent of which “bubble” equation is used; as long as one is concentrated on the slow expansion part of the cycle, the RP equation does as good a job as more elaborate (e.g., Keller) equations.

Figure 2 clearly shows the dramatic change of $R_{\max}/R_0$, as well as the modification of R_0 as P_0 is varied. While the change of $R_{\max}/R_0$ can be understood

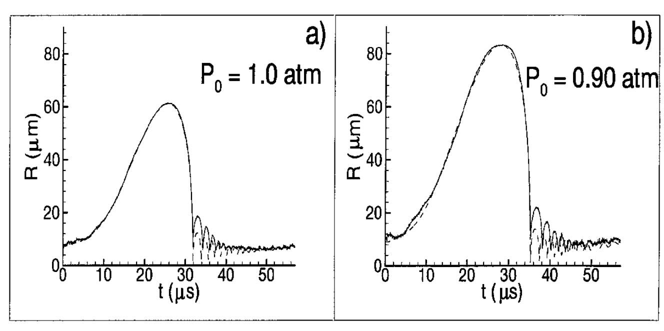

FIG. 3. The experimental results (broken lines) and the fits (solid lines) for two values of ambient pressure, P_0. In (a) the acoustic pressure amplitude, P_a, and R_0 are used as fitting parameters ($R_0 = 7.3\ \mu$m, $P_a = 1.29$ atm); in (b) we use $P_a = 1.29$ atm, and fit only $R_0 = 9.0\ \mu$m. The liquid viscosity is 0.021 cm^2/s, and surface tension is 69.4 dyn/cm.

based on the RP equation [15], the change of R_0 follows from the details of the mass transfer between the bubble and liquid.

It is the interplay between two competing mass flow mechanisms that leads to a stable (constant mass) bubble on a time scale long compared to the acoustic period. One mass flow process is standard diffusion that leads to the flow of the gas out of the bubble and does not depend on P_a. The other one is rectified diffusion, which leads to the flow in the opposite direction, and is due to the asymmetry of the bubble cycle. The bubble spends more time in the expansion part, so that more gas flows into the bubble during this time than escapes during the compression part [16–18]. This effect does depend on P_a, so that there is a value (or values) of P_a when equilibrium is obtained, leading to a time independent equilibrium bubble radius, R_0. R_0 also depends on P_0, and the degree of degassing, c_i/c_0. Here c_i is the concentration of gas in the liquid, and c_0 is the saturation concentration (which depends on P_0 through Henry's law).

The degree of degassing plays a significant role in determining how R_0 depends on P_a and P_0 [15]. If one assumes that the concentration of air in the liquid is relevant ($c_i/c_0 = 0.233$ in Figs. 2 and 3), then the theoretical results predict a *decrease* of R_0 when P_0 is decreased, and P_a kept constant, in obvious contradiction with Fig. 2. Further, the theoretically obtained equilibrium is unstable [15], whereas in our experiment we observe stability on a long time scale, measured in hours. On the other hand, assuming that only argon is left in the bubble, the relevant $c_i/c_0 = 0.0023$. In this case, the theory predicts an *increase* of R_0, as we observe in the experiment. This clear-cut result directly confirms the dissociation hypothesis [8].

Figure 2 (inset) shows experimental and two families of theoretical values for R_0 as a function of P_0. The theoretical values were obtained using $c_i/c_0 = 0.0023$ and the standard Eller-Flynn theory [16], where either isothermal ($\triangle$) or adiabatic ($\bigtriangledown$) conditions in the bubble were assumed. Neither of the theoretical results fits the experimental ones very well; still, we can conclude that the isothermal and adiabatic assumptions are the limiting cases and that the heat exchange between the bubble and the liquid puts the bubble somewhere between these two extremes. However, in both the experimental and theoretical results, there is an obvious trend of increase of R_0 as P_0 is decreased.

Let us now concentrate on the SL emission. Figure 4a shows the number of photons emitted in a single SL pulse, as P_0 is varied, while P_a is kept constant. Strong, roughly linear growth is observed as P_0 is decreased. This result could be qualitatively understood by inspection of Fig. 2. First, $R_{\max}/R_0$ increases as P_0 is decreased, leading to a stronger collapse. Second, there is a shift in the phase relative to the acoustic field as P_0 is reduced; the first minimum of the bubble radius, at which the SL pulse is emitted, happens later in the bubble cycle. Consequently, more time is left for energy storage.

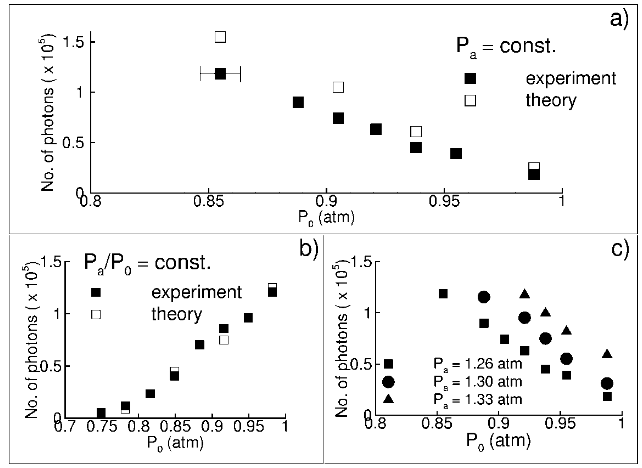

FIG. 4. Number of photons in a single SL pulse versus ambient pressure, P_0 (here $c_i/c_0 = 0.255$): (a) for constant acoustic pressure amplitude, $P_a = 1.26$ atm; (b) for constant ratio $P_a/P_0 = 1.41$; (c) with P_a as a parameter. Typical experimental uncertainty is also shown in (a) (the uncertainty of the number of photons is represented by a symbol size). Experimental results were not corrected for absorption in the liquid and flask walls; theoretical results were scaled down to experimental ones.

The calculation of the intensity of SL radiation is still a subject of debate. The only consistent calculations of SL radiation gave shock wave solutions in the gas during its compression phase and calculated SL intensity based on either bremsstrahlung [4] or radiative transport theory [5]. These studies have been criticized [14,19], since heat conduction between bubble and liquid was not included. The references [14,19] propose that simple adiabatic heating might be responsible for SL emission. This scenario, which predicts correlations between the time span and wavelength of emitted SL radiation, appears to be in contradiction with experimental results [1,20]. However, recent experiments [21], which find correlations for some liquid temperatures, but not for the others, suggest that the situation is more complex.

In this work, we use the approach presented in [5] to calculate the number of photons emitted for our experimental conditions. These results are also given in Fig. 4a; the experimental values for R_0 are used, and the parameters applicable to pure argon bubbles are assumed. Although the theory predicts a slightly faster increase of the number of photons than in the experiment, good qualitative agreement is found.

Another interesting question is the change of the SL emission as P_0 is varied, but the ratio P_a/P_0 is kept fixed. Figure 4b shows that both the experimental and theoretical results increase approximately linearly as P_0 is increased. This observation is consistent with the theoretically proposed increase of the expansion ratio as P_0 is increased, but P_a/P_0 kept constant [22].

Finally, we address the question of a possible increase of maximum SL emission as P_0 is varied. Since the SL intensity increases as P_0 is decreased and P_a kept constant (Fig. 4a), it appears that by decreasing P_0 one could obtain a stronger pulse. However, in the experiment we find that, if P_a is kept constant, and P_0 is decreased, for some critical value of $P_0 = P_0^{\rm crit}$, the bubble becomes unstable and disappears. We find that $P_0^{\rm crit} = P_0^{\rm crit}(P_a)$; for lower P_a it is possible to go to lower values of P_0, without inducing instability. Figure 4c shows the results for the number of photons emitted, for three different values of P_a, as P_0 is varied. For all values of P_a, we find that the maximum number of photons is approximately the same. It appears that, at least in the probed parameter range, a possible increase of the maximum SL output by changing P_0 is prevented by the limitations imposed by the stability requirements.

The intensity of steady-state SL emission is limited by three requirements: (1) that a bubble be stable with respect to the mass flow, (2) that it be stable with respect to shape instabilities [18,23], and (3) that its position in the liquid not change with time [24]. The instability due to mass flow is characterized by a rather slow process of bubble growth, possible breakup, and/or production of microbubbles [9]. We do not observe breakup of the bubble; as P_0 is decreased below a critical value, the bubble typically drifts away from the pressure antinode. If P_0 is further decreased, the bubble disappears. We conjecture that it is the combination of the requirements (2) and (3) above which limits the SL intensity. More detailed experimental results and theoretical analysis of this effect will be given elsewhere [25].

In conclusion, we report the first systematic measurements of the modifications of bubble dynamics and SL emission as the ambient pressure is varied. A strong increase of the number of photons emitted in a SL pulse is observed as the ambient pressure is reduced. Next, in agreement with the dissociation hypothesis, we observe that the equilibrium bubble radius increases as the ambient pressure is decreased. Further work in this direction will allow for even more precise comparison of our experimental results with theoretical predictions. In particular, our experiments will serve as an important test for a variety of SL theories.

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