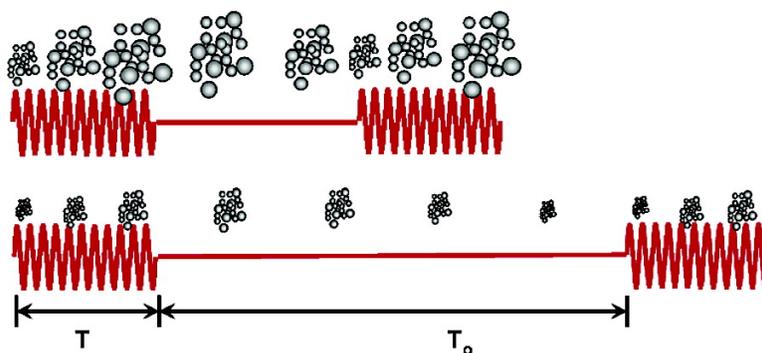


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## Determination of the Size Distribution of Sonoluminescence Bubbles in a Pulsed Acoustic Field

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The size of acoustic cavitation bubbles is difficult to measure due to the chaotic nature of acoustic cavitation. Various techniques, such as laser light diffraction,<sup>1</sup> active cavitation detection,<sup>2</sup> and phase-Doppler techniques,<sup>1,3</sup> have been employed in the past as a means of estimating the size of cavitation bubbles. Here we introduce a new method for determining the size of sonoluminescence bubbles in an aqueous medium.

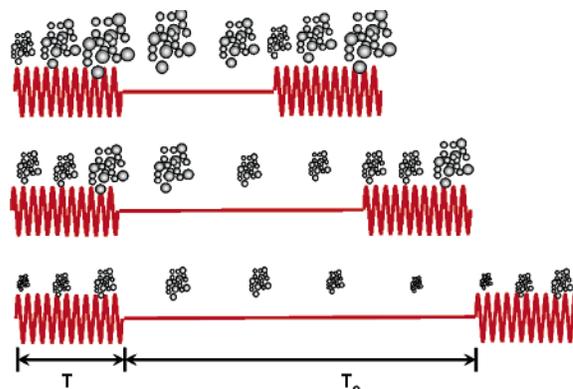
The experimental setup used for the pulsed sonoluminescence (SL) intensity measurements has been described in detail elsewhere.<sup>4,5</sup> Henglein et al.<sup>6</sup> have also studied the chemiluminescence from aqueous luminol solutions using pulsed ultrasound. The maximum SL intensity in water and SDS solutions as a function of the pulse off-time ( $T_o$ —the duration between the acoustic pulses) was measured for a fixed pulse on-time ( $T$ ) of 4 ms.<sup>4</sup> A frequency of 515 kHz and a sample volume of 50 mL were used. The power absorbed by the insonated solutions was 0.6 W/cm<sup>3</sup>, as measured by calorimetry.

It is known that the sonication of aqueous solutions, under certain conditions, generates cavitation bubbles that may violently collapse (active bubbles) to emit light (SL). In pulsed ultrasound experiments, we reported<sup>4</sup> that a “steady-state active bubble population” could be reached within a 4 ms ultrasound pulse at 515 kHz when a solution was exposed to a “train” of 4 ms acoustic pulses. During the pulse off-time, the bubbles can be expected to dissolve (or can be expelled from the liquid by buoyancy, if large enough). However, if the successive acoustic pulses can act upon these bubbles before they have time to dissolve, they can be grown again to be in the “active size” range. Thus, by varying  $T_o$ , the extent of dissolution of these bubbles can be controlled. This scenario has been depicted in Figure 1.

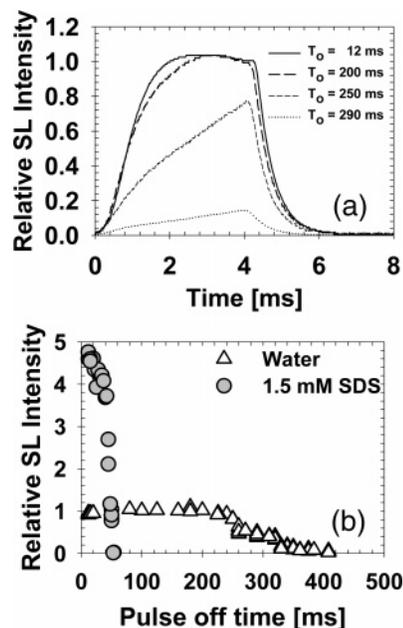
It follows that with increasing  $T_o$  a critical time will be reached, above which all the bubbles will have dissolved between the acoustic pulses.<sup>7</sup> As a consequence of the decrease in the size and population of cavitation nuclei, the SL intensity would be greatly diminished.

Figure 2a shows the SL pulse shapes in water at different  $T_o$ . The individual pulse is averaged over approximately 250 pulses and normalized relative to the SL intensity at  $T_o = 12$  ms. It shows that as  $T_o$  increases, the SL intensity decreases. The temporal profile of the SL pulses also indicates that a steady-state active bubble population has been reached within the 4 ms pulses for  $T_o = 12$  and 200 ms. This is not the case for  $T_o = 250$  ms and 290 ms and indicates that the rate at which the steady-state active bubble population reaches steady state strongly depends on  $T_o$ .

A plot of the maximum SL intensities as a function of  $T_o$  is shown in Figure 2b for water. It can be seen that the maximum SL signal initially remains constant until a  $T_o$  of about 200 ms, and there after it begins to decrease until the SL signal becomes undetectable



**Figure 1.** The effect of increasing the duration between the ultrasound pulses ( $T_o$ ) while keeping the pulse length ( $T$ ) fixed on the bubble size. As  $T_o$  increases, the bubbles generated are allowed to dissolve away, and this decreases the number of bubble nuclei available for cavitation in the successive pulse.



**Figure 2.** (a) Individual sonoluminescence pulse shapes in water averaged over about 250 pulses for various duration between pulses ( $T_o$ ). (b) Relative sonoluminescence intensity as a function of pulse off-time for water and 1.5 mM SDS. For both (a) and (b), the frequency, pulse width, and power absorbed were fixed at 515 kHz, 4 ms, and 0.6 W/cm<sup>3</sup>, respectively. The SL is normalized to the SL at pulse off-time of 12 ms.

at a time of about 408 ms. This decrease in the SL intensity with increasing  $T_o$  can be interpreted as the decrease in the size of cavitation bubbles due to the dissolution of bubbles during the pulse

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**Table 1.** Radii of SL Bubbles Generated in Water, Measured Using Different Techniques: The Theoretically Calculated Resonance Radii Are Also Included

technique	frequency (kHz)	theoretical resonance <sup>a</sup> $R_r$ ( $\mu\text{m}$ )	experimental $R_o$ ( $\mu\text{m}$ )
SL (this study)	515	5.8	2.8–3.7
active cavitation <sup>2</sup>	1100	2.7	0.9, 1.38
laser diffraction <sup>1</sup>	20	150	3.8
phase-doppler <sup>1,3</sup>	20	150	5.0

<sup>a</sup> Calculated using  $R_r F_r = 3$ .

off-time. Each increment in  $T_o$  above 200 ms would correspond to the total dissolution time<sup>7</sup> of a group of bubbles of a particular size.

The total dissolution time of the bubbles can be used to calculate the size of the SL bubbles quantitatively. Assuming that the bubbles are dissolving as if they were individual bubbles, the initial bubble size ( $R_o$ ) can be estimated using a simple equation developed by Epstein and Plesset<sup>8</sup> for a single dissolving stationary bubble.

$$\left(\frac{DC_s}{\rho_g R_o^2}\right)t = \frac{1}{3} \left(\frac{RT\rho_g R_o}{2M\gamma} + 1\right) \quad (1)$$

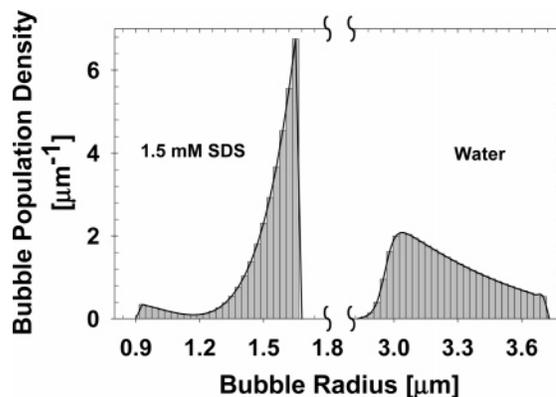
$D$  is the diffusion coefficient;  $C_s$  is the saturated dissolved gas concentration;  $\rho_g$  is the gas density in the bubble;  $R_o$  is the initial bubble radius;  $t$  is the total dissolution time;  $M$  is the molecular weight of the gas;  $\gamma$  is the surface tension of the liquid;  $R$  is the universal gas constant, and  $T$  is the temperature of the liquid. Thus, by replacing  $t$  with  $T_o$ , the  $R_o$  can be calculated.

The calculated radii of the SL bubbles in water are tabulated in Table 1 along with other bubble sizes determined using different techniques reported in the literature. The radii obtained for water are within the correct order of magnitude for a 515 kHz frequency, that is, at this frequency, the linear resonance radius ( $R_r$ ) is 5.8  $\mu\text{m}$  (see Table 1). The experimental bubble radii obtained are somewhat lower than the calculated linear resonance radius. It is known that, owing to the nonlinearity, the actual resonance radii of cavitation bubbles are smaller than the  $R_r$  calculated using the linear equation. This discrepancy with the theoretical radii has been reported in the past using other methods and is also shown in Table 1. It should be noted that the resonance size of the cavitation bubbles has a range rather than a single value for a given frequency,<sup>9</sup> which is what our experimental data suggest.

The data presented in Figure 2b can also be used to calculate bubble population density as a function of different bubble radii. Assuming that the SL intensity decreases linearly with a decrease in bubble population, the bubble population density could be estimated (Figure 3).

It has been qualitatively suggested in previous studies that the presence of small quantities of SDS in water leads to a decrease in the cavitation bubble size as well as a narrower bubble size distribution.<sup>10,11</sup>

The SL data shown in Figure 2b for 1.5 mM SDS clearly show that the bubbles dissolve within about 50 ms and the steepness of the drop in the SL intensity suggests that these bubbles have a narrower size distribution compared to water. For bubbles in the



**Figure 3.** The bubble population density versus bubble radius for water and 1.5 mM SDS, estimated using eq 1 for every bubble interval of 0.03  $\mu\text{m}$ . A sigmoid equation with five parameters was used to fit the decay of the SL intensity for water at  $T_o$  greater than 200 ms, giving a  $R^2$  value of 0.93. For 1.5 mM SDS, a cubic fit giving a  $R^2$  of 0.97 was used. These fits were used to calculate the bubble population density for every 0.03 mm increment in the bubble radius. Equilibrium surface tension values of 72 and 62 mN/m were used for water and 1.5 mM SDS, respectively.

presence of 1.5 mM SDS solution, the calculated bubble radii are in the range of 0.9–1.7  $\mu\text{m}$  (Figure 3).

In summary, it has been shown how a simple pulsed SL technique can be employed for determining the size distribution of SL bubbles in aqueous solutions.

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