Sensitivity of Sonoluminescence to Experimental Parameters

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Light-scattering measurements have enabled us to determine that the transition to sonoluminescence is characterized by a bifurcation in the dynamics of a trapped pulsating bubble. These experiments also reveal that in the sonoluminescence (SL) state, changes in bubble radius of only 20% are associated with factors of 200 in the intensity of emitted light. This sensitivity of SL suggests that it originates from the kind of singular behavior that arises from the implosion of a shock wave. Theoretical extrapolations of this model to energy scales for fusion are discussed.

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The radiation pressure of a resonant sound field in a liquid can trap a small gas bubble at a velocity node [1]. At a sufficiently high sound intensity the pulsations of the bubble are large enough to prevent its contents from dissolving in the surrounding liquid [2,3]. For an air bubble in water, a still further increase in intensity causes these pulsations to become so enormous and nonlinear that the supersonic [4] inward collapse of the bubble concentrates the acoustic energy by over 12 orders of magnitude [5] so as to emit picosecond flashes [6] of broadband light which extend well into the ultraviolet [7] and which furthermore are synchronous [8] with the sound field to picosecond accuracy.

We now use light scattering techniques to determine the dependence of the light emitting mechanism on the bubble dynamics. In particular we find that the transition to sonoluminescence (SL) involves a sudden decrease in the bubble's size. In the SL state changes in experimental parameters which vary the bubble radius by 20% cause a hundredfold increase in light emission. Measurements of the bubble's dynamic susceptibility suggest that while the parameter spacer for SL is sharply delineated, the establishment of a steady state involves long time scales on the order of seconds. Our calculations suggest that this extremely sensitive dependence of sonoluminescence on bubble dynamics originates from the singularity which forms when a shock wave implodes [9]. Idealized theoretical extrapolations indicate that as the shock radius passes through 60 Å the temperatures and densities are high enough for fusion.

The extreme sensitivity of SL to external parameters such as the water temperature and the sound field amplitude, is indicated in Fig. 1 which shows that, as the water temperature decreases from 40 °C to 1 °C, the intensity of the light emission increases by a factor of over 200. (At 1 °C the purple light emitted by the bubble is so bright that it can be seen by the unaided eye even in the presence of external lighting [10], but at 40 °C the SL is barely visible even in a darkened room.) Since an understanding of this remarkable sensitivity would provide insight into the as yet unexplained mechanism of light emission (as well as providing a critical test of theoretical models), we have employed a recently described light-scattering technique [4] to determine the corresponding temperature dependence of the parameters which characterize the highly nonlinear pulsations of the bubble. These include its maximum radius R_m, the dynamic acoustic pressure amplitude P_a at the location of the bubble, and the ambient radius R_0 (when its contents are at 1 atm). According to our results, which are shown in Fig. 1, the more than hundredfold increase in light intensity is accompanied by changes of only 10%–20% in the key physical quantities that describe the motion of the trapped bubble of air.

Even when the fluid temperature is fixed the intensity of SL is a rapidly increasing function of P_a until, as shown in Fig. 2, an upper threshold is reached. With the goal of understanding the dynamical effects that limit the extent to which sound can be converted into light, we have displayed in Fig. 2 our measurements of radius versus time for a trapped bubble as a function of a slowly increasing drive level. Visible in Fig. 2 are the following

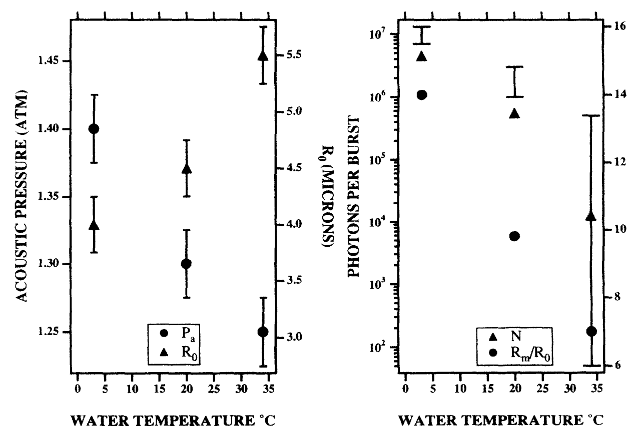

FIG. 1. Values of the intensity of sonoluminescence, sound field level P_a, maximum bubble radius R_m, and ambient radius R_0 as a function of water temperature for a trapped bubble of air. The number $\mathcal{N}$ of photons per burst (with wavelength greater than 200 nm) is measured in each case near the maximum achievable value. The bars are the ranges of intensities calculated from the shock wave theory when the uncertainty in the experimental input parameters is allowed for.

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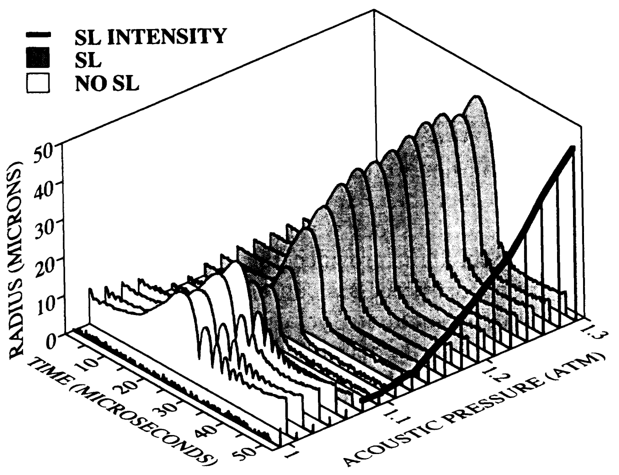

FIG. 2. Bubble radius versus time for about one cycle of the imposed sound field as a function of increasing drive level. The shaded area represents the light emitting region. The relative intensity of emitted light as a function of drive level is indicated by the solid line ramp. For the unshaded region, the bubble is trapped but no light is emitted. At drive levels below the unshaded region the bubble dissolves over a long time (~1 s). The lowest amplitude sweep (no bubble present) indicates the noise level.

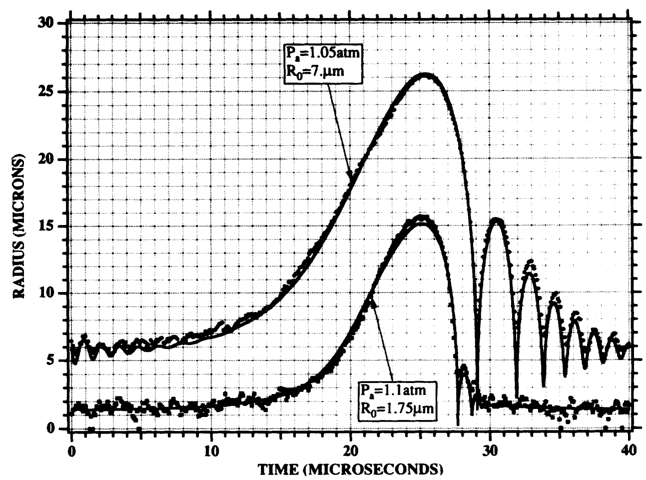

FIG. 3. Plot of radius versus time for bubble motion just above and below the threshold for the onset of sonoluminescence. The data have been matched to the Rayleigh-Plesset equation using the procedure discussed in Refs. [3] and [9]. The frequency of the resonant sound field is 26.4 kHz and its quality factor is about 1000.

regimes: (a) at the lowest amplitudes shown, the sound field can trap the bubble but the oscillations are not sufficiently violent to make light; (b) as the amplitude is increased the bubble abruptly becomes significantly smaller while the collapse ratio R_m/R_0 becomes significantly larger; (c) a still further increase in amplitude leads to a more violent collapse and a stronger light emission until an upper threshold is reached, above which it is impossible to maintain a stable bubble of any radius. The data for Fig. 1 were taken at the top of the SL regime where the number $\mathcal{N}$ of photons per flash is nearly its maximum.

The actual establishment of the light emitting mechanism is also extremely sensitive to small changes in P_a. A detailed comparison of the radius versus time curves just above and just below the abrupt transition between the regimes (a) and (b) is shown in Fig. 3. It can be seen that, as the acoustic drive is increased by a few percent, the bubble adopts a new steady state with a significantly smaller ambient radius; also, the collapse (first studied by Rayleigh [11]) becomes more violent, as is evidenced by a lessening of the afterbounces. The low drive side of the threshold is characterized by an easily noticeable jiggling or "dancing" of the bubble's position through a few multiples of R_m [12], whereas on the SL side of the threshold the bubble is extremely stable. With other liquids, such as low viscosity silicon oil, we have been able to trap a bubble in the non-SL regime but have been unable to achieve the transition shown in Fig. 3. An understanding of this threshold will extend to liquids other than water, the parameter space in which SL can be observed. In view of the sensitivity of SL to small parametric variations there are sure to be some remarkable surprises ahead.

Although the threshold for establishing SL is sharp and

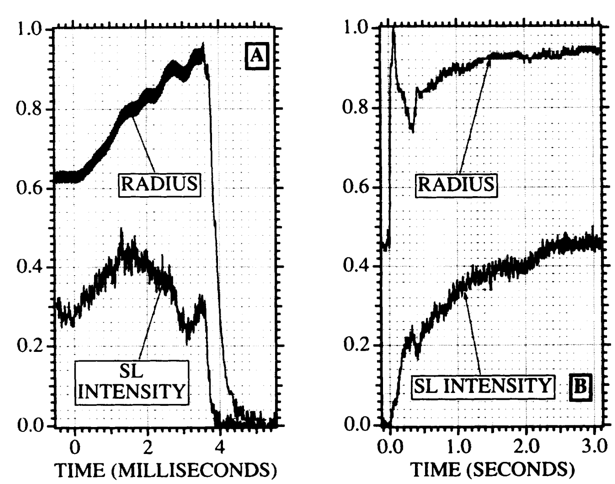

FIG. 4. Dynamic response of the intensity of sonoluminescence and of the maximum bubble radius to a sudden change in drive level. In (a), the drive is boosted above the upper threshold and, after becoming brighter for a short time, the bubble disappears. In (b), the drive is boosted from weak to strong sonoluminescence. After "gagging" [which can also be seen in (a)] the bubble achieves a new steady state during a long time scale measured in seconds. The signal has been averaged with a time constant of 100 μs for (a) and 10 ms for (b). Strictly speaking the results labeled "radius" signal are proportional to the product of R_m and the duration of the expansion.

well defined, the time required for the bubble to reach the SL steady state is very long, typically on the order of 10^5 cycles of the imposed sound field. Displayed in Fig. 4(b) is the response of a bubble to a sudden jump in the sound intensity which takes it from a state of low (or zero) SL to one of high SL. In this case the short term response is followed by a long time response on the scale of seconds during which the bubble seeks a steady state characteristic of a more violent collapse. One physical process with this long time scale is mass diffusion for which a typical time is $t_d = \rho_g R^2/DC_0$, where ρ_g is the gas density, R is a typical bubble radius, $D = 2\times 10^{-9}$ m^2/s is the coefficient of mass diffusion for air in water, and C_0 is the saturated concentration of air in water in the close vicinity of the ambient bubble. Because of the possible importance of mass diffusion, the data for Fig. 1 were taken at similar concentrations, in the range of 5%–10% of saturation for dissolved air in water. The dynamical method for determining the concentration has been described elsewhere [3]. That the water should be degassed was an important aspect of the discovery of single bubble SL [12]. The transient response of the bubble to a sudden increase in P_a at the upper threshold is shown in Fig. 4(a). In this case the bubble disappears on the (faster) time scale required for the sound field to exceed the upper threshold appreciably, but not before an initial increase in the maximum radius and SL intensity is seen.

In addition to challenges posed by the above data, theoretical attempts to interpret SL will also be motivated by the observation [4] that the collapsing wall of the bubble attains a supersonic velocity just prior to light emission. Thus it has been suggested [9] that SL is due to thermal bremsstrahlung emitted from a plasma generated by an imploding spherical shock wave. As shown in Fig. 1 the shock wave theory also yields SL intensities that are extremely sensitive to the drive parameters and furthermore the calculated values are in reasonable agreement with experiment. Since this model [9] does not allow for mass exchange of air between the interior of the bubble and its surroundings, it cannot determine R_0 and cannot explain the two thresholds described above. For this reason a comparison of this model to the experimentally observed SL intensity requires the use of data for R_0 and P_a as obtained from our light scattering measurements. The range of values attached to the theoretical results shown in Fig. 1 arises not from the model but from uncertainties of roughly 0.025 atm for P_a and 0.25 μm for R_0 in the experimental input parameters. The integrated spectrum for this model is proportional to $\lambda^{-1.5}$ where λ is the wavelength of the emitted light. This is a somewhat weaker dependence on λ than that observed, but corrections to the formula for the spectrum of bremsstrahlung, due to the fact that the light is emitted from a region smaller than the wavelength of light, may explain this. Furthermore the flash widths of 150 ps, as determined by this model [9], are reasonably consistent with experiment.

In another model of SL [13], light emission arises from the change in zero point energy as a hole is filled in by a medium with a different dielectric constant; the emission is due to the entire transit from R_m to the collapse. The first approximation to this model yields a spectrum that is somewhat steeper than experiment.

According to the shock wave model the temperature increases without limit provided the shock remains spherical and transport processes can be neglected. For example, a simple calculation for an ideal gas indicates that the shock attains a temperature of 3×10^8 K when its radius returns through 10 Å just after the moment of focusing. To see this we note that, in the limit of large Mach numbers, a self-similar solution of Euler's equations can be found [14] in which the radius of the shock is

$$R_s = At^\alpha , \tag{1}$$

where t is the time to the moment of focusing and α depends upon the equation of state (for air $\alpha \approx 0.7$). The determination of A requires knowledge of the launch conditions which we take to be that the shock is moving at Mach 1 (relative to c_0, the ambient sound velocity in the gas) when the bubble is collapsing through its ambient radius R_0 [3]. In fact energy conservation implies that the collapse rate of the bubble obeys [10]

$$\frac{\dot{R}(R_0)}{c_0} \approx \left[\frac{2}{3\gamma} \frac{\rho_0}{\rho} \left(\frac{R_m}{R_0} \right)^3 \right]^{1/2} = O(1) ,$$

where γ is the heat capacity ratio and ρ_0/ρ is the ratio of densities of the ambient gas and the liquid. Applying the launch conditions to (1), we see that

$$M = \dot{R}_s/c_0 = (t_0/t)^{1-\alpha}, \quad R_s = R_0 (t/t_0)^\alpha , \tag{2}$$

where t_0 is the time that elapses between the moment when the bubble radius is R_0 and the instant when the shock focuses,

$$t_0 = \alpha R_0/c_0 . \tag{3}$$

The temperature jump across a strong shock is proportional to the square of the Mach number [14]. But after reflection from the origin the outgoing shock moves into the gas previously heated by the incoming shock, and the increase in temperature after focusing is approximately given by $T/T_0 = M^4$, where $T_0 \approx 300$ K is the ambient temperature. In this way one verifies that, 0.1 ps after focusing, $M \approx 30$, $R_s \approx 10$ Å $= R_n$ (say), and $T = 3\times 10^8$ K. Assuming that these estimates would apply to a gas bubble which contains a mixture of deuterium and tritium, and that they remain physically valid at such minute length and time scales, the repetitive SL implosions generate about 1 n/s in such a mixture. Changes in the equation of state that these temperatures would bring about have also been ignored in making these estimates.

The neutron emission, N, was obtained from the standard formula [15]:

$$N = n^2 \overline{\sigma v} R_n^3 \Delta t_n / \tau_a , \qquad (4)$$

where n is the number density of atoms ($\approx 10^{23}/\mathrm{cm}^3$ at focusing), R_n is the radius of the hot region, Δt_n is the length of time during which the temperature is high enough for significant fusion to occur, τ_a is the acoustic period which determines the number of implosions per second, σ is the reaction cross section, and v is the relative velocity of reacting nuclei; the overbar indicates the statistical average of σv:

$$\overline{\sigma v} = 4\times 10^{-12}(\tilde{T}^{-2/3})\exp(-20\tilde{T}^{-1/3})\mathrm{cm}^3/\mathrm{s} , \qquad (5)$$

where $\tilde{T} = T/1.16\times 10^7$ K. The maximum value of (4) occurs when $\tilde{T}\approx 10$; this motivated our choice of T in the preceding estimates. Calculations [12] for the case $P_a = 1.425$ atm, $,_0 = 4$ μm, $1/\tau_a = 25$ kHz show that, for an air bubble modeled as a van der Waals gas (for which $\alpha \approx 0.5$), a temperature of 10^8 K is attained at a distance of $R = 60$ Å from the center of the bubble and lasts for a time of order 0.1 ps. This computation yields about 40 n/s, but the results are very sensitive to the launch conditions, in part because α depends strongly on these conditions in a van der Waals gas [16]. For instance, at $P_a = 1.375$ atm, the computed yield is less by a factor of 10. These computations neglect the emission of an outgoing shock by the bubble surface and the fusion rate given by (4) and (5) may have to be modified for a dense system where binary collisions may not make the dominant contribution to N. A parallel exists between the shock wave model of SL and efforts aimed at developing inertial confinement fusion. In each case the level of energy concentration which can be attained is limited by the stability of an imploding shock wave.

The sensitivity of sonoluminescence to small changes in bubble parameters indicates that this phenomenon is linked to a kind of singular behavior such as occurs when a spherical shock wave implodes. If this is the case, the robust synchronicity displayed by SL is that much more amazing. Although sensitivity to input parameters is an essential aspect to the theory of SL, the upper energy at which the singularity remains intact can only be determined by further experimentation.

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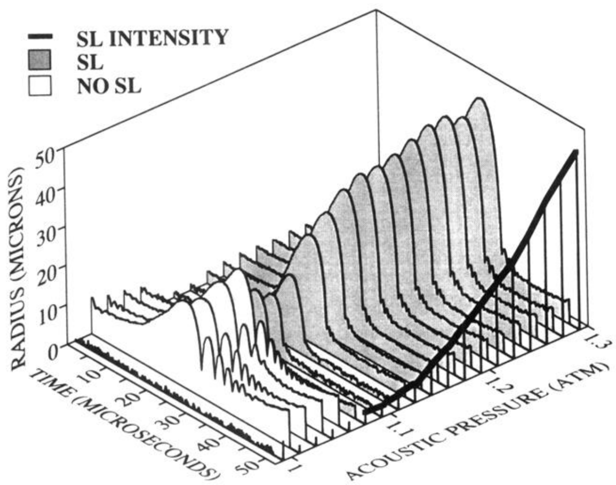

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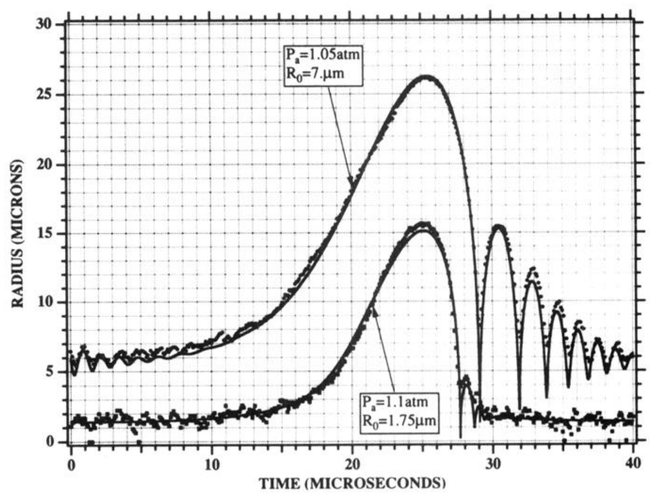

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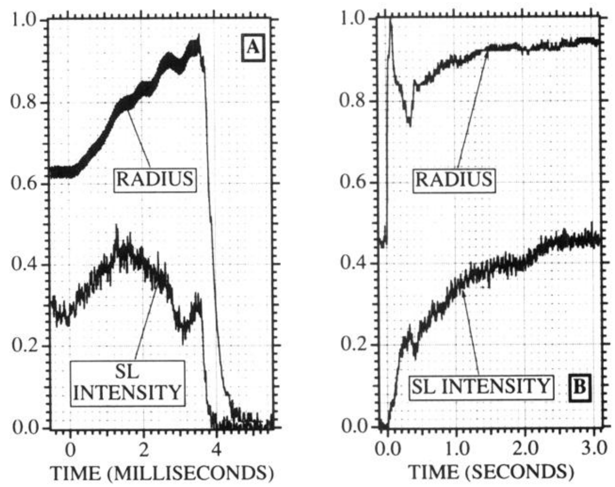

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