Pulsed Mie Scattering Measurements of the Collapse of a Sonoluminescing Bubble

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Measurements of light scattered off a bubble that is illuminated by a train of short pulses enables one to resolve the strongly supersonic collapse of a sonoluminescing bubble. We find that the collapse is faster than Mach 4 (relative to the ambient speed of sound of the gas in the bubble) and that the flash of sonoluminescence is emitted within 500 ps of the minimum bubble radius, at about which time the bubble's acceleration is greater than 10^{11} g. [S0031-9007(97)02516-7]

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Figure 1 shows the relative timing of a simultaneous measurement of three key processes which characterize sonoluminescence (SL), the transduction of sound into light by the high amplitude pulsations of a gas bubble in a fluid. Displayed are: (*a*) the intensity of laser light scattered into a photodetector by the bubble [1], (*b*) the signal recorded on a hydrophone 1 mm from the bubble [2], and (*c*) the response of a photomultiplier tube (PMT) to the flash of light emitted by the bubble [3]. To the accuracy of this measurement, the acoustic spike and SL flash are delta functions in time and the slope $\dot{R}$ of the bubble's radius is vertical as the moment of light emission is approached.

Experimentalists are faced with the crucial challenges of obtaining better temporal resolution of these effects and determining the spectrum of SL [4,5] beyond 6 eV photons, which is the cutoff imposed by water. To date the upper bound on the width of the SL flash is 50 ps [3], the bandwidth [see Fig. 2(A)] of the outgoing acoustic spike launched by the high compression in the bubble is 30 MHz [2] (or 10 ns rise time), and it arrives at the hydrophone approximately 1 μs after the SL flash (which is about the time required for sound to propagate 1 mm in

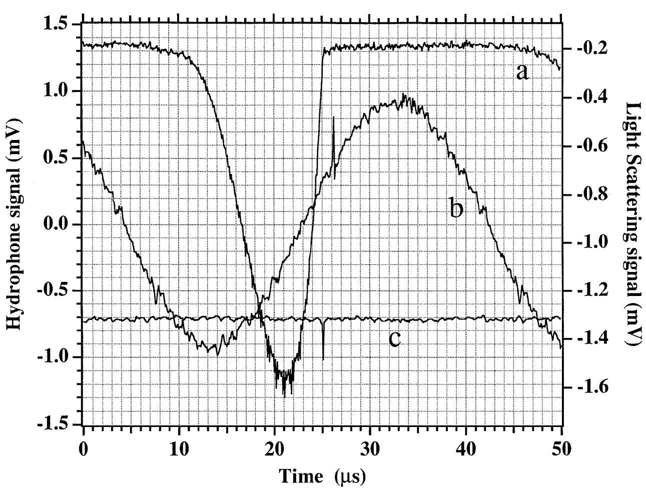

FIG. 1. Relative timing of the PMT response to sonoluminescence (*c*) from the stressed interior of a collapsing air bubble whose radius squared is proportional to the magnitude of the intensity of scattered laser light (*a*). The high pressures reached during the collapse launch an outgoing spike riding on the driving sound field (*b*) recorded by a needle microphone (Precision Acoustics) about 1 mm from the bubble. The scale for SL has been offset and the phase of the 26 kHz sound wave has been shifted by 3 μs to correct for the phase delay introduced by the ac-coupled preamplifier.

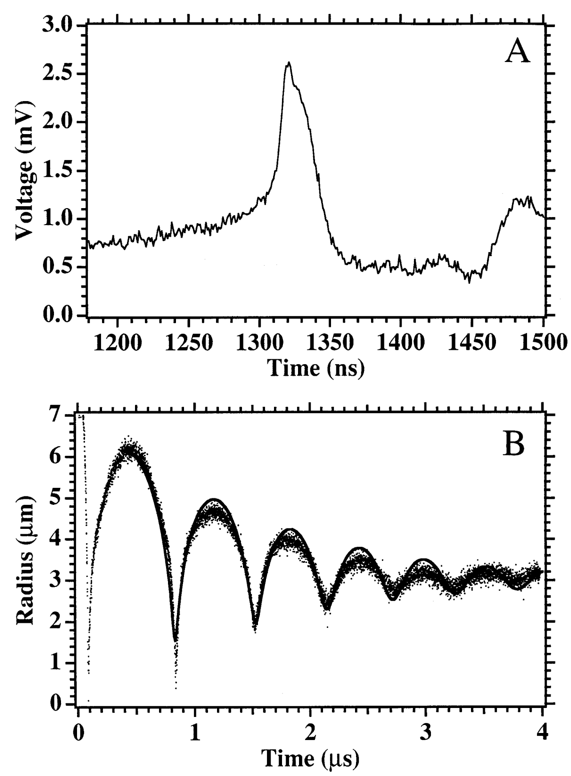

FIG. 2. (A) Detail of trace (*b*) in Fig. 1 showing the instrument limited rise time of 10 ns for the outgoing acoustic spike. According to the calibration of the hydrophone (5 mV/MPa at 3.5 MHz), the signal at 1 mm from the bubble is about 3 atm. (B) Comparison of the light scattering signal and a fit to the Rayleigh-Plesset (RP) equation for the afterbounces of a 150 torr 1% argon in oxygen bubble using parameters $f_A = 40$ kHz, $R_0 = 4.0$ μm, and $P_a = 1.45$ atm. To account for impurities in the water we interpret the viscosity as an effective damping and take its value as 0.03 g cm/s (three times the tabulated viscosity of pure water), and for surface tension the value 50 dyn/cm is taken. Details of the procedure whereby data is fit to the RP equation are given in Ref. [2].

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water). For the speed of collapse of the bubble, previous work found about Mach 1 (relative to ambient gas at 1 atm) as a lower bound [1] and so suggests that the collapsing bubble launches an imploding shock wave [6,7] which further focuses the acoustic energy to such an extent that uv light is emitted [4,5]. To probe this model we have used femtosecond Mie scattering to measure more accurately the bubble dynamics near the moment of light emission. We find that the bubble (as theoretically predicted [6]) is collapsing at speeds higher than Mach 4, and that the flash of light is emitted within 500 ps of the moment of minimum radius which is determined by the van der Waals hard core of the gas molecules. At that moment the measured acceleration $\ddot{R}$ is greater than 10^{11} g.

Light scattered by a bubble into a large solid angle is approximately proportional to the square of its radius [1,8]. Fast PMTs with the requisite dynamic range respond to an infinitesimally short light pulse typically with a rise time of 1–3 ns and so convolve values of R on this time scale. The huge changes in R that accompany SL exacerbate this difficulty. The resulting distortion of the measured time dependence of the radius can on these fast time scales affect the relative timing of the minimum radius relative to SL [1,2]. This experiment overcomes the limitations of finite time response detectors by using 200 fs pulses of laser light scattering off the bubble at a precisely timed instant. As before, a PMT is used to collect the scattered light, but now its integrated (slow) response can be ascribed to scattered light at the instant of the laser pulse. Since the repetition rate of the laser ($\sim$13 ns) and the acoustic frequency are incommensurate the time of the laser pulse with respect to the SL flash (which sets the zero of time for every acquisition) wanders through all possible values and in the course of an experiment maps out a complete radius vs time curve.

These measurements were performed on a gas bubble levitated in a sealed, spherical quartz flask filled with water and acoustically driven at its breathing resonance [3,9]. Light produced from a pulsed Ti:Sa laser (Coherent Mira 900) illuminated the bubble. The output of this laser after passing through a frequency doubling nonlinear crystal and an acousto-optic modulator resulted in a steady stream of blue (410 nm) pulses each 200 fs in length repeating at about 76 MHz with an average power of around 26 mW (0.3 nJ per pulse) striking the bubble. The light scattered by the bubble out of the beam is collected around 60° from the forward by a 2 in diam lens. This light passes through a laser line pass filter and an aperture on its way to a PMT (Hamamatsu R-580). The output of the tube (shown in Fig. 3) is acquired in real time on a digital oscilloscope (HP 54542A). The oscilloscope is triggered by another PMT with appropriate filters to respond to the flash of SL and not the laser light. This trigger which sets the zero of time for all acquisitions has been shown to be good to 50 ps [3].

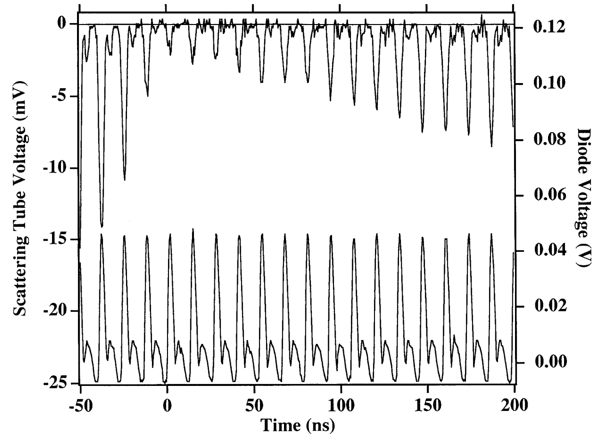

FIG. 3. Pulsed light scattering from a collapsing bubble as probed by 200 fs pulses of light with a repetition rate of 76 MHz [upper trace]. The scattering data is generated by a single sweep. The noise floor is obtained from 512 averages. Essential to this method is that the response of the detector return to the noise floor between pulses. In this way each peak can be ascribed to a single, precisely timed scattering event. Shown on the lower trace is the output of the photodiode that tracks the phase of the laser.

Two problems arise due to the fact that the scattered light signal is the smallest when the bubble radius is a minimum: (1) this small signal immediately follows the large signal generated at the maximum radius, and (2) this small signal is accompanied by a broad band flash of light which is the SL. The acousto-optic modulator allows the first to be overcome by deflecting the laser beam from the bubble during the time that the radius is near its maximum [1]. The beam is then switched to hit the bubble about 100 ns before the minimum radius is achieved; long enough to set up a stable response. The second problem is minimized by using laser flashes that are brighter than the SL flash (0.1–0.5 pJ per flash broad band into 4π) in combination with the narrow band laser line filter. In addition, the use of a polarizer between the bubble and the scattered light detector reduces the light from the unpolarized SL flash while leaving the polarized scattered signal unaffected.

For the purpose of averaging many traces the time of an integrated scattering event is ascribed to its peak. The precise time ($\tau = 13.23$ ns) between laser pulses can be used to synchronize a string of events with respect to the SL trigger more accurately than a single peak can be located. If $V(t_i)$ is the voltage of the ith 0.5 ns bin of the oscilloscope, then the value t_{phase} which extremizes

$$\sum_{n=0}^{\Delta t/\tau} V(t_{\text{phase}} + n\tau)$$

(where Δt is the record length) determines which of the 26 ($\approx 13.23/0.5$) possible values is the desired phase. Still more accuracy can be achieved by using this method to first determine the run independent phase (from electronic and optical delays) between the scattering event and the laser flash as detected by a photodiode sampling a tiny fraction of the beam (lower trace Fig. 3) and then adding in the run dependent phase between the diode and SL. Acquisition continued until each of the 26 phases had been averaged a minimum of 64 times. The computer acquired about thirty 500 point time records per second so that averaging required 3–5 minutes.

Comparison of the measured hydrodynamical motion of the bubble to the Rayleigh-Plesset equation [1,10] was used to calibrate the radial values. The acquisition showing the high detail around the minimum radius was immediately preceded by a run using 16 000 point time records which were acquired at the low rate of a few frames per second. Therefore this curve was averaged only about eight hits per bin, but that was sufficient to resolve the microsecond ringing motion after the collapse [as shown in Fig. 2(B)]. This data was fit to theory with the ringing period, decay envelope, and ratio of height of the first bounce to that ambient value constraining the fit. As this data was acquired with the same gain and optical alignment as the zoom on the minimum, the proportionality constant of this fit was used to calibrate both. These runs were bracketed by runs at lower PMT gain where the laser was on the entire acoustic cycle [1]. These curves were independently fit to hydrodynamics. The calibration was accepted only when all these fits agreed, which overall are trusted to about 10%. Final results generally ranged from 0.4–0.8 μm for the minimum radius and 1200–1600 m/s for the slope near the minimum.

As the observation angle and radius change, Mie theory predicts huge variations of the light scattered by a small sphere [8]. These diffraction fringes are smoothed into approximately an R^2 dependence of the scattered intensity by collecting light from a large solid angle [1]. Hence once a background is removed, taking the square root converts this data into values proportional to radius. Numerical calculation of the Mie theory for the polarization and color of light and angles of collection used here show for $2.5 < R < 4$ μm the observed radius is smaller than actual radius by 3%–5% and for $0.7 < R < 2.5$ μm the observed radius is larger than the actual one by 5%–10%. If corrections were made to account for this effect, the slope of the data near the minimum radius in Fig. 4 would *increase*.

The precise temporal resolution of the experiment allowed the timing of the SL flash with respect to the minimum radius to be measured. The two curves in Fig. 5 were generated by acquiring (i) the radius time curve as described above and (ii) the same signal with the laser turned off, the laser line filter removed and not integrating around the peak. Since (i) and (ii) were triggered by the SL flash, and since the optical and electronic propagation delays from the bubble to the oscilloscope were the same, the relative timing of the two curves is identical. The apparent width of the SL flash is due to the bandwidth of the PMT. As with the radial data, the time of the SL flash must be assigned to its peak voltage. So according

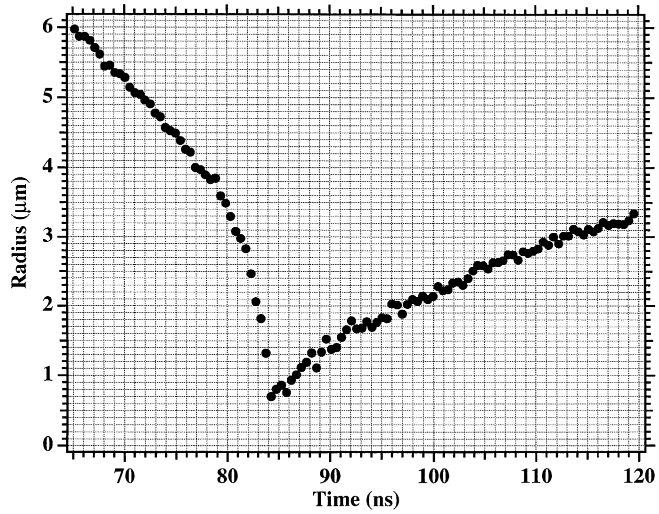

FIG. 4. Radius of a sonoluminescing bubble (1% argon in oxygen at 150 torr) as the moment of collapse is approached. These points are obtained by averaging together many traces of the type shown in Fig. 3, for various phases of the laser relative to the flashes of SL. According to this data the bubble is collapsing with a speed about 4 times the ambient speed of sound in the gas.

to Fig. 5 the emission of SL occurs ±500 ps of the minimum radius.

Using this technique it was also observed in some but not all of the data that the standard deviation in the detected scattered light at a given radius increased after the collapse [Fig. 6]. This effect was most often observed in larger bubbles (3–5 torr Xe and 150 torr 1% Xe doped diatomic gases), whereas air bubbles typically had the same size standard deviation at equivalent voltages on

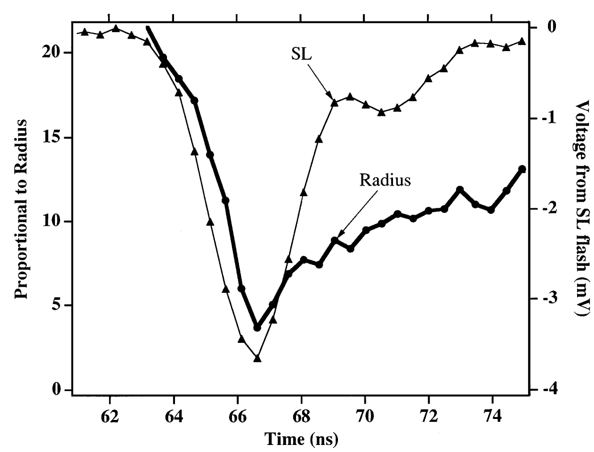

FIG. 5. Flash of SL and light scattering as resolved by the same photomultiplier tube. Each radius measurement is ascribed to that point in time when the response shown in Fig. 3 is a maximum. Thus for comparison the flash of SL must be ascribed to that point in time when the response to the flash is a maximum. As shown by this data the flash occurs within 500 ps of the minimum radius.

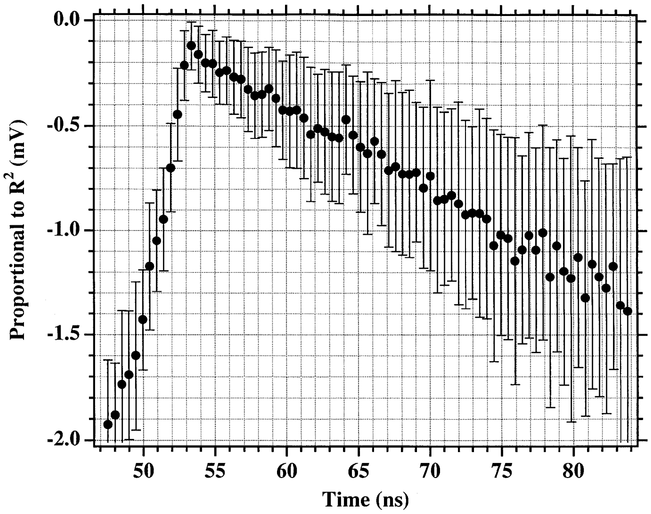

FIG. 6. Standard deviation in the intensity of light scattered from a bubble near the moment of light emission. The average value determines the radius, and the bar determines the standard deviation in a given 500 ps bin. (Note the standard deviation of the mean is down by about a factor of 8 from the plotted values.) The larger standard deviation after collapse can be interpreted as due to nonspherical bubble oscillations whose orientation relative to the laser varies from shot to shot. For some gases the standard deviation does not increase as a result of the collapse. This data was taken for 1% xenon in oxygen at 150 torr.

both sides of the minimum. This variation can be interpreted in terms of an implosion generated nonsphericity. As the decay time for surface oscillations is about 1 μs they damp out before the next crash [11] so as to allow the dynamics to be highly repetitive, as is observed.

Through use of pulsed Mie scattering we have resolved that sonoluminescence is emitted within 0.5 ns of the minimum radius and that just prior to this moment the bubble is collapsing faster than Mach 4 and accelerating over 10^{11} g. Taken together with measurements of photonic angular correlations [12] (which have been interpreted as indicating that the light emitting region is well inside the bubble) the current data provide the strongest evidence for the shock wave model of SL. It must be emphasized, however, that even if this model is correct its current formulation lacks predictive value, in that it fails to determine or explain the allowed ambient radii [13] or acoustic drive levels, and the effect of ambient temperature [4,14] or doping with a noble gas [5]. In any event, a direct observation of the imploding shock has yet to be carried out.

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