Angular correlations in sonoluminescence: Diagnostic for the sphericity of a collapsing bubble

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Deviations from isotropic emission in sonoluminescence (SL) have been resolved to about one part per thousand. States with larger dipole components are characterized by large fluctuations in the intensity of SL. When dipole components exceed the threshold of detectability their magnitude decays on a long time scale. This data can be interpreted as monitoring the degree of nonsphericity of the bubble collapse that leads to SL. [S1063-651X(96)50609-4]

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Among picosecond light sources sonoluminescence is unusual in that the flash intensities are uniform over a spherical shell [1]. Furthermore, theoretical [2] extrapolations of measurements [3] of the radius of the pulsating bubble that generates the sonoluminescence (SL) indicate that the radius of the light emitting region is smaller than the wavelength of the outgoing light. In order to characterize the details of this light source and possibly learn about the light emitting mechanism we have used multiple photodetectors to measure angular correlations in SL. Although we observe dipole components that reach 10% of the total emitted intensity they can be interpreted as being due to the refraction of light by a nonspherical bubble wall that separates the hot gas near the center of the bubble from the surrounding host liquid. According to this interpretation, angular correlations in SL provide a diagnostic for the sphericity of the bubble collapse. Since a more spherical collapse is more violent this diagnostic should prove useful to attempts to reach higher levels of energy concentration with sonoluminescence.

Angular dependence in the intensity of SL would be characterized by a nonzero value of the correlation,

$$\Delta Q_{AB}=\frac{1}{\overline{Q_A Q_B}}\langle(Q_A(i)-\overline{Q_A})(Q_B(i)-\overline{Q_B})\rangle_i \qquad (1)$$

as a function of the angle θ_{AB} formed by the detectors A and $B$and the bubble which is reckoned to sit at the vertex. In Eq. (1),$Q_A(i)$ is the total charge recorded in detector A on the ith flash, $\overline{Q_A}$ is the running average of $Q_A(i)$, and $\langle\ \rangle_i$ denotes an average over i. For detection we use standard photomultiplier tubes (Hamamatsu R1463-01) with a rise time of 2 ns. In addition to tubes A and B there is a trigger tube which monitors the SL so as to gate a digital oscilloscope (HP54542A) which then acquires the tube outputs in sequential mode. At an acquisition rate of 1 Gigasample/s 30 points are used to digitize each flash in each tube and $Q(i)$ is the area of the curve generated by these points. In sequential mode the oscilloscope ignores the long ($\sim 40\mu$s) dead time between events. So in this mode one can typically acquire 1,000 flashes ''on the fly'' during a time spanned by 25 000 cycles of the sound field (i.e., about 1 s). From this data set the average and the correlation (1) are calculated. Twenty such data sets generate a ''result'' and the average and standard deviation of three results generate the plotted point and ''error'' bar at each angle. As experimental configurations were varied the signal recorded in each phototube ranged from 5–8 photoelectrons per flash of SL.

Two types of resonators were used for these experiments: (i) two sizes of spherical quartz flasks (one with a free surface and the other sealed, with fundamental resonances at 26.4 and 40 KHz, respectively) [1,3] and (ii) various sealed cylindrical cells (with resonances at 33 and 23 KHz)[4,5]. For the sphere, data was taken when tubes A and B where located along either a latitude or a longitude. Use of a third detector C enabled us to simultaneously measure correlations along both the longitude and latitude. In each apparatus and for each set of control parameters the correlations were found to depend only on the angle between the detectors and not their absolute location relative to the laboratory or the resonator.

The solid line in Fig. 1 displays the angle dependent correlation that is observed in most runs. It can be attributed to a dipole component in the detected photon field. If $\overline{\Omega}_A$ is the fraction of solid angle subtended by detector A, and N_I, N_D are the numbers of photons arriving isotropically, and as a dipole then the total number of photons to strike the detector is

$$N_A(i)=\overline{\Omega}(N_I+3N_D\cos^2\theta_{iA}) \qquad (2)$$

(and similarly for B), where θ_{iA} is the angle between the direction of the dipole on flash i and the detector A. If one assumes that over time there is no preferred direction then a physical dipole of strength N_D leads to a correlation:

$$\Delta N_{AB}=\frac{\langle\{N_A(i)-\overline{N_A}\}\{N_B(i)-\overline{N_B}\}\rangle_i}{\overline{N_A N_B}}$$

which upon substitution of Eq. (2) yields

$$\Delta N_{AB}=\frac{{N_D}^2}{(N_I+N_D)^2}\left(\tfrac{1}{5}+\tfrac{3}{5}\cos 2\theta_{AB}\right). \qquad (3)$$

So according to the sine wave fit to the data in Fig. 1 this observed field has a dipolar component, N_D/N_I, of 7%.

In order to determine whether the dipole is due to the mechanism whereby the stress of implosion is converted into

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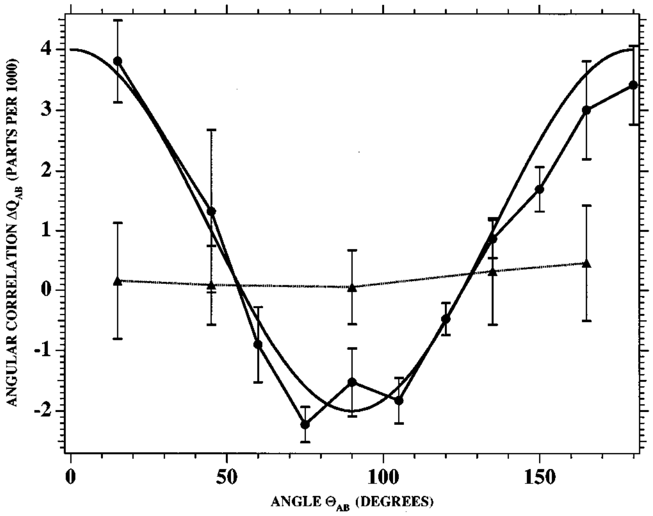

FIG. 1. Correlation of light intensity between two phototubes subtending an angle θ_{AB} with respect to a sonoluminescing bubble. The solid line corresponds to an SL bubble whose flash to flash intensity has a large variation. The flash to flash fluctuations for the dotted line are much less and are furthermore consistent with Poisson statistics. Note the appearance of a negative correlation at 90°. The maximum dipole that we have observed is about ten parts per thousand. If the dipole is due to refraction of light at the gas fluid interface of the bubble, then the ellipticity of the bubble in the state with large fluctuations is about 20%. The sine wave fit to the data is $(0.001)(1+3\cos 2\theta)$; and the least squares fit [not plotted] to the dotted data is $(0.001)(0.21+0.12\cos 2\theta)$.

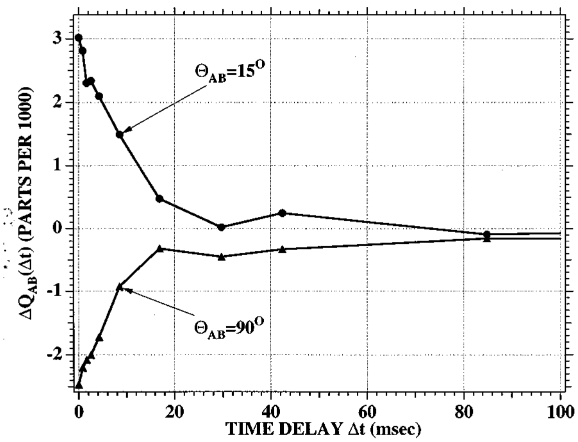

FIG. 2. Result of correlating SL flashes in tube A with flashes in tube B at a later time. The time delay $\Delta t = nT$, where n is an integer and T is the acoustic period. The long term memory of the dipolar component of emission is comparable to the lifetime of the driving sound field. The autocorrelation of a tube with itself $\Delta Q_{AA}(\Delta t)$ also decays to zero on this time scale. Typical quality factors for these sound fields range from 500 to 1000.

light we measured the correlation (1) as a function of a time delay Δt between acquisitions in tubes A and B. Since our method of data acquisition yields a labeled time sequence of events in each tube these dynamical correlations could be easily calculated. In the event that the dipole is due to the light emitting mechanism we would expect each flash to be independent of the previous flash so that the correlation should fall abruptly to zero for $\Delta t \neq 0$. As shown in Fig. 2, however, the dipole correlation $\Delta Q_{AB}(\Delta t)$ has a long memory which in fact is about four times the free decay time of the sound field.

The long time decay of the angle dependent correlation indicates that the dipole component is due to some aspect of the hydrodynamic motion. Such motions rearrange themselves on the same time scale for which the sound field changes. Various possibilities include (i) jitter in the location of the bubble, (ii) bending of the emitted light by the sound field in the bulk of the fluid, and (iii) refraction of the SL rays by the surface of a nonsperical bubble. Jitter in the location of the bubble would tend to increase the light in the detector towards which it is moving while decreasing the light in the other detector, and so cannot explain our observation of a positive correlation at 180°. The same is true of light bent by a $\cos\theta$ dependence in the resonant sound field. In addition, such an effect would at most yield a contribution to ΔN_{AB} that is proportional to M^2 (where $M \sim 10^{-5}$ is the Mach number of the bulk resonant sound field).

We propose that the measured angular correlation is due to the refraction, by the nonspherical bubble wall, of light that is emitted uniformly from a point source within the bubble's interior. Figure 3 shows how light from a point source would be refracted by passage through an elliptical boundary in the ray optics limit. For an interface where the index of refraction jumps from 1.0 (on the gas side of the bubble's surface) to 1.35 (in the water) a 7% dipolar component would require a 20% ellipticity. (We define ellipticity as $a/b - 1$, where a,b are the major, minor axes.) For demonstration purposes Fig. 3 was constructed for a jump in the index of refraction from 1.0 to 2.0 and a ratio of major axis to minor axis given by 2. According to our interpretation the measurements reported in this paper provide evidence that SL originates from the interior of the bubble as opposed to the surface of the bubble.

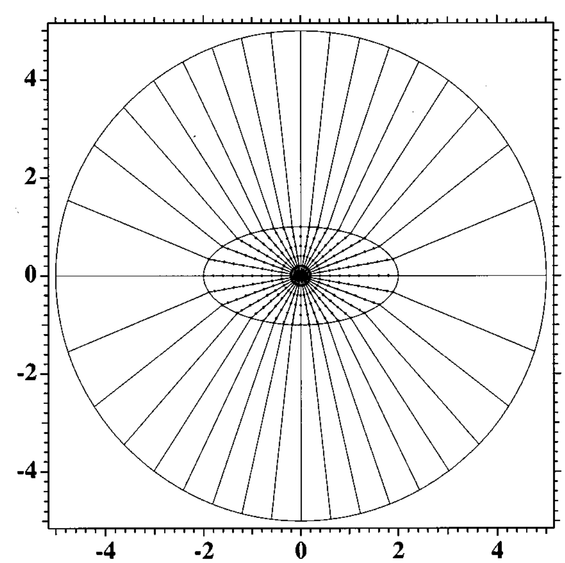

FIG. 3. Refraction of light from a point source by a surrounding elliptical interface. For purpose of demonstration this figure was generated with an ellipticity of 2 and a jump in the index of refraction from 1 to 2. Such a surface introduces a dipole component into the far field.

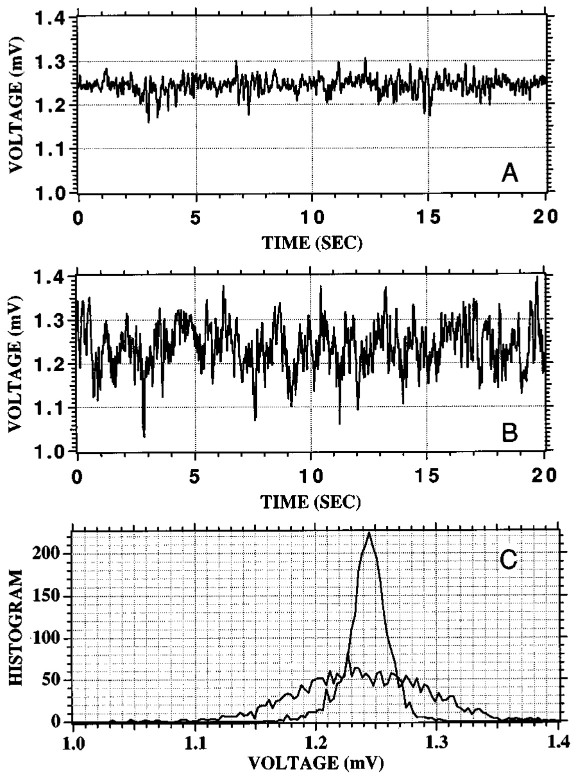

FIG. 4. Comparison of fluctuations in SL intensity for states with large *B* and small *A* dipolar emissions (as measured in the far field). The time records are displayed in (A) and (B) and the pulse height distributions are compared in (C). For this arrangement the driving sound field has a frequency of 26.4 KHz and the detectors each record an average of 5 photoelectrons per flash. The time constant for binning of the data is 10 ms.

Another insight into the origin of these correlations is provided by the observation that on some runs the dipole component vanishes, as shown by the dashed line in Fig. 1. According to Fig. 4 we see that these states are in one to one correspondence with narrow pulse height distributions. The time record and histogram of SL flash intensities indicates that the state with a 7% dipolar component (and ellipticity of 20%) has over an order of magnitude more spread in flash intensities. (We define the spread as the maximum divided by the width at half maximum). For the state with a narrow pulse height distribution the dipole component (as determined by a least squares fit) is 1%, which corresponds to an estimated upper bound on ellipticity of about 3%. Using three detector tubes we were also able to verify that the angular dependent and angular independent states occurred in the vertical and horizontal planes simultaneously. That is, when the longitude showed a large dipole so did the latitude, and similarly for the case of no dipole.

Control of the key parameters that determine whether the collapse is elliptical has been elusive as the system falls into and out of this state. Various candidates are imperfections in the sound field, such as coupling to nearby modes of the resonator, thermal drift, scattered sound biting back on the bubble, or dust particles in the vicinity of the bubble. Any hydrodynamic state that is coupled to the sound field will tend to wander on the time scale determined by its bandwidth (which is also the decay time of a transient). In this

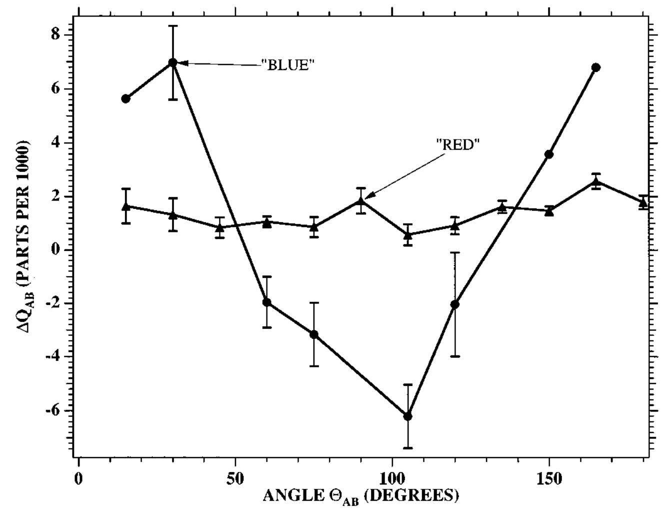

FIG. 5. Angular correlation for the long and short wavelength parts of the SL spectrum. According to the model presented in Fig. 3 the absence of correlation at long wavelengths is attributed to the dominance of diffraction over refraction.

way the direction of the measured dipole will change in space on the time scale shown in Fig. 2. Remarkably we have found that the jitter (50–100 ps) in the time between flashes is the same for the two states shown in Figs. 1 and 4; so these particular effects do not appear to explain the chaos observed in other experiments [6].

By inserting glass filters between the photomultiplier tube (PMT) and the bubble the dependence of the angular correlations on color was investigated. Figure 5 shows the angular correlation for ''red'' ($\lambda > 500$ nm; filter GG495) and ''blue'' (260 nm$<\lambda<380$ mm; filter UG11) light for the same bubble state. The fact that the red correlation is suppressed indicates that these longer wavelengths diffract out of the bubble's interior on their way to the detector. This implies that the radius of the bubble at the moment of light emission is about equal to the wavelength of red light, consistent with light scattering measurements [3].

Through use of a straightforward light intensity correlation measurement technique we have been able to resolve the deviation from isotropic emission to about one part per thousand. Sonoluminescent states with larger dipole components (say six parts per thousand peak to peak) are characterized by intensity fluctuations that are over a factor of 10 greater than states with dipole components of about one part per thousand or less. Both states display the same flash to flash synchronicity. States with a dipole component tend to lose their preferred direction on a time scale determined by the decay time of the resonant sound field driving SL. Finally when SL falls into a state where the entire emission exhibits a dipole component the ''red'' part of the spectrum does not display this correlation.

Our findings set the stage for attempts to learn about SL [7] from the methods of Hanbury-Brown and Twiss (HBT) [8]. In that theory filtered light from a uniform emitter can display angular correlations in the intensity. This effect has been used to measure the radius of stars. If the dipole component in our broadband measurements is indeed due to the asphericity of the collapse, then this source of correlation will have to be monitored or eliminated in attempts to apply HBT to SL [9]. (We note that preliminary measurements of single photon correlations in SL display the same states as shown in Fig. 1.)

According to our interpretation the observed dipole provides a probe of the degree of nonsphericity of the collapse. Ellipticity is the leading order, quadrupolar, form of a convolution instability. Such instabilities have been studied with regard to bubble and shock wave motion [10] and inertial confinement fusion [11]. They have also been implicated in the upper threshold of SL [12]. Another type of asphericity that occurs in a collapsing bubble is the formation of a jet [13,14].

Through measuring angular correlations one would hope to probe potential symmetry breaking mechanisms that convert the spherically generated stresses of bubble collapse into a flash of light (remembering of course that radial accelerations of shperically symmetric charge distributions do not radiate [15]). While such correlations may exist, measurements (at our current level of accuracy) can be interpreted as providing a diagnostic for the degree of sphericity of an imploding bubble.

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