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Direct observations of single sonoluminescence pulses

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Abstract

Previous reports have described experimental measurements and theoretical descriptions of sonoluminescence without clearly resolving the physical events that underlie this phenomenon. Although incomplete, these results have led to suggestions that sonoluminescence might be such an extreme process that it could serve as an unusual inertial confinement source of fusion neutrons. Such a possibility depends on physical details of individual sonoluminescence events such as the source temperature, diameter, and density. The present report describes attempts to measure the diameter and duration of single sonoluminescence flashes. In both cases the results were limited by the resolution of the instruments, giving diameters of the order of 3 μm or less and durations of the order of 12 ps, or less.

1. Introduction

Sonoluminescence (SL) is the emission of flashes of light by imploding air bubbles in water. Initially, SL was observed as random flashes of light during studies of cavitation phenomena [1]. More recently, procedures have been developed to produce SL with repetitive emission under relatively stable, reproducible experimental conditions [2]. The excellent stability of SL from single acoustically levitated bubbles has made possible detailed studies of the emission characteristics [2–5]. However, since each flash emits only about one million photons, these measurements generally have required that the characteristics be averaged over many (> 10000) flashes.

This paper describes attempts to measure the images and histories of single SL events. If possible, it is important to know whether these quantities differ substantially from their average values. Clearly, the spatial distribution and temporal history are fundamental to probing the basic nature of SL. Furthermore, given the optical flux from a SL event, the duration and size of the source relate directly to its energy density and thus bear directly on remote possibilities such as inertial confinement fusion [4–6].

Sonoluminescence is observed readily by the unaided eye, but its intensity is low enough to make difficult any detailed study of the characteristics of single flashes. Measurements of emission duration have been particularly frustrating. As the emission pulse has been measured with faster and faster detectors, the duration has continued to be shorter than the response limit of the system. The fastest previously reported measurements used a sampling oscilloscope and a detector having a response time of about 0.5 ns to infer that the SL pulse duration was less than 50 ps [7].

The time-dependent collapse of single SL bubbles has been recorded with Mie scattering. For example, averaged measurements of time-dependent Mie scattering have been used to study the unusual nonlinear oscillatory implosion of an acoustically levitated bubble [3]. These measurements indicate that the SL emission occurs when the bubble radius is of the order of 1 μm. However, the resolution limit of this technique is reached precisely at that time when the bubble radiates. Thus, it is not clear whether the radiation event is associated with bubble collapse to even smaller volumes, or whether some other process takes place.

Similarly, measurements of averaged emission spectra have encountered significant frustrations [8]. In this case, difficulties arise from the fact that SL spectra typically extend aggressively into the ultraviolet region where increasing material absorption and decreasing detector sensitivity conspire to make accurate absolute measurements extremely difficult to perform. In spite of these difficulties, spectral data have provided convincing evidence, based on interpretation in terms of a black-body emission spectrum, that SL is a source having an effective temperature of several eV, or more [8].

On the other side of the issue, theoretical descriptions have been outstanding both for their successes and limitations with predicting the behavior of SL. One of the

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outstanding successes has been the remarkably accurate prediction of the extremely nonlinear and repetitive collapse of a bubble under the influence of an oscillatory acoustic driving field [2,3]. This is true both for traditional and more modern Monte Carlo analyses [5,9,10]. Thus far, only the Monte Carlo descriptions have been able to address that very small fraction of the acoustic cycle where the truly microscopic and high-frequency SL emission events occur. However, theoretical calculations to date have not provided a prediction of a fundamental signature of the SL emission process that could be verified by experiment.

The basic difficulty is that, at the instant of SL emission, the bubble is so small (diameter ≈ 1 μm), it is collapsing so rapidly [3] (wall velocity $\approx$ mach 1), and parameters such as pressure ($P \gg 1$ atm) and effective temperature [2,5,8] ($T > 10000$ K) are changing so rapidly that the standard theories or experimental techniques are incapable of showing how these conditions combine to produce the very brief SL flashes. Until the gap between theory and experiment is closed further, it is unlikely that the SL emission process will be understood fully.

The spectral data have also provided some useful insights into the nature of the SL source. The spectra often show increasing intensity into the UV, sometimes with a broad peak in the near UV. When compared with the Planck distribution of black-body radiation, these spectra indicate source temperatures of 10000 to 25000 K, and higher [5,8]. This simple interpretation may not be entirely valid, but it is a straightforward technique that can be used to compare results from different measurements.

Even modest improvements in data quality can make substantial contributions to our understanding of SL. In order to see this, note that the total optical emission provides an indication of the product of the area and duration of the light source. In the present context, the Stefan–Boltzmann law can be written:

$$A\Delta t\; T^4 \approx 8.46 \times 10^{-2}\ \mathrm{cm^2\ s\ K^4}, \tag{1}$$

where T (K) is the effective source temperature. Eq. (1) models SL as thermal emission of 10^6 photons with an average energy of 3 eV radiated in a time Δt from a surface with area A. This model provides a simple relationship between the basic source parameters. Previous experimental results are consistent with the relationship defined by Eq. (1) [2,7,8]: With $\Delta t = 50$ ps and a spherical source of radius 1 μm, Eq. (1) implies a source temperature of 10000 K (consistent with black-body interpretations of measured spectra). Measurements of smaller emission times or source sizes would imply correspondingly higher source temperatures.

2. Experiment

With these considerations in mind, the present experiments attempted to measure the duration and size of the sonoluminescence source. A schematic diagram of the system is shown in Fig. 1. A 50 mm diameter quartz flask filled with degassed water and fitted on the outside with a tetrahedral arrangement of acoustic piezoelectric drivers provided the acoustic resonator. When this assembly was excited at a "breathing mode" resonant frequency of about 26 kHz, a single acoustically levitated bubble of air produced stable trains of SL flashes. Manual injection of a bubble of air near the center of the flask initiated the SL process. The water was a solution of 20% glycerin, by weight, (the glycerin seems to enhance the stability of the SL emission) and 80% distilled water. A continuous stream of chilled air cooled the flask to a temperature of about 10°C.

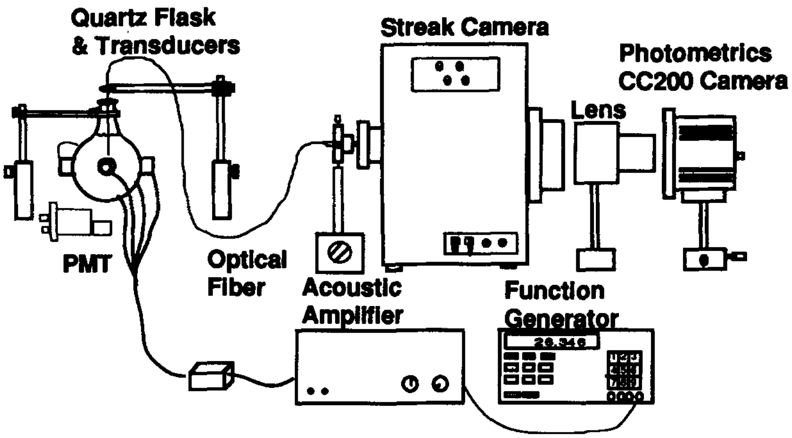

Fig. 1. Schematic layout of streak camera measurement. The SL emission is collected by a 600 μm optical fiber and injected at the input slit of the camera. A combination of optical and electrical signals trigger the streak camera.

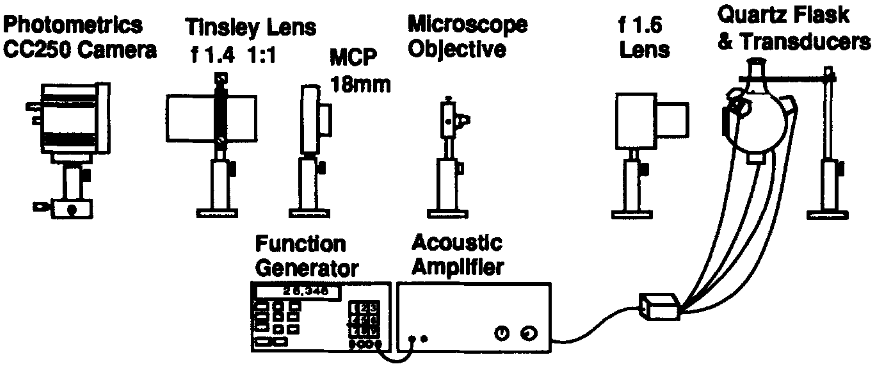

Fig. 2. Schematic layout of imaging measurements. Here, a compound optical system images the SL source onto a camera. The camera and MCP gating can be varied to select one or a number of SL flashes for a given image.

A 600 μm optical fiber that was inserted through the vertical neck of the flask and positioned to within 2 mm of the radiating bubble provided optical coupling between the SL bubble and the streak camera. This fiber collected approximately 0.07 sr of the SL and guided it to the input slit of an E.G.&G. model L-CA-24 intensified streak camera (SC). At the same time, a lens-coupled photomultiplier tube (PM) that measured SL pulses directly through the wall of the flask produced electrical pulses that triggered the control electronics for initiating the SC and data recording electronics.

The actual streak camera trigger pulses were timed with respect to the sinusoidal 26 kHz electrical "drive" for the piezoelectric transducers. Because of inherent delays, the system recorded the SL pulse that followed the receipt of an appropriate trigger pulse from the PM. This approach resulted in a jitter of about 100 ps (mostly due to the electronics) in the apparent position of the SL pulse on the streak image. Images on the output phosphor of the SC were recorded by a Photometrics CC200 Camera and stored on a Macintosh fx computer.

Except for some differences in the flask, the system for recording images of single SL flashes, shown in Fig. 2, was similar. The flask was a 250 ml laboratory boiling flask with a circular area ground away from the side and resealed with a flat quartz disk. This flat "window" allowed much improved observation of the radiating bubble, but it degraded the basic symmetry of the SL arrangement. This flask had a "breathing mode" resonant frequency of about 40 kHz. By varying the electrical drive to each transducer, the position of the bubble was adjusted to compensate for asymmetries which otherwise would cause the bubble to be displaced from the center of the flask.

A compound telescope consisting of a 44 mm F1.6 lens and a variety of microscope objective lenses imaged the SL light from the window onto a microchannel plate intensifier (MCP). The microscope lenses allowed quick changes of system magnification from 15 × through 120 ×. A 1:1 lens relayed the image from the MCP to a Photometrics CH250 camera. The imaging system recorded the SL images with a series of increasing magnifications. The magnification of the system was calibrated with images of a 100 μm ruling on glass. The gross features of these images showed no unusual distortions. In addition to demonstrating the magnification, these images showed that the sharpness of the line edges of the order of 10 μm.

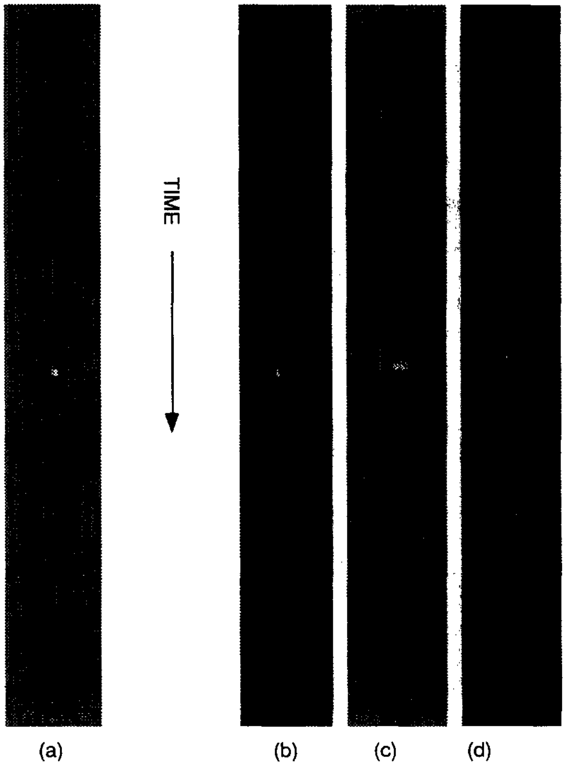

Fig. 3. Sonoluminescence streak data. This figure compares the recorded streak of a SL flash with a 10 ns streak duration (a) and a 2.3 ns streak duration (b) with a 13 ps laser pulse (c) and a 6 GHz "comb" sweep calibration (d), both with 2.3 ns sweep duration.

The images were recorded with two approaches to gating: gate times that guaranteed that only one flash would be recorded, and gate times that integrated over a number of pulses. The camera controller gated the camera for 200 ms, but the MCP was gated independently for times that varied from 1 μs to 2.5 ms for recording of the images. The MCP gating time controlled the number of SL pulses that would contribute to a given image: 1 μs yielded a time-integrated image of a single pulse, while 2.5 ms integrated more than 100 pulses. The gating allowed control of the tradeoff between minimizing spurious scintillation from the MCP, recording a single pulse, and using maximum MCP gain.

The overall imaging sensitivity was approximately 4.5 counts per photon (average energy = 2.75 eV) incident on the MCP. For this calibration, an E.G.&G. #550-1 radiometer/photometer measured the transmitted optical power of a tungsten lamp, filtered (Schott 4-96 broadband green) and injected into a 100 μm core fiber. A gated image of the light from the calibrated fiber output, when placed at the center of the SL flask, yielded the overall system calibration. This approach is only approximate (±30%), as it ignores differences between the spectral dependences of the radiometer sensitivity, the spectral response of the S-20 MCP photocathode and the spectrum of SL.

3. Results

3.1. Temporal measurements

The streak camera recorded the emission signature for a series of increasing sweep speeds. In all cases, the emission duration was indistinguishable from the impulse response of the camera. In the case of the highest sweep speed, approximately 4 ps per pixel, the SL pulse had an apparent width of about three pixels, or 12 ps. This recorded time was comparable to the fastest signal that previously had been observed by this camera, a 13 ps laser pulse (as determined independently with an autocorrelation technique).

Fig. 3 shows a comparison of the SL pulse (recorded with 10 and 2.3 ns sweep durations), the 13 ps laser pulse, and a 6 GHz "comb" signal that is used to calibrate the camera sweep speed. The SL flash duration cannot be distinguished from the impulse response of the system, as there is no significant difference between the streaks recorded with the 10 and 2.3 ns sweep times. With the faster streak, the SL flash is somewhat shorter than the 13 ps laser pulse (FWHM ≈ 4 pixels/16 ns). These results suggest that the SL flash duration is less than the approximately 12 ps response time of the streak camera.

We recognize that this result is somewhat beyond what we expected to be able to measure, due largely to modal and material dispersion in the optical fiber. Each of these broadening mechanisms should be expected to contribute about 30 ps to the response function of the streak camera. However, the reported results were observed repeatedly and consistently during many independent trials. One partial explanation for this inconsistency is that the experimental geometry may have excited a small fraction only of the fiber modes. With respect to the system bandwidth, the low intensity of the source and spectral dependences of the system components might have narrowed the overall effective bandwidth of the measurement significantly.

3.2. Spatial measurements

The single-flash images show two qualitatively different kinds of regions: a diffuse region having a size of the order of 30–100 μm, and a smaller, brighter region with a diameter of the order of 3 μm, or less. Integration of measured intensities gave single pulse emission totals of about 1 to 5×10^5 photons.

The images tend to emphasize the larger diffuse region, due to the speckle behavior of the intensifier tube. This is due to the poor collection efficiency of the optics (solid angle ≈ 0.07 sr), the quantum efficiency of the MCP photocathode (≤ 15%), and the high gain of the MCP intensifier. Thus, the larger diffuse region takes on the appearance of a "speckled" area, with a brighter peak typically evident somewhere near the middle of the diffuse region.

Fig. 4 shows a typical image. Here, the peak intensity of the image corresponds to about 180 counts/pixel, with

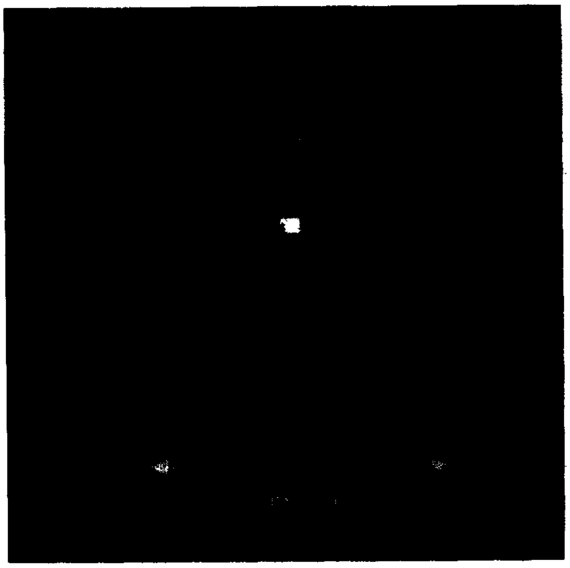

Fig. 4. Image of a single SL flash. Single-flash images typically show a bright spot with a resolution-limited 3 μm diameter surrounded by a diffuse 100 μm region of dimmer spots. The images correspond to total emission of about 1 to 5×10^5 photons/flash.

a background level of about 80 counts. The background level is produced by thermal excitation of image tube elements during the camera gassing period.

The diffuse region often displayed extreme changes in intensity and geometry on successive recordings of single pulses (separated by ≈ 1 s). At other times, this region was roughly circular and relatively consistent from image to image. The appearance of circular regions tended to correlate with overall stability of the sonoluminescence experiment.

Surprisingly, the multiple-pulse images tend to show a very bright, central spot surrounded by a more uniform dimmer region. The central peak is more than ten times brighter than the dim region. Furthermore, the FWHM of the central peak still is only 3 μm, while the diameter of the dimmer region is 50–100 μm. Thus, not only is the central peak extremely small, but its position is extremely stable on a pulse to pulse basis. This suggests that photons from the bright spot were recorded in almost exactly the same position in at least a large fraction of the consecutive flashes. In contrast, the random distribution of detected points from the dimmer region leads to a rather uniform brightness that tends to decrease as distance from the bright spot increases.

The smaller region, for all magnifications, was never observed to be larger than the resolution limit of the system. Thus, the smaller region had a diameter of less than 3 μm. Surprisingly, the position of this region was extremely stable, as recordings that integrated up to 100 pulses still showed this region to have a diameter still of the order of 5 μm or less. Thus, over short time periods, the position of this region was stable to within a few μm.

We were unable to determine whether the larger diffuse region truly was characteristic of the source, or whether it was associated with some optical aberration. It was difficult to image the SL source precisely, because acoustic-induced flexing of the flask and window perturbed the effective focal length of the system. Furthermore, the macroscopic motions of the bubble introduced significant uncertainty as to whether or not the bubble was positioned at the desired focal point for any given single-pulse image.

Although the SL bubble often showed excellent long-term position stability, there were also times when it exhibited jittery "dancing" [2] motions that were evident to the naked eye. These "macroscopic" motions were another fickle aspect of the SL. The "macroscopic" motions appeared to be orbital motions with frequencies from a few to tens of Hz, and diameters from hundreds to thousands of μm. Needless to say, such motions tended to interfere with the imaging measurements by making it difficult to focus the optics or to position the bubble for good placement on the image. Qualitatively, the system often appeared to be metastable with respect to these oscillations. At times the bubble position was steady, but at other times the oscillations would commence unpredictably. Sometimes, but not always, slight changes in acoustic frequency or amplitude would terminate the oscillations and return the bubble to a stable position.

4. Conclusions

The streak camera results indicate that the SL duration is of the order of, or less than, 12 ps. There are significant uncertainties associated with this result, but it seems clear that single-pulse durations are substantially less than the 50 ps result that was reported previously for pulse-averaged measurements [7].

The spatial images show a bright spot in the source with a diameter of about 3 μm, or less, and a larger diffuse region with a diameter of 50 to 100 μm. Again, these results are consistent with pulse-averaged Mie scattering measurements [3]. They also demonstrate that the position of the SL source can be extremely stable (to within 3 μm) on a pulse-to-pulse basis.

The purpose of the present study was to use modern diagnostics to measure the spatial and temporal distributions of single-pulse SL with the highest resolutions possible. The measurements have at least verified the results of multiple-pulse measurements. The results suggest further that the duration and size of SL sources might still be substantially shorter and smaller than has been measured.

These results leave open the possibility that the SL source is substantially hotter than the 10000 K that was implied above by Eq. (1). Recent theoretical results have come to similar conclusions. If the source temperatures actually are substantially hotter than 10000, then, as numerous authors have speculated [4–6], SL might prove to be a fascinating approach to inertial-confinement fusion. We would hope that continued experimental and theoretical work will illuminate the nature of sonoluminscence further.

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