Spectrum of Synchronous Picosecond Sonoluminescence

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The clocklike emission of picosecond pulses of light with a peak power over 30 mW has been observed to originate from a bubble trapped at the velocity node of a resonant sound field in water. The spectrum of this bright sonoluminescence is broadband and our measurements show that if a spectral peak exists, it lies at photon energies above 6 eV.

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We have measured the spectrum of the picosecond pulses of light that are synchronously generated by the extremely nonlinear motion of a small cavity which is trapped in a sound field in water. As shown in Figs. 1 and 2 the spectrum extends from above 700 nm to below 190 nm. The portion which we have been able to calibrate lies in the range 240 to 620 nm. Most remarkable is our observation that the intensity of emitted light (sonoluminescence or SL) is increasing as the ultraviolet cutoff of water is approached. The spectral peak of SL appears to be located at photon energies above 6 eV. For our sound fields (Mach number $\approx 10^{-5}$ and sound energy level of order 10^{-11} eV/atom) the defect (i.e., cavity) is therefore capable of concentrating the ambient energy by more than 12 orders of magnitude.

For an air bubble trapped in filtered, distilled, degassed water we also find that (a) there is no evidence of spectral lines at a resolution of 1 nm; (b) the relative spectral density is independent of the frequency of the sound field f (which is also equal to the number of SL flashes per second) for 60 kHz $> f >$ 25 kHz; (c) the relative spectral density (at room temperature) is independent of the integrated light output for fivefold variations in SL intensity that are affected by changing the level of acoustic excitation; and (d) the intensity of SL increases by over a factor of 10 and becomes even more heavily skewed towards the UV when the water is cooled below 10 °C. Sonoluminescence would appear to be an ideal candidate for a broadband calibrated light source (190 nm $< \lambda <$ 750 nm).

The experimental arrangement for achieving stable SL has been described elsewhere [1,2]. Basically the output of a synthesizer drives piezoelectric transducers that in turn excite breathing resonances in the water-filled spherical quartz flask to which they are attached. Above a threshold acoustic amplitude a bubble can be trapped and maintained at the velocity node ($r=0$). As the amplitude is increased further a regime is reached where one can observe a blue light with the unaided eye. Measurements indicate that this light actually consists of flashes

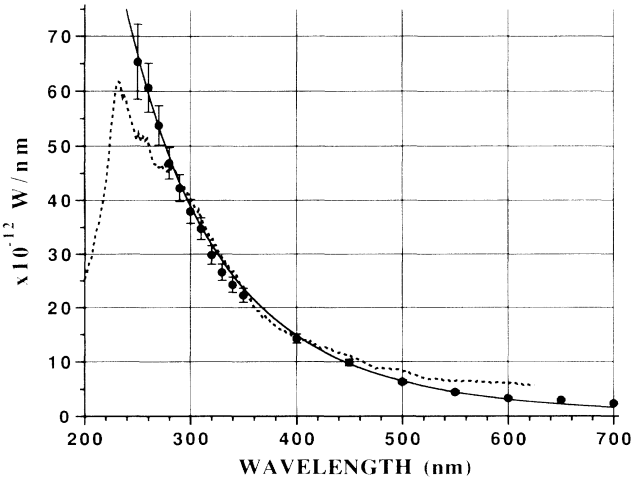

FIG. 1. Calibrated spectral density of the synchronous picosecond flashes of sonoluminescence at 22 °C. The average spectral energy density of a single flash can be obtained by dividing by the acoustic frequency of 27 kHz. The dotted line was obtained via the D lamp calibration. The points with error bars were obtained by calibrating our apparatus with a QTH standard of spectral irradiance. The solid line is a 25000 K blackbody spectrum.

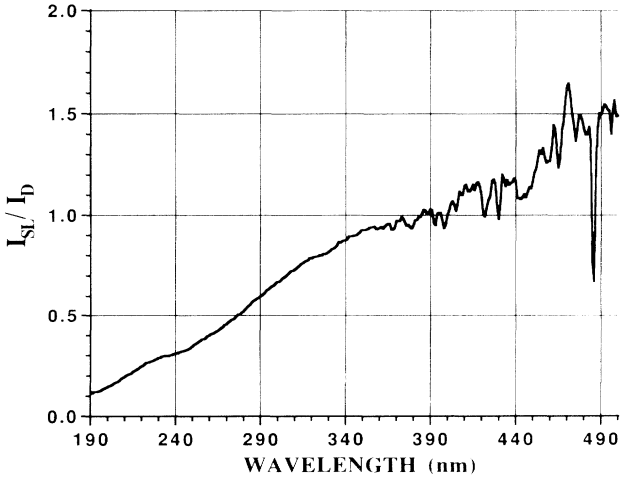

FIG. 2. Ratio of spectral density of sonoluminescence I_{SL} to that of the deuterium lamp I_D at 22 °C. The peaks and valleys are due to the deuterium lamp. The ratio has been normalized to unity at 400 nm.

that are less than 50 ps long and repeat at the frequency of the sound field, with a jitter (in the time between successive flashes) that is also less than 50 ps [2].

The spectrum of these flashes was obtained by collecting light by three different methods: (1) The emission from a bubble (trapped in a 45 or 72 mm outer diameter quartz flask) went straight into an ARC 275 mm spectrometer at an entrance angle of $F/8$; (2) a quartz fiber (0.8 mm core) was positioned in the water so that one end was about 2 mm from the bubble and the other end was at the entrace slit of the spectrometer; and (3) an $F/0.85$ 51 mm biconvex quartz lens was used to image the SL onto the input slit of the spectrometer. In methods (1) and (2) the entrance and exit slits were open over 2 mm so that the resolution was limited to about 10 nm. In method (3) the slits were pinched down to 25 μm and so the resolution for this case is better than 1 nm. In view of chromatic aberration this last method is not useful for calibrating relative spectral densities. However, method (3) is of value in searching for the presence of sharp spectral lines.

The output of the spectrometer was measured with a broadband photomultiplier tube, PMT (Hamamatsu R2027). The signal-to-noise ratio was improved by collecting the output of the photomultiplier tube with a lock-in amplifier synchronized to the frequency of the sound field. Since each cycle of sound creates one flash of light, the acoustic oscillations play the role of light chopper. Each data point was acquired by averaging for 1 s and between each acquisition there was a dwell time of 1 s. During each run a second PMT was used to monitor the total intensity of the light emitting bubble. Since our data are obtained by a scanning technique, temporal variations in SL intensity could incorrectly modify the spectrum. Use of the reference tube enabled us to select reliable runs where the intensity drifted by less than 5%. Use of order-sorting filters (at 305 and 495 nm) that pass long wavelengths was necessary above 400 nm. The gratings (1200 rulings/mm) were blazed at 300 and 500 nm. Our typical signal is about 50 times the noise floor and the $\sqrt{N}$ fluctuations introduced by the finite averaging time are about 5%.

The broadband calibration of the flashes of SL was obtained by comparison with (1) a calibrated quartz tungsten halogen (QTH) lamp standard of spectral irradiance and (2) a deuterium light source (Oriel 6316/Hamamatsu L1403 as well as a Hamamatsu L1128). Figure 2 shows the ratio of the spectral density of SL and the D lamp (Oriel 6316) as a function of wavelength (normalized to 1 at 400 nm). The D lamp calibration of SL (dotted line in Fig. 1) was obtained from Fig. 2 by using the calibration provided by Oriel which is valid to about 5%–10% between 230 and 600 nm. The peaks in Fig. 2 (e.g., 485 nm) are due to the D lamp. The sharp dropoff in the SL spectrum (Fig. 1) for wavelengths $\lambda < 230$ nm follows from the Oriel (but not Hamamatsu) calibrations of the D lamp. The signal-to-noise ratio is also rapidly deteriorating as 200 nm is approached. Overall, the two methods of calibration yield results that are well within the known limits of accuracy of the QTH and D lamp sources.

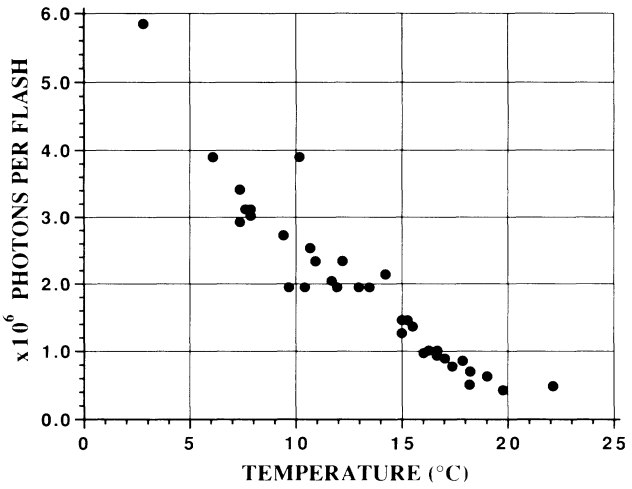

FIG. 3. Number of photons emitted per flash of SL as a function of temperature. At each temperature we recorded the output of the brightest bubble attainable.

At room temperature a variation in the sound field amplitude can cause the intensity of SL to change by about a factor of 5 [2]. However, we have also found that by lowering the temperature to below 10 °C the total emission from a single bubble can be enhanced by over a factor of 10. Figure 3 shows a plot of the number of photons per flash of SL as a function of temperature for an excitation frequency that varies from $f = 27\,127$ Hz at $T = 7$ °C to $f = 27\,348$ Hz at $T = 22$ °C (as the acoustic resonance varies slightly with temperature).

A comparison of the relative spectral densities of SL emission from a cavity in water at 10 and 22 °C is shown in Fig. 4. The raw data taken by method (2) are uncorrected for the effect of the grating, PMT, and transmission coefficient of the fiber. The curves have been normalized to equal area. Note that the 10 °C curve has a much greater spectral density in the UV.

Our motivation for measuring the effect of a lowered temperature on SL comes from earlier experiments on transient sonoluminescence [3]. In these works an intense sound field created a stream of cavities which emitted light in an irregular and unpredictable fashion as they collapsed. The finding that transient SL emitted more light upon cooling led us to perform the above experiment. The enhancement of stable SL, however, was not guaranteed. The effect of temperature on transient SL was originally [3] interpreted as being due to its effect on the distribution of nucleating cavities and not the dynamics of a single trapped bubble. We believe that the increase in intensity of SL as the water is cooled is due to the fact that water dissolves about twice as much air at 0 °C as at room temperature [4]. We hope to show that

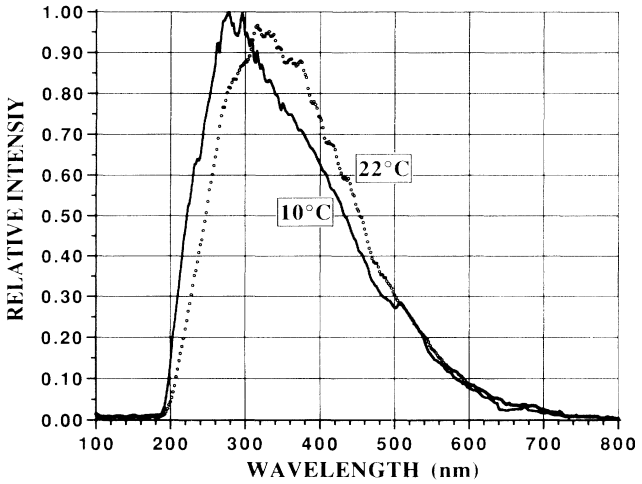

FIG. 4. Raw data for the spectral density of SL at 10 and 22 °C. The peaks have been chosen so that the curves have equal area. These curves have not been corrected for the fiber grating, or PMT. The grating is blazed at 300 nm.

in this case the collapse of a bubble is more violent and so leads to a greater energy focusing [5]. The spectrum shown in Fig. 1 has differences from those measured previously [6] for transient SL. (For a review of transient SL see Ref. [7].) This effect could be due to the means of calibration, the frequencies used, or possibly the fact that stable SL may be a different phenomenon.

A single flash of SL is too short to be measured in one shot, but the response of a fast PMT to a flash of SL can be averaged by a sampling oscilloscope. At room temperature this method has established an upper bound of 50 ps on the flash width. The resolution with which we could use this technique to measure the width of a flash of cold SL was hampered by shortcomings in temperature control and an increase in unwanted vibrations that are parametrically excited by the imposed sound fields which are larger than at room temperature. Nevertheless, our use of a 34 GHz sampling scope (HP54123) and a microchannel plate PMT (Hamamatsu 2809U) enabled us to establish a very conservative upper bound of 150 ps on the width of the more intense flashes that occur at 10 °C. This already implies a lower bound on the peak power of individual flashes of 30 mW. Whether the increased width is due to a noise-induced variation in pulse heights or a fundamental dynamical effect remains to be seen.

As shown by the solid line in Fig. 1 the spectrum of room-temperature SL is fitted quite accurately by the tail of a 25000 K blackbody spectrum [8]. Similarly the 10 °C spectrum can be compared to a blackbody spectrum in excess of 50000 K. Future experiments with other liquids or gases will help determine whether the broadband contribution to the SL spectrum is due to a Planck distribution.

Sonoluminescence is a phenomenon whereby energy entering a continuum at the macroscopic level spontaneously focuses down to the molecular-atomic-electronic degrees of freedom so as to generate light. This transduction is quite efficient: The emitted light energy is comparable to the acoustic energy that would be damped away by the shear viscosity in the absence of the bubble. Theoretical explanations for this phenomenon, especially as regards the fast time scales, is lacking. Here we have reported that the spectral peak lies at photon energies which are higher than those required to dissociate the molecules comprising the medium in which the energy concentration occurs.

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[8] Our observations can be contrasted with those of E. B. Flint and K. S. Suslick [J. Am. Chem. Soc. **111**, 6987 (1989); Science **253**, 1397 (1991)]. They assign SL a temperature of 5000 K and furthermore claim that the continuum contribution to SL is not blackbody. Their investigations focused on Silicone oil which is surely different from water. But, more importantly, they studied the variety of SL which originates from unknown distributions of cavitating transient bubbles. This "transient SL" may also be a different physical phenomenon from the synchronously repeating stable SL that originates from the extraordinarily nonlinear motion of a single trapped bubble as reported in our paper.