Single-Bubble Sonophotoluminescence

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Acoustic cavitation involves the formation, growth, and collapse of microbubbles in a fluid. This process may lead to the emission of light from the collapsing bubbles, referred to as sonoluminescence (SL).[1] Despite the fact that SL from a bubble field (multibubble sonoluminescence (MBSL)) has been extensively investigated, there is still some debate over the mechanism involved in the light emission from the acoustically driven bubbles.[1−3] The relatively recent discovery of single-bubble sonoluminescence (SBSL)[4] has led to several experimental and theoretical investigations owing to the "controlled" experimental conditions under which it is produced.[1−8] The key features of SBSL are the featureless emission spectrum and the synchronous emission of the SL pulse on every acoustic cycle.[4,7−9] The pulse width of SBSL has been measured to be in the range of ∼35−350 ps.[9]

We have recently reported on the use of MBSL to excite fluorescent solutes in water and nonaqueous solvents.[10] The ensuing emission, referred to as, sonophotoluminescence (SPL), possesses all of the emission characteristics normally associated with the direct photoexcitation of the solutes. In the present report, we show that SBSL can also be used to photoexcite fluorescent solutes. In addition, SBSL can act as a pulsed picosecond light source suitable for monitoring the temporal emission profile of the excited solute.

The experimental system used to levitate a single-bubble was similar to the one described by Matula.[11] A 250 mL beaker was used as the SBSL cell. The bottom of the cell was attached to a hollow-cylindrical transducer (American Piezo Ceramics), which was driven by a HAMEG function generator (HM8131-2) through a Krohn-Hite model 7500 amplifier at ∼25 kHz. The bubble was levitated at an acoustic pressure of ∼1.2−1.3 atm (measured by a calibrated needle hydrophone (Precision Acoustics, HPM1/1)). SL/SPL spectra were recorded by placing the SB cell in the cell compartment of a spectrofluorimeter (Hitachi, F-4500). A full scan (200−800 nm) could be recorded in about 2 min. The SL/SPL pulse width measurements were made with a Hamamatsu (R647-04) photomultipler tube (PMT). The rise time of the instrumentation setup was about 2−3 ns. All solutions were made with Milli-Q water and used on the day of preparation. High purity samples of pyranine (Molecular Probes Inc.), fluorescein (Sigma), and eosin (Sigma) were used as received. The temperature of the solutions remained in the range of 20−22 °C while spectra were recorded.

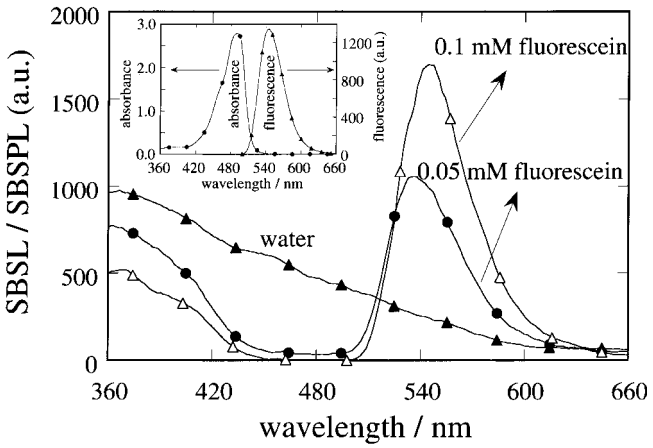

Figure 1. Single-bubble sonoluminescence (SBSL) spectrum of water and single-bubble sonophotoluminescence (SBSPL) spectra of 0.05 mM and 0.1 mM aqueous fluorescein solutions. The insert shows the absorption and fluorescence (photoexcitation $\lambda = 480$ nm) spectra of a 0.025 mM fluorescein solution.

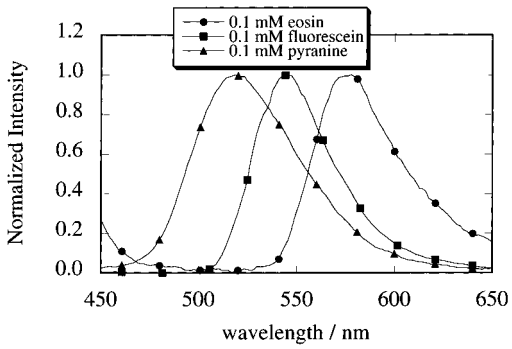

Figure 2. Normalized (to the individual maximum intensities) SBSPL spectra of 0.1 mM aqueous solutions of eosin, fluorescein, and pyranine.

A single sonoluminescing bubble was levitated in sufficiently degassed water and in aqueous solutions containing the dissolved fluorescent solutes. The uncorrected SBSL spectrum of water and the emission spectra obtained from 0.05 mM and 0.1 mM fluorescein solutions are shown in Figure 1. The key features to note are: (1) SBSL has been absorbed, almost completely, in the wavelength region 420−500 nm—on examining the absorption spectrum of fluorescein (see insert of Figure 1), it is clear that the absorption of SBSL is due to the added fluorescein; the absorption of the SBSL also increases as the concentration of the solute increases; (2) a new emission band with an emission maximum of ∼540 nm has appeared—comparing the emission spectrum (see insert of Figure 1) of fluorescein, generated by conventional photoexcitation, it can be concluded that the new emission observed is due to the fluorescence from fluorescein. These results are consistent with the SPL observation that we have reported in multibubble systems.[10] In keeping with the terminology of our MB work, the SPL from the single-bubble system will be referred to as SBSPL.

SBSPL was also observed with other fluorescent solutes, eosin, and pyranine. The normalized emission spectra of 0.1 mM

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aqueous solutions of eosin, fluorescein, and pyranine, that are shown in Figure 2, illustrate that the SBSL can be shifted to any desired wavelength, by choosing an appropriate fluorescent solute. The primary mechanism involved in the generation of SBSPL is the same as in the MBSPL system. The SBSL that is absorbed by the dissolved fluorescent solutes leads to the in situ vibronic excitation of these molecules, which then fluoresce.

The SB system has also allowed us to estimate the lifetime of the SBSPL emission. As mentioned earlier, SBSL has a pulse width of ~35−350 ps.[9] The impulse response of the detection system that we have used cannot directly measure the true pulse shape of the SBSL. However, since the fluorescence lifetime of the solutes used is in the nanosecond regime, the measurement of the PMT response in the absence (SBSL from water) and in the presence of the solutes has enabled us to calculate the SBSPL lifetime.

Typical impulse response curves of the PMT to the SL and SPL (fluorescein) pulses are shown in Figure 3. It is very clear that the decay of the SPL pulse is slower than that of the SL pulse. Deconvolution of these data yields a lifetime of 5.0 ns for the fluorescein emission decay. This value is in excellent agreement with the reported fluorescence lifetime (5.0 ns) of this solute.[12]

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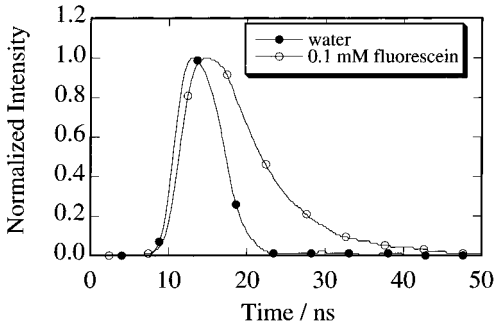

Figure 3. Impulse response of the PMT to the emission pulse (an average of 100 individual pulses have been taken) in the absence and in the presence of 0.1 mM fluorescein. Rise time of the instrument was about 2−3 ns.

In summary, SBSL in water can be used to photoexcite fluorescent solutes yielding SBSPL. SBSPL may itself be used as a light source for generating short light pulses of variable wavelength by selecting appropriate fluorescent solutes.

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