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Near-infrared emissions in single-bubble and multibubble sonoluminescence

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Comparisons of light emissions from multibubble and single-bubble sonoluminescence in the near-infrared band extending from 800 to 1050 nm have been investigated. In argon-water mixtures, single-bubble and multibubble spectra are similar in appearance. In sodium chloride-water mixtures, the multibubble spectrum shows evidence of the $3d$-$3p$ sodium emission line, whereas the single-bubble spectrum shows no such emission. For single bubbles, the near-infrared emissions change linearly with driving pressure. No evidence of near-infrared emissions are observed below the visible luminescence threshold.

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Sonoluminescence refers to the acoustically driven violent collapse of bubbles in a liquid resulting in the emission of light. Two general cases of sonoluminescence exist: Single-bubble sonoluminescence (SBSL), whereby a single levitated bubble is made to emit light, and multibubble sonoluminescence (MBSL), whereby individual bubbles within a typically dense cloud or field of bubbles emit light. The degree to which these two systems are related is still unknown, although the commonality of cause (acoustic cavitation) and effect (light emission) suggests an association in the underlying physics.

Measurements of the light properties show both similarities and differences in the UV-VIS spectrum. MBSL spectra typically contain line and band emissions that extend above a continuum. Except for a few isolated cases [1–3], SBSL spectra show only a continuum structure, devoid of line and band emissions. Using band emission information from MBSL spectra, the average temperature of an MBSL bubble interior has been measured to be between 3000 and 5000 K [4,5]. This measurement is an average in the sense that individual bubbles within the cavitation cloud may show large deviations [6]. It has even been suggested that some individual bubbles within a cavitation cloud may yield much more extreme conditions than exists in SBSL [7], where the temperatures have been computed to be greater than 10 000 K [8,9], although recent computations suggest that when water vapor is incorporated, the maximum temperature in SBSL is significantly lowered [10]. Although still controversial, recent computations suggest that the light emission mechanism for both systems is due to heating of the bubble interior [9,11,12].

Whereas MBSL can be achieved in many fluids, at frequencies ranging from approximately 20 kHz to over 1 MHz, SBSL has so far only been achieved over a relatively small region in the parameter space of liquids, frequencies (≈ 7–70 kHz), and pressures (≈ 1.2–1.4 bar). Direct comparisons between MBSL and SBSL continue to provide researchers with information about the mechanics and kinetics associated with the light emission mechanism. Ashokkumar *et al.* [13] showed that alcohol quenching of SBSL is similar to previous studies of alcohol quenching in MBSL [14], suggesting a common mechanism for quenching. Togel *et al.* [15] interpreted the effect as being due to a lowering of the effective gamma of the gas. Matula *et al.* [16] found that the pulse duration of sonoluminescence flashes in a glycerin-water MBSL systems were not too different from SBSL pulse durations. However, the pulse duration of sodium emissions in NaCl solutions apparently is significantly increased [17]. In another study involving NaCl solutions, Matula *et al.* [18] showed that the continuum characteristics exhibited similar trends of increasing intensity toward lower wavelengths, and whereas MBSL spectra had distinct sodium emission lines near 589 nm, SBSL showed no such lines. Recently, band emission from SBSL was shown to occur for unstable, dim noble gas bubbles [3]. These emissions appear similar to emission bands observed in MBSL. Parenthetically, one might also expect that dim SBSL ‘‘air’’ bubbles should also generate band emissions if driven off resonance.

In this paper, we extend the comparison of the emission characteristics of SBSL and MBSL to the near-infrared region (800–1050 nm). The spectral properties in argon/water mixtures and NaCl solutions are examined. Furthermore, the emission intensity of SBSL in the near-infrared (NIR) region is measured in order to determine if NIR emissions can be observed below the visible luminescence threshold (which might be expected if SL is thermal in nature).

The experimental apparatus for MBSL included a 20 kHz acoustic horn (Sonics and Materials, Inc.) threaded into a custom made flow cell. A stainless steel cylinder cell with multiple ports for fluid circulation, gas sparging, liquid sampling, and static pressure and temperature control was used [19]. Once introduced to the cell, solutions were sparged with argon gas for 30 min and thermostated to $5 \pm 0.5\,^{\circ}\mathrm{C}$ by flow at 4 l/min through a coil submerged in a cooling bath. The presence of noble gases and reducing the solution temperature increased the violence of cavitational collapse, brightening SL intensity. Solutions were then sonicated with an average power density of 12.5 $\mathrm{W/cm^2}$, as measured by a custom complex power meter. Data collection began after

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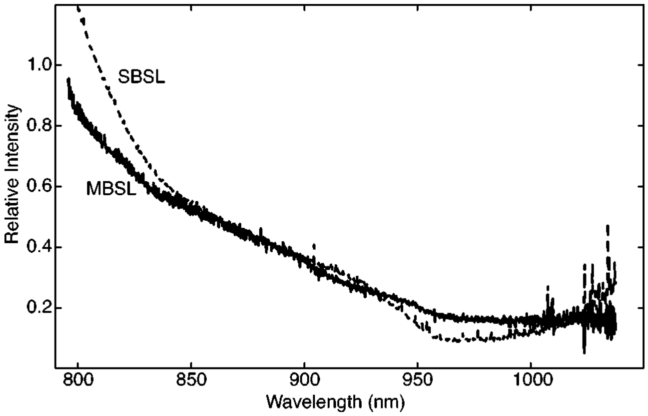

FIG. 1. Comparison of NIR spectra collected for SBSL (dashed line) and MBSL (solid line). The features near 1050 nm are noise.

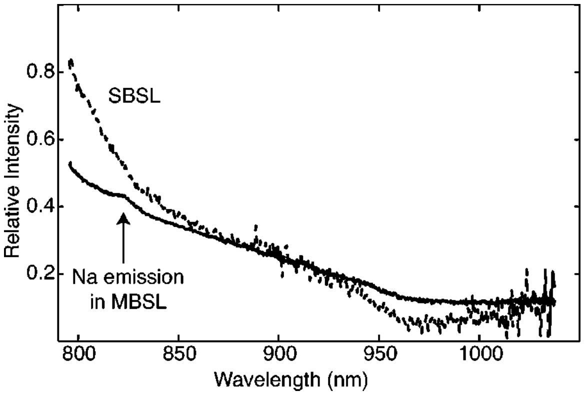

FIG. 2. Comparison of NIR spectra in 0.1 M NaCl collected for SBSL (dashed line) and MBSL (solid line).

approximately 10 min, when spectral intensity stabilized. A fiber optic probe inserted into the cell was used to collect the light.

The SBSL system included a round-bottom spherical flask, argon-filled (≈ 3 mmHg), filtered water, situated in a light-tight enclosure. A bubble was levitated at the center of the flask. A 1-inch $f/2$ achromatic lens was used to focus the sonoluminescence light onto the fiber tip. A broadband light source was used to align one focal point of the lens with the levitated bubble. Then, using laser light scattered from the bubble, the fiber was aligned with the other lens focal point by maximizing the signal intensity of a photomultiplier tube (PMT) at the other end of the fiber. The PMT was then replaced with the spectroscopy system.

A Kaiser HoloLAB 5000 Raman system was used to collect the data, operated with the Raman laser turned off. The internal optics are optimized for a fiber optic input. Two in-line transmission gratings transmitted the incident light in the first diffraction order ($\approx 80\%$ maximum efficiency) onto a 1024×256 thermoelectrically cooled charge coupled device (CCD) array. To increase light throughout a 500 μm slit was used (resulting in a resolution of ≈ 2.3 nm). Kaisers HoloGRAMS software was used to collect data, subtract background, and remove cosmic ray signals. The raw data was then flat-field corrected to account for the light gathering differences of the two gratings.

Figure 1 shows normalized spectra for MBSL and SBSL in water/argon mixtures collected with this system in the range 800–1050 nm. Data for the SBSL system was integrated over a 15 min interval while the MBSL data was integrated over a 10 min interval. The SBSL spectrum shows a faster rate of increase in intensity at lower wavelengths. In addition, there is a dip in the SBSL spectrum near 960 nm, corresponding to a water absorption band in this region. The MBSL spectrum shows qualitatively similar spectral characteristics. The dip in the MBSL spectra is not as pronounced, due to the large void fraction of bubbles generated in the MBSL system; the light had less travel distance through water. Although typical spectra in the VIS region show a relatively flat profile at the longer wavelengths [18], it is shown here that in actuality, the spectral intensity continues to decline at lower energies. However, except for the water absorbance feature in SBSL near 960 nm, there do not appear to be any features of the spectra that differ from spectra observed in the visible region.

To observe distinct features in the spectra, we added NaCl (0.1 M) to the solution. Figure 2 shows how the spectra change. The MBSL spectrum shows a sodium emission band near 819 nm, resulting from the $3d$-$3p$ transition from neutral sodium. The SBSL spectrum shows no such emission, to the resolution of our apparatus. This result is to be expected, since no sodium emission has been observed in the visible region of the spectrum [18]. The commonly observed sodium doublet emission in the visible region near 589 nm results from the $3p$-$3s$ transition, with an excitation energy of $\Delta E = hc/\lambda \approx 2.1$ eV. Its emission intensity is approximately of an order of magnitude greater than the emission intensity for the $3d$-$3p$ transition near 819 nm, which has an excitation energy of ≈ 3.6 eV [20].

To complete our studies of the NIR emissions, we measured the total integrated light intensity of an air/water SBSL system in the wavelength region extending from 800 nm to 1700 nm. The light was collected with an optical fiber (IR sensitive, 400 μm diameter tip) probe connected to an IR femtowatt photoreceiver (New Focus model 2153) [21]. Simultaneous measurements in the visible region (350–650 nm) were made with a Photomultiplier tube (Hamamatsu model 2027) [22]. From the intensity data, the total number of photons emitted in each region were calculated and are shown in Fig. 3.

The number of emitted photons in the visible and NIR regions is linear with drive pressure (measured using a calibrated Dapco needle hydrophone). Because the position of the SBSL bubble will change with pressure [23], the fiber-to-bubble distance was maintained (at 3 mm) by viewing the fiber tip and bubble together using a microscope and CCD camera, and adjusting the cell vertically each time the pressure amplitude was changed. The total distance moved during the experiment was approximately 2 mm. The distance to the PMT (≈ 33 mm) remained relatively constant throughout the experiment.

A comparison of the total number of emitted photons in the visible and NIR regions is slightly misleading. Spectral studies have shown that the light intensity is much greater toward the UV, and quickly dies off into the NIR. Therefore,

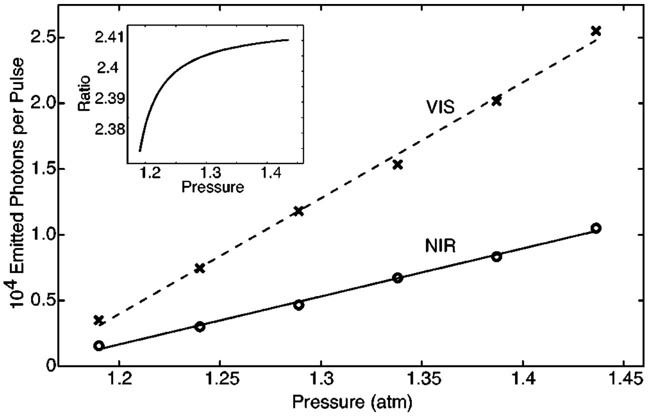

FIG. 3. The integrated light intensity in the NIR and UV-VIS were converted into photons emitted by the bubble per flash, and plotted here. The lines are a linear least squares fit to the actual data points shown. NIR data are shown as circles, UV-VIS data are shown as squares. The inset shows the ratio of visible to NIR photons, using the linear fit curve.

the relative number of photons in the visible should be much greater than Fig. 3 suggests; however, the size of the detectors' wavelength window must accounted for. The much larger window of the NIR detector results in a higher photon number.

As the pressure is increased, a slightly higher ratio of the visible-to-NIR photon yield is observed, as shown in the inset. Here, the curve is obtained from a linear fit to the data, and not from the data itself, owing to the large variation in the data. Again, this ratio will depend critically on the wavelength region of the detector.

Finally, a thermal origin to SBSL suggests that IR emissions may be observed below the visible luminescence threshold. We tried, unsuccessfully, to measure NIR emissions below the visible luminescence threshold. This was due, in part, to noise introduced by a preamplifier, as well as unstable bubble motion in the subluminescence region.

In conclusion, we have performed NIR studies of sonoluminescence emissions, comparing and contrasting the MBSL system with the SBSL system. The spectral characteristics of each system show a continuum emission, decreasing into the IR. When 0.1 M NaCl is added, sodium emission from the $3d-3p$ transition is observed in the MBSL system, but not in the SBSL system (increasing the NaCl concentration is precluded because of instabilities in SBSL). The NIR emission intensity was measured and found to vary linearly with drive pressure. However, no NIR emissions below the luminescence threshold were observed.

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