

## Conditions during Single-Bubble Sonoluminescence

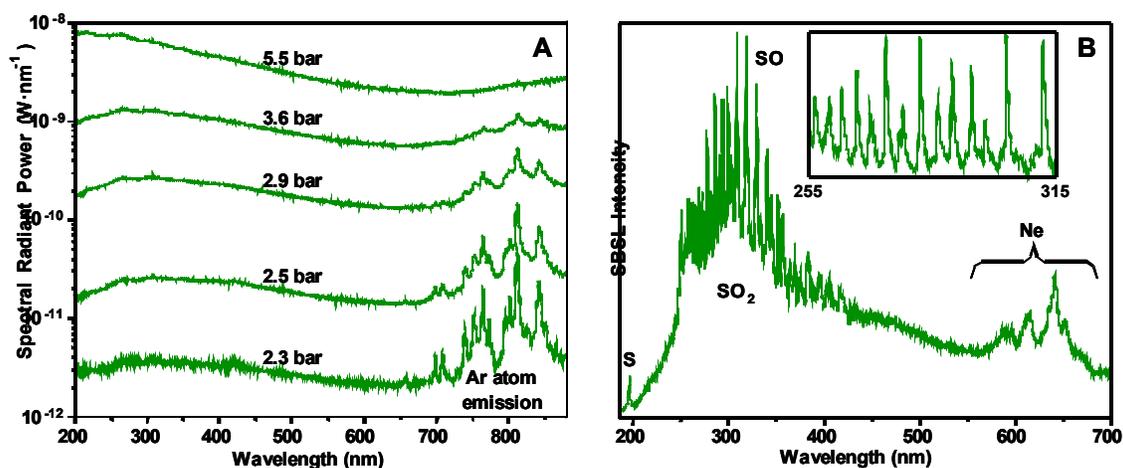
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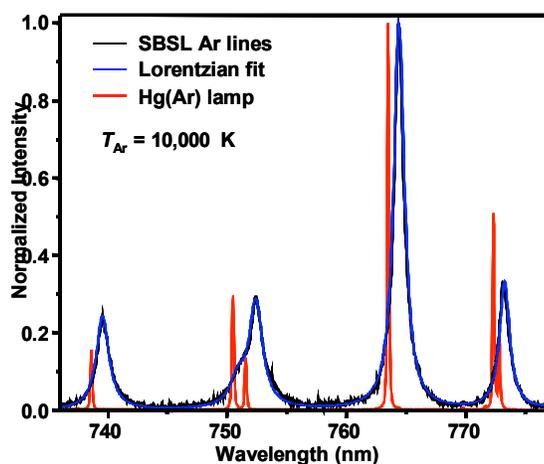
A single bubble can be levitated in a liquid and driven into highly nonlinear radial oscillation by application of a standing acoustic wave. The nonlinear oscillation of the bubble consists of a slow volume growth during the rarefaction phase of the acoustic cycle followed by a runaway implosion that is initiated well into the compression phase of the cycle. As the bubble collapses the contents are rapidly compressed and heated quasiadiabatically to the point that a subnanosecond flash of broadband light is observed (single-bubble sonoluminescence, SBSL).<sup>1,2</sup> If the net mass exchange across the interface is zero during the oscillation, the nonlinear radial motion will be repeated each and every acoustic cycle, and the light flash will be synchronous with the sound field having a phase shift of less than  $\pm 0.001\%$ .<sup>3</sup>

Though SBSL has been vigorously studied for some time, little is definitively known about the light-emitting mechanisms, intra-cavity physical conditions, and chemical processes. Spectroscopic studies of SBSL from water have revealed little more than featureless continua that increase in radiant power from the near-IR to the mid-UV.<sup>4</sup> The rapid bubble collapse,<sup>5</sup> narrow SBSL flash width,<sup>6</sup> and featureless spectra have led to speculation that the intra-cavity conditions generated near the point of maximum implosion may be extreme enough to support nuclear fusion reactions.<sup>7</sup> Though speculations of extreme conditions during SBSL are ubiquitous within the field, experimental evidence supporting even the generation of a plasma is lacking, and recent claims of the observation of acoustic inertial confinement fusion have been met with severe skepticism.<sup>8,9</sup> Remarkably, water continues to be the liquid most often used to generate and study SBSL in spite of the limited information gained.



**Figure 1.** (A) SBSL from degassed 85% H<sub>2</sub>SO<sub>4</sub> regassed with 40 torr Ar as a function of the applied acoustic pressure amplitude (bar). The Ar lines arise from transitions within the  $4p - 4s$  manifold. (B) SBSL from degassed 85% H<sub>2</sub>SO<sub>4</sub> regassed with 50 torr Ne. The Ne lines arise from transitions between states having energies of nearly 20 eV. The inset is an expansion of the region between 255 and 315 nm showing SO ( $B^3\Sigma^- \rightarrow X^3\Sigma^-$ ) emission bands.

In order to gain insight into the physical and chemical processes at work during SBSL, nonvolatile liquids such as concentrated sulfuric acid ( $\text{H}_2\text{SO}_4$ ) were studied. Liquids having negligible vapor pressure are attractive for SBSL studies because the bubble will be essentially devoid of molecular species; the energy of the bubble collapse will not be partitioned into molecular degrees of freedom, so the peak intra-cavity energy densities will be higher than for a relatively volatile liquid such as water.<sup>10,11</sup> The SBSL radiant powers from  $\text{H}_2\text{SO}_4$  aqueous solutions are well over  $10^3$  times larger than those typically seen from water.<sup>12</sup> In addition, the emission spectra contain extensive bands and lines from various molecules, atoms, and ions (Figure 1).<sup>13,14</sup> The observation of the population of high-energy states of atoms (20 eV) and ions (37 eV) provides definitive experimental evidence of the formation of a plasma during cavitation, and is key in supporting specific theories on the light-emitting mechanisms of SBSL. By using molecular and atomic probes, standard methods of plasma diagnostics, and spectrometric methods of pyrometry, quantification of the heavy particle temperatures,<sup>12</sup> heavy particle densities,<sup>15</sup> and plasma electron densities<sup>16,17</sup> generated during SBSL from  $\text{H}_2\text{SO}_4$  was achieved (Figure 2). It was found that for an Ar bubble in  $\text{H}_2\text{SO}_4$ , temperatures of 15,000 K, heavy particle densities of 4,000 atm, and electron densities comparable to laser-generated plasmas ( $10^{18} \text{ cm}^{-3}$ ) were achieved. These measurements indicated that while the conditions generated during SBSL from  $\text{H}_2\text{SO}_4$  were extraordinary, they were well in-line with the predictions put forth by classical bubble dynamics.<sup>18</sup>



**Figure 2.** SBSL Ar emission lines compared to a simulated spectrum with Lorentzian line-shapes and the Ar lines from a Hg(Ar) calibration lamp. Comparison of the relative intensities of the 763 and 738 nm lines results in an estimate of the heavy particle temperature of 10,000 K.

In addition to spectroscopic studies, far-field microscopic and stroboscopic studies of bubble dynamics were conducted in order to gain more insight into the effect of bubble dynamics on SBSL. By conducting simultaneous stroboscopic and spectroscopic studies on single-bubble cavitation in solutions of  $\text{H}_2\text{SO}_4$  containing sulfate salts of the alkali-metals, it was found that dramatic changes in the bubble dynamics led to emission from involatile species such as Na and K atoms.<sup>19</sup> The major mechanism producing these effects was attributed to the development of surface instabilities at the bubble interface with increasing translational velocity of the bubble.<sup>20</sup> The surface instabilities then resulted in the injection of droplets into the hot bubble interior where they were pyrolyzed during implosion, thus producing the observed effects.

## References

1. Brenner, M. P.; Hilgenfeldt, S.; Lohse, D. Single-Bubble Sonoluminescence. *Rev. Mod. Phys.* **2002**, *74*, 425-484.
2. Putterman, S. J.; Weninger, K. R. Sonoluminescence: How Bubbles Turn Sound into Light. *Annu. Rev. Fluid Mech.* **2000**, *32*, 445-476.
3. Barber, B. P.; Putterman, S. J. Observation of Synchronous Picosecond Sonoluminescence. *Nature* **1991**, *352*, 318-320.
4. Hiller, R. A.; Putterman, S. J.; Weninger, K. R. Time-Resolved Spectra of Sonoluminescence. *Phys. Rev. Lett.* **1998**, *80*, 1090-1093.
5. Pecha, R.; Gompf, B. Microimplosions: Cavitation Collapse and Shock Wave Emission on a Nanosecond Time Scale. *Phys. Rev. Lett.* **2000**, *84*, 1328-1330.
6. Gompf, B.; Günther, R.; Nick, G.; Pecha, R.; Eisenmenger, W. Resolving Sonoluminescence Pulse Width with Time-Correlated Single Photon Counting. *Phys. Rev. Lett.* **1997**, *79*, 1405-1408.
7. Moss, W. C.; Clarke, D. B.; Young, D. A. Calculated Pulse Widths and Spectra of a Single Sonoluminescing Bubble. *Science* **1997**, *276*, 1398-1401.
8. Taleyarkhan, R. P.; West, C. D.; Cho, J. S.; Lahey, R. T.; Nigmatulin, R. I.; Block, R. C. Evidence for Nuclear Emissions During Acoustic Cavitation. *Science* **2002**, *295*, 1868-1873.
9. Reich, E. S. Evidence for Bubble Fusion Called into Question. *Nature* **2006**, *440*, 132.
10. Didenko, Y. T.; Suslick, K. S. The Energy Efficiency of Formation of Photons, Radicals and Ions During Single-Bubble Cavitation. *Nature* **2002**, *418*, 394-397.
11. Flannigan, D. J.; Hopkins, S. D.; Suslick, K. S. Sonochemistry and Sonoluminescence in Ionic Liquids, Molten Salts, and Concentrated Electrolyte Solutions. *J. Organomet. Chem.* **2005**, *690*, 3513-3517.
12. Flannigan, D. J.; Suslick, K. S. Plasma Formation and Temperature Measurement during Single-Bubble Cavitation. *Nature* **2005**, *434*, 52-55.
13. Flannigan, D. J.; Suslick, K. S. Plasma Line Emission during Single-Bubble Cavitation. *Phys. Rev. Lett.* **2005**, *95*, 044301.
14. Flannigan, D. J.; Suslick, K. S. Molecular and Atomic Emission during Single-Bubble Cavitation in Concentrated Sulfuric Acid. *Acoust. Res. Lett. Onl.* **2005**, *6*, 157-161.

15. Flannigan, D. J.; Hopkins, S. D.; Camara, C. G.; Putterman, S. J.; Suslick, K. S. Measurement of Pressure and Density inside a Single Sonoluminescing Bubble. *Phys. Rev. Lett.* **2006**, *96*, 204301.
16. Flannigan, D. J.; Suslick, K. S. Plasma Quenching by Air during Single-Bubble Sonoluminescence. *J. Phys. Chem. A* **2006**, *110*, 9315-9318.
17. Flannigan, D. J.; Suslick, K. S. Electron Density of the Plasma Generated during Single-Bubble Cavitation. **2007**, (In preparation).
18. Hilgenfeldt, S. Sonoluminescence - Sound Basis for Light Emission. *Nat. Phys.* **2006**, *2*, 435-436.
19. Flannigan, D. J.; Suslick, K. S. Metal Atom Emission during Single-Bubble Sonoluminescence. *Phys. Rev. Lett.* **2006**, (Submitted).
20. Suslick, K. S.; Hammerton, D. A.; Cline, R. E. The Sonochemical Hot Spot. *J. Am. Chem. Soc.* **1986**, *108*, 5641-5642.