Energy Focusing in a Converging Fluid Flow: Implications for Sonoluminescence

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Cavitation induced by a converging flow leads to such a powerful concentration of energy that subnanosecond flashes of light with an ultraviolet spectrum are emitted. Light intensity is enhanced a factor of 5000 by using xenon rather than helium. This xenon enhancement is similar to that seen with surface bubbles and acoustically driven bubbles in nonaqueous fluids, but contrasts with acoustically driven bubbles in water where xenon is only 5 times brighter than helium. This system has been studied with the goals of obtaining sonoluminescence from diatomic gases and learning about those aspects of sonoluminescence which are properties of a single collapse.

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Cavitation is a prime example of energy focusing in the far off equilibrium motion of fluids. Flow energy injected at long wavelengths and low energy densities forms bubbles whose subsequent collapse focuses the energy by many orders of magnitude. Already in 1917 Rayleigh [1] studied cavitation in connection with the damage caused by bubbles to ships' propellers. More recently, the high quench rates achieved in acoustically driven cavitation have been used to synthesize new materials [2] and have been implicated in the facilitation of surgical procedures [3]. Measurements of the repetitive collapse of a single bubble in a water filled acoustic resonator reveal that energy can focus by over 12 orders of magnitude to create a clocklike string of picosecond flashes of ultraviolet light [4–7]: sonoluminescence (or "SL").

The most spontaneous form of cavitation (and energy focusing) occurs when a fluid is accelerated, such as when it is forced through the constriction of a venturi tube at velocities that create pressure drops in excess of one atmosphere [8–10]. We have revisited this form of cavitation in an attempt to gain insights into the physical mechanisms that underlie sonoluminescence [11]. We find that luminescence from cavitation induced by a converging flow is dramatically strongest for xenon gas [12]. In contrast with resonant SL, we find that in luminescence from a converging flow light emission from the other noble gases is down by orders of magnitude. However, we find that the spectrum (Fig. 1) and flash width [9] (Fig. 2 inset) of flow-induced cavitation match within experimental accuracy the properties of resonant SL. Also as found for resonant SL the intensity of light emission increases strongly as the ambient temperature of the water is decreased (Fig. 2).

Our overarching observation is that cavitation luminescence (at least for fluids that are partly degassed) divides into two categories: (i) systems which are greatly enhanced by the presence of xenon gas and (ii) isolated bubbles surrounded by water in a resonant sound field. In the category of xenon-facilitated luminescence sit the phenomena of flow-induced cavitation [9], resonant sonoluminescence from single bubbles in nonaqueous fluids [13], isolated "hemispherical" bubbles that spontaneously appear on the boundaries of resonators [14], and multibubble SL from a dipole sound source [14]. For an isolated bubble trapped at the velocity node of a resonant sound field in water the luminescence is strong for all noble gases [4]. We shall call this last case resonant or "clocklike" SL, since the gas concentration can be adjusted so that the flash times display a remarkable synchronicity that distinguish this case from all other cases [15].

A key aspect of flow-induced luminescence is that the bubbles form and collapse on a "one shot" basis. Therefore these measurements may provide clues as to which aspects

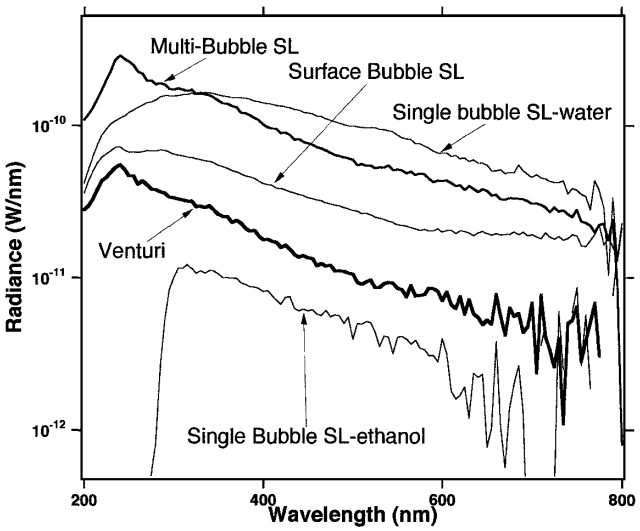

FIG. 1. Spectrum of light emitted from xenon bubbles in a venturi tube at 28 °C and 50 Torr partial pressure. Also shown for comparison are xenon spectra from surface bubble SL [14] (300 Torr, 12 °C), single bubble SL in ethanol [13] (325 Torr, −12 °C), single bubble SL in water [26] (3 Torr, 20 °C), and multibubble SL from saturated water (760 Torr, 16 °C) generated by the dipole sound field of the Contour Genesis ultrasonic-assisted liposuction device (Mentor Corporation) [14]. All data are acquired with the same equipment (10 nm FWHM resolution) and are corrected for grating and detector responses. The venturi and multibubble data are multiplied by an arbitrary factor for presentation on the graph.

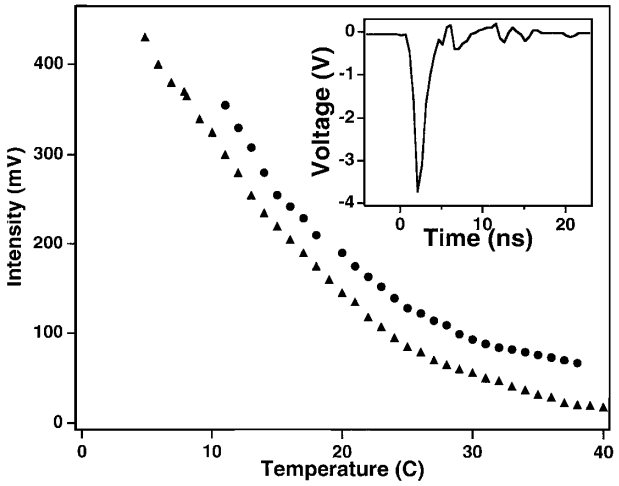

FIG. 2. Circles show the temperature dependence of the light emitted by venturi cavitation of water with 50 Torr xenon dissolved in. The flow entering the venturi is held constant at 1.26 m/s and intensity is recorded as rms current through 10 kOhms generated by a PMT observing the bubbles. The triangles show another run of 50 Torr xenon but with 100 ppm by volume 1-butanol mixed in water and a flow of 1.25 m/s. The intensity of light for single shot collapses does not appear to be diminished by drops of alcohol as it is for synchronous SL [13,27]. The increase in intensity as liquid temperature is decreased is the same seen in single bubble SL. Inset: Single shot response of a Hamamatsu H5783-03 PMT to the flash of light from bubble collapse in the venturi (50 Torr, 20 °C). The fall time for this 100 photoelectron event is 918 ps, and deconvolution with the typical single photoelectron event (700–800 ps) gives an upper bound on the venturi flash width as 550 ps.

of resonant sonoluminescence are ascribable to short time scales (~one acoustic cycle) or long time scales.

Experiments are carried out with a $\frac{3}{4}$ hp swimming pool pump that circulates water around a closed loop constructed from $\frac{1}{4}$ in. wall tygon tubing and polyvinylchloride (schedule 40) pipe of inner dimension (ID) $1\frac{3}{8}$ in. In the loop is a handblown quartz venturi tube with an entrance/exit of ID 34.5 mm tapering in at 40° full angle to a 5 mm long constriction of ID 9 mm and then tapering out at a full angle of 10°. Also in the loop are a flow meter, thermocouple, static pressure gauge, and heat exchanger. In parallel with the venturi tube in the loop was a bypass route with a variable flow restrictor, thus allowing the flow rate in the venturi to be varied for fixed pump speed. Maximum flow rates that could be achieved at the entrance to the tube were measured to be about 1.5 m/sec. Water was degassed and then mixed with the desired combination of gases in a separate container. The venturi loop was evacuated before the water was transferred in, and then returned to about one atmosphere static pressure and sealed.

At the highest flow velocities Bernoulli's law yields pressure drops in excess of an atmosphere. In this situation bubbles nucleate and expand as they pass through the constriction and then collapse as they pass again into the region of ambient pressure. Measurements were therefore carried out a few cm downstream from the constriction. For xenon gas dissolved into the water at a partial pressure of about 100 Torr, light is emitted from a 1 cm^3 diffuse cloud and individual pinpoints near the center, as seen by the unaided eye. A PMT (photomultiplier tube) was centered on the cloud for high flow rates. A piezoelectric ceramic microphone glued to the wall could correlate individual flashes with pulses of acoustic emission. Figure 3 shows the intensity of light emission as a function of flow rate for several concentrations of xenon in water. Flow-rate-induced variations in cloud location relative to the PMT were small, giving at most 20% corrections to a few low flow rate points.

The strongest light emission observed from argon is down by a factor of 50 from the weakest observed emission from xenon. When 50 Torr solutions are compared it is found that argon is down by a factor over 100 and helium is down by a factor of at least 5000 from xenon. We also found that D_2, O_2, and air all yield signals that are down from Xe by a factor of at least 5000, which places a possible signal (at 50 Torr) within our noise floor.

Since emission from a cavitating hydrodynamic flow depends not only on the strength of an individual bubble's implosion but also on the number of bubbles formed by the flow we have attempted to characterize the cloud of bubbles formed in these experiments. Figure 4 shows the counts recorded by a photomultiplier tube in a fixed interval of time due to laser light scattered from the collapse

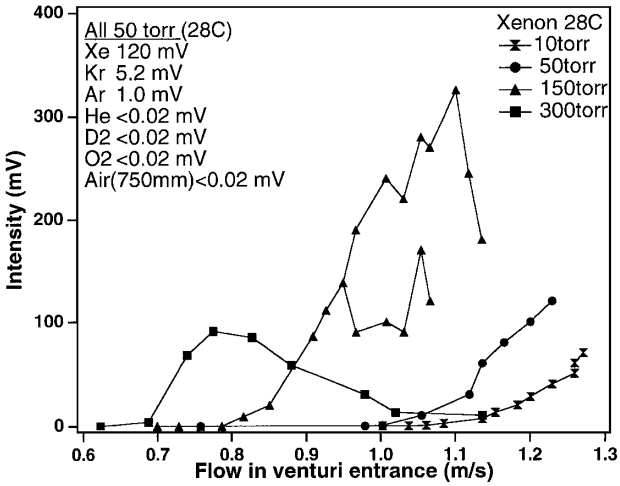

FIG. 3. Intensity as a function of flow rate for venturi cavitation with xenon in water at 28 °C and various levels of saturation. At low concentration we cannot rule out the possibility that the intensity continues to increase with flow rate. Although the 150 Torr system is the brightest, it was undesirable because on the scale of seconds it would alternate between two states of different intensities at a fixed flow. Shown as a table are the maximum intensity for various gases at 50 Torr and maximum flow. For helium, deuterium, oxygen, and undegassed water any possible signal was not detectable over our noise floor (0.02 mV).

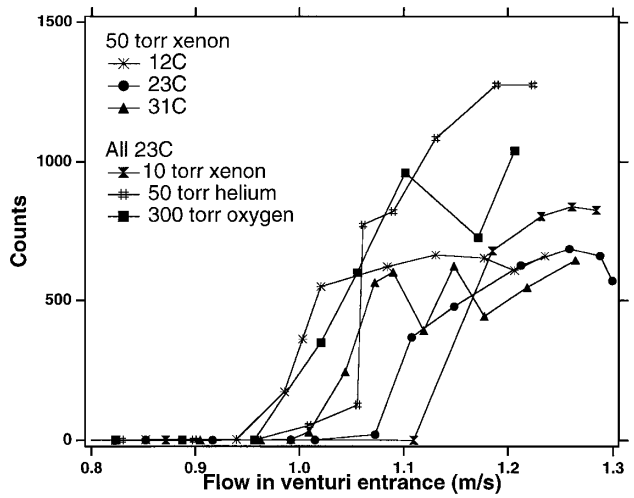

FIG. 4. Laser scattering to characterize the bubble cloud generated by flow in the venturi with various gases dissolved in the water. A 1 mW HeNe laser is passed through the center of the bubble cloud. Light scattered out of the beam at 90° is collected by a PMT (attenuated with neutral density filters to single photon level) and time traces were recorded by an oscilloscope. Individual scattering events are counted for 650 μs and the average of 20 such runs is plotted.

region. Flows with dissolved He and O_2 nucleate about the same number (or slightly more) bubbles than 50 Torr mixtures of Xe in water. Nevertheless light emission from He and O_2 is down by more than a factor of 100 from the 10 Torr Xe mixtures. Similarly 50 Torr Xe at 12 °C and 23 °C nucleate comparable clouds of bubbles. It appears that variations in light emission relate to the properties of individual cavitation events. This conclusion is also supported by measurements of the pulse height distribution by Peterson and Anderson [9].

Mixtures of xenon and oxygen have been studied with the goal of determining whether flow cavitation displays a noble gas doping effect [16]. As shown in Table I small concentrations of Xe yield correspondingly smaller light emission intensities in contrast with clocklike SL, where a 1% Xe concentration already yields an emission that is about equal to a pure Xe bubble [4]. It remains to be seen whether the absence of a noble gas doping effect is a property of luminescence from a single collapse or whether it carries over to the other cases of xenon-facilitated luminescence, such as resonant SL from an isolated bubble in a wide range of *nonaqueous* fluids.

TABLE I. Maximum intensity (mV) for mixtures of xenon in oxygen from venturi cavitation at 28 °C for 50 and 300 Torr total saturation of water.

Xenon in oxygen	50 Torr	300 Torr
0% xenon	0.02 mV	0.5 mV
20% xenon	3.3 mV	16 mV
100% xenon	120 mV	92 mV

One of the striking features of clocklike SL is its sensitivity to ambient temperature; the maximum acoustic drive and light emission increase as the temperature is lowered [17]. This upper threshold of SL has been interpreted in terms of a shape instability that builds up over many cycles via a (parametric) resonance [18–20]. While shape instabilities [21] such as bubble pinch-off [15] play a role in SL, the observation of a light emission that is strongly dependent on temperature for single shot cavitation suggests that the upper threshold may not be determined by an instability that requires many cycles to build up [22].

According to the (adiabatic) hot spot theory of cavitation of SL greater emission from Xe can be ascribed to its lower thermal conductivity [10]; according to the shock wave theory [23,24] the lower speed of sound would enhance the implosion; according to the plasma picture its low ionization potential would enhance the bremsstrahlung [4,23]. These processes apply to clocklike SL and venturi tube luminescence. Although Xe is strongest in both cases its emission exceeds He by over a factor of 5000 in flow cavitation and by only a factor of 5 for clocklike SL.

Xenon has a larger solubility in water than does helium (as well as most other gases) which could lead to larger (transient) bubbles when xenon is dissolved in the water as compared to helium. To address these issues we used a 0.5 mm ID needle inserted parallel to the flow about 1.5 in. upstream from the constriction to introduce bubbles of known gas content (from a gas reservoir with a solenoid valve) into a flow of highly degassed water. With a microscope these bubbles were found to have a radius of about 0.3 mm upon leaving the needle. The single flashes of light detected by a PMT beyond the constriction from xenon were 1.5 times larger than krypton, and 200 times larger than argon. We detected no light after injecting helium bubbles, although a microphone glued downstream of the constriction registered similar responses for all of the collapsing bubbles. Diffusive bubble growth does not seem to explain the various light intensities since for a gas bubble injected into degassed water the high solubility of xenon in water should cause the bubble to dissolve more than helium bubbles before they reach the collapse region. The opposite is true of bubbles spontaneously cavitated in partially saturated water, although the trends in light intensity as a function of gas species are the same.

One of the surprises of clocklike SL is the poor behavior of pure diatomics [4,25] (O_2, N_2 are dim and unstable, whereas H_2 can be stabilized at low drive levels). We now see that in regards to luminescence from a single pass not only the diatomics but also the lighter noble gases are poor light emitters. An explanation could be sought in the possibility that cavitating bubbles have radii and expansion ratios (the ratio of maximum to ambient bubble radius) that place them right at the

threshold of light emission so that sensitivities such as outlined above are magnified. Such an explanation appears to be challenged by the observation of similar spectral densities [26] for the xenon bubbles in each experimental arrangement (see Fig. 1). In any case, future experiments should measure the size and distribution of cavitating bubbles, as the data of Fig. 4 respond to a combination of these parameters.

In conclusion we have verified that the spontaneous focusing of energy that is achieved for flowing water (with dissolved xenon) [9] is comparable to that achieved for clocklike SL. If we are justified in interpreting luminescence from flow cavitation as being due to a single bubble collapse then various insights can be gained with regard to clocklike SL. First, observation of similar temperature dependence suggests that the maximum intensity of SL is limited by a physical process that takes about one (acoustic) cycle to set in. This challenges theories that account for the temperature dependence of SL in terms of a parametric shape instability [19] and diffusional changes in bubble size [28]. Second, the relative intensities observed for He, Ar, and Xe in the xenon-facilitated cases are consistent with the weak degree of ionization expected from compressive heating to about 20 000 K. This highlights the contrast with clocklike SL, where He emission is nearly as strong as Xe, perhaps indicating a much higher temperature and a strong degree of ionization.

Another means of creating bubbles on a one shot basis is provided by laser generated cavitation [29]. Should this phenomenon fall into the category of luminescence facilitated by heavy noble gases, then scaling to the results reported in this paper would suggest that the execution of those experiments with xenon instead of air would lead to a signal that is greatly enhanced. Finally there remains the highly practical form of cavitation from gas saturated chemical solutions driven by cell disruptors [30]. Its precise relation [31] to the cases studied here requires further study. Perhaps accelerating flows will yield a means of scaling up industrial processes that rely on cavitation.

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[1] Lord Rayleigh, Philos. Mag. **34**, 94 (1917).
[2] K. S. Suslick *et al.,* Nature (London) **353**, 414 (1991).
[3] M. Topaz, Aesthetic Surgery **18**, 19 (1998).
[4] B. P. Barber *et al.,* Phys. Rep. **281**, 65 (1997).
[5] S. Putterman, Sci. Am. **272**, No. 2, 32 (1995).
[6] S. Putterman, Phys. World **11**, 42 (1998).
[7] L. Crum, Phys. Today **47**, No. 9, 22 (1994); B. Gompf *et al.,* Phys. Rev. Lett. **79**, 1405 (1997).
[8] P. D. Jarman and K. J. Taylor, Br. J. Appl. Phys. **16**, 675 (1965).
[9] F. B. Peterson and T. P. Anderson, Phys. Fluids **10**, 874 (1967).
[10] A. J. Walton and G. T. Reynolds, Adv. Phys. **33**, 595 (1984).
[11] Contemporaneous responses to this work were negative: R. Hickling, Phys. Fluids **11**, 1586 (1968); see also Ref. [6].
[12] Experiments with radon have not been attempted.
[13] K. Weninger *et al.,* J. Phys. Chem. **99**, 14 195 (1995).
[14] K. Weninger *et al.,* Phys. Rev. E **56**, 6745 (1997); K. R. Weninger, C. Camara, and S. Putterman (to be published).
[15] B. P. Barber *et al.,* Phys. Rev. Lett. **74**, 5276 (1995); B. P. Barber *et al.,* J. Acoust. Soc. Am. **91**, 3061 (1992).
[16] R. Hiller *et al.,* Science **266**, 248 (1994).
[17] B. P. Barber *et al.,* Phys. Rev. Lett. **72**, 1380 (1994).
[18] S. Putterman and P. H. Roberts, Phys. Rev. Lett. **80**, 3666 (1998); M. P. Brenner *et al.,* Phys. Rev. Lett. **80**, 3668 (1998).
[19] S. Hilgenfeldt *et al.,* Phys. Rev. Lett. **80**, 1332 (1998); S. Hilgenfeldt *et al.,* Phys. Rev. Lett. **80**, 3164 (1998).
[20] C. C. Wu and P. H. Roberts, Phys. Lett. A **250**, 131 (1998); Phys. Fluids **10**, 3227 (1998).
[21] G. Holt and F. Gaitan, Phys. Rev. Lett. **77**, 3791 (1996).
[22] Other authors have scaled vapor pressure to the effects of changing ambient temperatures, see Ref. [10].
[23] C. C. Wu and P. H. Roberts, Phys. Rev. Lett. **70**, 3424 (1993); Proc. R. Soc. London A **445**, 323 (1994); Phys. Lett. A **213**, 59 (1996); Q. J. Mech. Appl. Math. **49**, 501 (1996).
[24] W. C. Moss *et al.,* Phys. Fluids **6**, 2979 (1994); H. P. Greenspan and A. Nadim, Phys. Fluids A **5**, 1065 (1993).
[25] D. Lohse and S. Hilgenfeldt, J. Chem. Phys. **107**, 6986 (1997).
[26] These experiments use an SL spectral facility set up at UCLA by R. A. Hiller, Ph.D. thesis, UCLA, 1996.
[27] R. Hiller *et al.,* Phys. Rev. Lett. **77**, 2345 (1996).
[28] V. Q. Vuong *et al.,* J. Acoust. Soc. Am. **104**, 2073 (1998).
[29] C. D. Ohl, O. Lindau, and W. Lauterborn, Phys. Rev. Lett. **80**, 393 (1998).
[30] K. S. Suslick and E. B. Flint, Nature (London) **350**, 553 (1987); K. S. Suslick *et al.,* J. Am. Chem. Soc. **119**, 9303 (1997).
[31] T. J. Matula *et al.,* Phys. Rev. Lett. **75**, 2602 (1995).