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LUMINESCENCE FROM CAVITATION BUBBLES

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Abstract: *Luminescence from laser-produced cavitation bubbles in water is investigated experimentally. The main parameters of this study are the maximum bubble radius and the ambient pressure of the liquid surrounding the bubble. The luminescence pulse was determined using a very fast photo-multiplier-tube (PM) and the maximum temperature of the bubble was determined using a spectrum analyzer. The results indicate that the luminescence pulse is of the order of nanoseconds while the maximum temperature of the bubble at the minimum volume is about 9000 K.*

Key words: *cavitation, laser-induced bubbles, luminescence.*

1. Introduction

The mechanism which produces luminescence pulses from collapsing bubbles is still not well understood. The adiabatic compression of the gas in the bubble produces fairly high temperatures, enough to ionize a small fraction of the gas, but the details of exactly how the light is produced is not yet very clear. There are also significant unexplained differences between multi-bubble and single-bubble sonoluminescence (MBSL [6] and SBSL [5]). To try to understand better the nature of the light emission, we have undertaken studies of the light emission from freely-collapsing bubbles in water and in cryogenic liquids. The bubbles are created from a focused laser pulse, which allows the study of bubbles whose maximum radius is between 0.2 and 2 mm, considerably larger than the 0.05 mm maximum radius of SBSL bubbles. This phenomenon of luminescence from laser-induced bubbles (LIBL) was first discovered in Russia in 1971 [4], and then after the advent of SBSL it has been considerably further investigated [1-3], [7], [8].

2. Experimental

To create the bubbles a Nd:YAG laser producing 6 ns pulses with a maximum energy of 600 mJ at 1064 nm is collimated and focused to a point about 10 μm in diameter in the sample liquid, as previously described [1-3]. The laser energy is adjusted to the threshold for just creating a single bubble, typically of order 100 mJ. Absorption of the laser energy is initiated by an impurity in the liquid, and hence the bubble size varies with each laser shot, depending on the impurity size. For the water sample the sealed stainless-steel cell shown in Figure 1 has quartz windows for monitoring the emitted light with a photomultiplier, and the radius of the bubble versus time can be monitored by a shadowgraph technique and by

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pulsed-laser photography through a long-distance microscope. Calibration of the radius is found by measuring the time to the collapse point and comparing with Rayleigh's equation for the collapse time of a vacuum bubble. The luminescence is monitored with two fast PMT's with rise times of about 1.2 ns, both of which can fitted with filters in front to pass different spectral regions. The luminescence can also be sent to a 0.3 m spectrum analyzer for measurements in the visible wavelength range 250-700 nm. Since our measurements showed that dissolved air in the high-purity deionized water sample had no effect on the luminescence [1], we do not degas the water, and dry nitrogen gas is used to pressurize the sample.

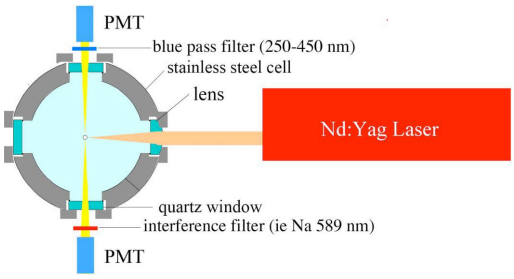

Fig. 1. *Experimental apparatus for LIBL measurements*

The laser pulse ionizes the liquid in a volume about 10 μm in diameter, and this grows to a hot plasma 50-100 μm in diameter with a temperature of order 16,000 K [2] that glows for 50-100 ns before recombining. The energy absorbed from the laser then appears as the energy of an expanding bubble. The details of the gas in the bubble are not known with any certainty, but it is likely to primarily be the recombined atoms making up the liquid. For water this would be atomic hydrogen and oxygen, and since the bubble collapse time is less than 200 μs and the gas pressure is very low over most of that time, it is unlikely that there will be any appreciable molecular recombination. There will also be water vapor at the saturated pressure of about 25 torr in the bubble.

The spectrum of the luminescence is measured by collecting the light with paraboloidal mirrors, and focusing onto the slits of a 0.3 m spectrometer. An intensified CCD detector reads out the light intensity over a 200 nm range for each bubble, with a resolution of about 1 nm. It is necessary to average over 25 bubbles to get a reasonable signal to noise ratio in the spectra, where only bubbles in a given range of radii 0.65 ± 0.05 mm in the present measurements are included in the average. The measured spectra are divided by a calibration curve to account for the absorption in the optical path and the quantum efficiency of the detector, derived by illuminating a pinhole at the bubble position with quartz and deuterium lamps. There should be little optical change in the windows and the water as pressure is applied, and the same calibration curve is used for all the pressures.

3. Results and Discussion

Figure 2 shows the growth and collapse of a bubble in ambient 1 bar water after it is created with the laser pulse, as monitored with the shadowgraph technique. A sharp luminescence pulse is observed precisely at the collapse point of the bubble. The further bubble oscillations are quite small, since the acoustic shock emission at the collapse point radiates away much of the bubble energy. Figure 3 shows a photograph of the bubble collapse taken with a series of backlighting laser pulses, and the bright spot at the middle is the luminescence. The size of the hot spot producing the luminescence is about 25 μm. We also note that the bubble keeps the spherical symmetry until the very late stages of its collapse.

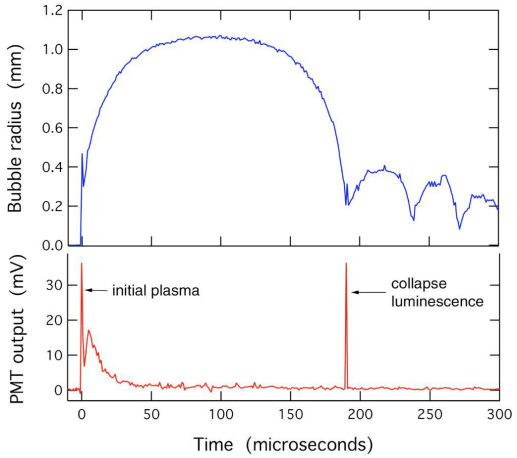

Fig. 2. *Bubble radius as a function of time and the light output recorded by a PMT*

We find that the duration of the emitted luminescence pulse is several nanoseconds [1-3], and that it increases linearly as the bubble size increases, shown in Figure 4. This is quite consistent with the shorter pulse widths (200-300 ps) found for the smaller bubbles of SBSL (shown as the open symbol). As also shown in Figure 4, the pulse duration increases with increasing hydrostatic pressure applied to the sample [3]. Fits to the pressure dependence show that the slope of the fit lines in Figure 4 varies as a power law of the pressure, $p^{0.37}$. This is true both for small and larger values of the hydrostatic pressure.

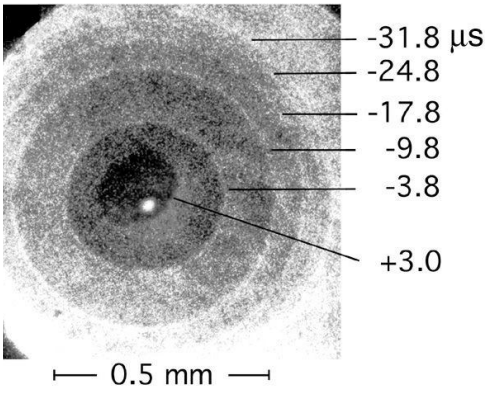

Fig. 3. *Photograph of a collapsing bubble at different times from the collapse point*

Figure 5 shows the number of photons emitted in the luminescence pulse, which appears to vary as the square of the bubble size. Unfortunately, none of these results have been explained by theory. One might have expected the pulse duration to decrease with increasing pressure, since the solutions of the Rayleigh-Plesset (RP) equation governing the bubble wall motion have faster dynamics as the pressure increases and the bubbles are smaller.

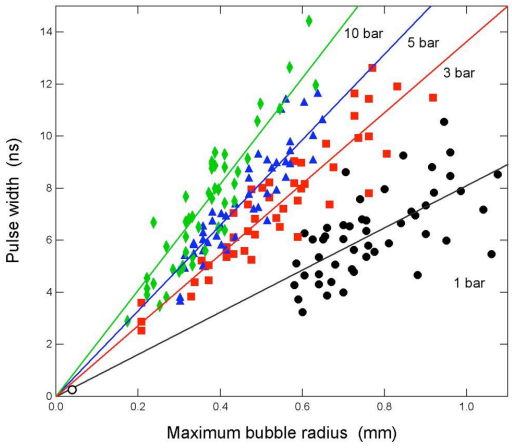

Fig. 4. *Luminescence pulse width as a function of the maximum bubble rdius and the applied hydrostatic pressure*

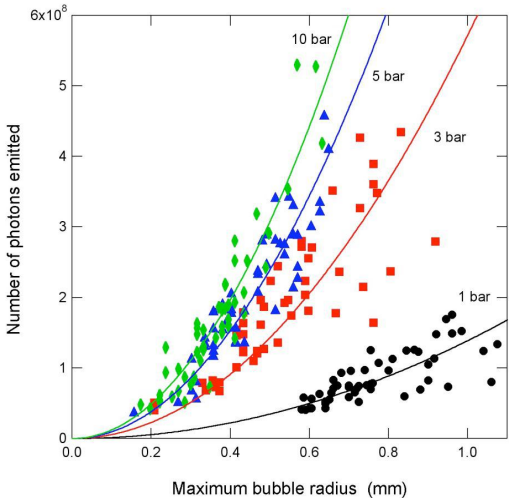

Fig. 5. *Number of photons emitted as a function of the maximum bubble radius, with square-law fits*

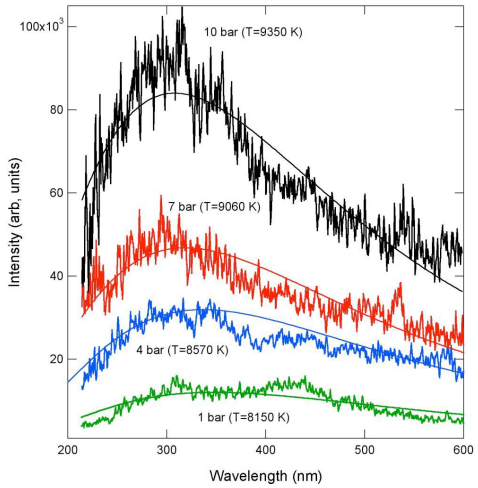

Fig. 6. *Spectrum of luminescence from bubbles with radius 0.6-0.8 mm*

Analysis of the spectrum of the luminescence shows reasonable fits to a blackbody spectrum at about 8000 K for bubbles with a maximum radius in the range of 0.6-0.8 mm at 1 bar ambient pressure [2], [3]. It is necessary to average the spectrum over 25-50 bubbles to get sufficient signal for the spectrometer, even with an intensified CCD detector. With applied hydrostatic pressure this temperature increases somewhat [3], to about 9400 K at 8 bars, as shown in Figure 6. At pressures above this the temperature levels off and even decreases above 12 bars, but it is likely that this is the result of instabilities in the bubbles where they split into two during the collapse [1-3].

Figure 7 shows that as the bubble radius is increased above about 1 mm, a new factor is observed [2] superposed on the blackbody spectrum: a peak at 310 nm that is probably associated with molecular band emission of an excited state of the OH molecule. By using a 310 nm interference filter on one of the PMT's, we found that this OH* emission is coincident in time with the blackbody emission to within our resolution of 1 ns. A similar peak is also seen in MBSL [6] and in SBSL when the bubble is unstable. The origin of the OH* band is not clear for any of these systems, though it may be associated with bubble instabilities, which is probably the only feature common to all of them. We find an increase in the number of bubbles with an unstable collapse as the radius is increased above 1 mm, and although we do not include such bubbles in the spectral data set (these bubbles usually show two luminescence flashes since the bubble has split into two [1-3]), but the unstable bubbles may not always be readily identifiable.

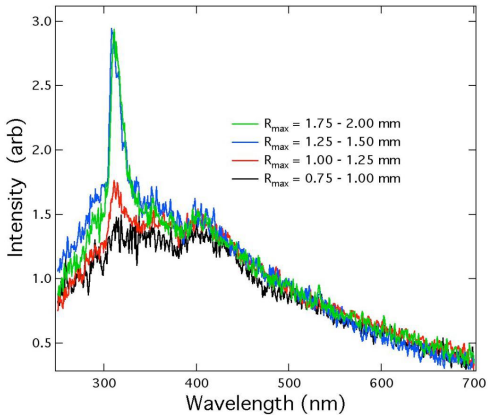

Fig. 7. *Luminescence from larger bubbles, at 1 bar*

To investigate the effect of the viscosity of the liquid on the luminescence, we have undertaken studies in water-glycerol mixtures. Pure glycerol has a viscosity about 1400 times that of water, so mixtures with water enable a wide range of viscosity, with only secondary variations in other parameters such as density and surface tension. We find that the bubble luminescence in the mixtures displays very similar properties to that found in pure water: the pulse duration is proportional to the maximum bubble size, and increases with applied pressure. Figure 8 shows the slope of the linear fits to the pulse duration versus bubble size as a function of the viscosity of the mixtures and the pressure. The pulse

duration increases by more than a factor of two as the viscosity increases to 4 cP (about 4 times the viscosity of water, a mixture of 33% glycerol by volume). At viscosities above that the pulse duration remains constant up to viscosities of 15 times that of water (a 60% concentration of glycerol). The probability of a bubble emitting luminescence at this concentration has fallen drastically from the lower concentrations, however, and by 75% glycerol concentration we can no longer see any luminescence at all, even though bubbles are still being generated by the laser.

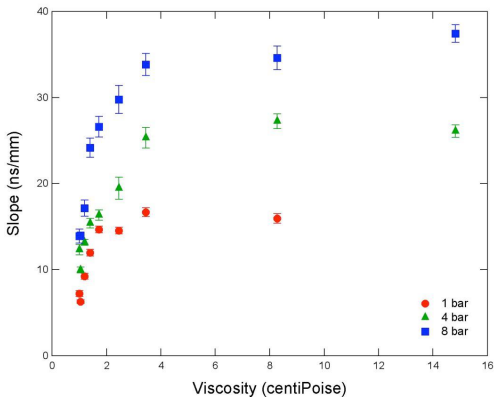

Fig. 8. *Slope of the pulse duration versus bubble size, as a function of viscosity of the mixtures and the pressure*

There is no theory for this observed doubling of the pulse duration with viscosity, similar to the lack of theory for our previous observations such as the linear increase of the duration with bubble size. The problem is that it is quite difficult to model the details of the bubble motion in the region of the collapse point, since at that point there is a very rapid increase in the temperature, pressure, and density of the gas in the bubble, and the bubble wall undergoes very high acceleration. It is very doubtful that the simple RP equation is adequate to model the behavior on the nanosecond time scales of the luminescence emission. One can guess, however, that the probable effect of viscosity is to somewhat slow down the motion of the bubble wall in the vicinity of the collapse point. If there is still enough compression of the gas to high temperatures, then slower dynamics at the collapse point might keep the temperature higher for a longer time, resulting in the longer pulse durations.

We also note that with increasing viscosity both the size and duration of the bubble oscillations increases, which may indicate that the dynamics at the collapse point are slowed by viscosity, since this will mean that less energy is lost to acoustic shock emission. With a further increase in viscosity, the oscillations then start to decrease in amplitude as the direct viscous damping becomes dominant, and the oscillations become more sine-like.

When alternative energy sources are mentioned, the image of bubbles rarely comes to mind. Yet two closely related phenomena, sonoluminescence (SL) and laser-induced luminescence (LICL), may shift the spotlight onto bubbles and their potential use in fusion, a promising energy source requiring the union of two nuclei, which in turn requires millions of Kelvin to occur. In SL, an acoustic field forces a bubble to oscillate in sync with the field and in doing so, the bubble emits light. In LICL, it is a laser beam that induces the growth and collapse of a bubble, upon which light is emitted as well. Compared to SL, LICL yields greater light intensity and allows for a spherical bubble collapse, which may prove useful in understanding the nature of cavitation luminescence. The remarkable ability of LICL to focus energy into a tiny volume on the micrometer scale makes it a pleasantly unexpected candidate for fusion techniques.

Of course, 9400 K (~9700 degrees Celsius) may seem scorching to a person, but that temperature is nowhere near the millions of Kelvin required for fusion. Furthermore, much work remains to be done in

understanding the precise bubble dynamics and mechanism of light emission before trying to engineer a bubble fusion plant. Nonetheless, these experiments can be modified in myriad ways that may enhance the force of bubble collapse. For instance, scientists could experiment with more volatile solutions to make the collapse of the bubble even more violent. An even more drastic change would be to use multiple lasers firing slightly asynchronously to induce collapsing concentric bubbles, thus making the phenomena even more forceful. In this sense, the versatility and energy concentration power of luminescence make it a promising choice for fusion energy.

4. Conclusions

The behaviour of the luminescence pulse emitted during the collapse of cavitation bubble is investigated experimentally. The present results indicate that the luminescence pulse emitted during the collapse of single laser-produced cavitation bubbles in water is of the order of nanoseconds. The temperature of the bubble at the minimum volume fits the blackbody radiation spectrum and is about 9000 K. The present experiments give insight into the nature of the luminescence, but, however, a number of features are still not explained by theory.

Although our investigations shed light on the behaviour of the luminescence pulse emitted during the collapse of single cavitation bubbles, the complex behavior of the bubble during the very late stages of its collapse is not yet fully explored. Numerical simulations of the bubble collapse could yield a valuable contribution to a better understanding of the bubble behavior by providing pressure contours and velocity vectors inside the bubble kinetic energy of the gas which are not easily accessible through experiments. For example, noncondensable gas might also be produced by plasma recombination at the initial stage of bubble expansion after laser-induced breakdown and by chemical reactions during all the period of bubble dynamics. Thus, to clarify the problem of non-condensable gas presence in a laser-induced cavitation bubble, plasma recombination phenomena, and chemical reaction kinetics should be investigated carefully.

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