PHYSICAL REVIEW E VOLUME 58, NUMBER 3 SEPTEMBER 1998

Measured pulse width of sonoluminescence flashes in the form of resonance radiation

Asis Giri and Vijay H. Arakeri*
Department of Mechanical Engineering, Indian Institute of Science, Bangalore 560 012, India
(Received 7 May 1998; revised manuscript received 14 July 1998)

Recent studies have shown that the measured flash widths from single and multibubble sonoluminescence are in subnanosecond or even picosecond regime. Here, we provide conclusive evidence for the existence of nanosecond multibubble sonoluminescence. This has become possible by our ability to find a medium from which exclusive sodium *D* line resonance radiation as a form of sonoluminescence is possible. The measured flash width of this emission is found to be in the range of tens of nanoseconds and is sensitively dependent on experimental parameters. Our finding is important since all the earlier pulse width measurements have been limited to emission with the physical source or species responsible for observed optical radiation not being clearly identified. We propose that the presently observed resonance radiation is from ''soft'' bubble collapse as analyzed by V. Kamath *et al.* [J. Acoust. Soc. Am. **94**, 248 (1993)]. [S1063-651X(98)51709-6]

PACS number(s): 78.60.Mq, 43.25.+y

One of the striking features of single-bubble sonoluminescence (SBSL) has been its measured and estimated optical pulse widths being in the picosecond regime [1]. More recent studies have shown that the timing characteristics of other types of sonoluminescence (SL) flashes are also similar to that of SBSL [2,3]. One common aspect to these experimental observations is that they have all been associated with light emission, which is known to have a broadband character [3–5]. Since the pulse width of SL flashes is now considered to be a key parameter in understanding the physics of SL, it occurred to us that if this parameter is measured for SL where the emitting species is clearly identified, it could be considered to be a significant advance in the subject. With this motivation we looked for a medium from which SL in the form of exclusive line emission is possible. From a careful study of the existing SL spectra measured by previous investigators it appeared to us that multibubble SL in the form of alkali-metal line emission from an appropriate nonaqueous medium could be a potential candidate [6]. Our expectations were proven to be correct, when we visibly observed (in a darkened room) golden yellow sodium *D* line emission from acoustically excited medium of argon saturated 1*N* ethylene glycol sodium chloride solution. We find SL from this medium to be relatively very bright; the estimated maximum number of photons per burst being 4×10^6, which is almost 20 times the standard SBSL intensity at room temperature, and this proved to be a major advantage in our studies. Low resolution spectra of the presently observed SL with salt solution is shown in Fig. 1 and it is clear that the emission consists exclusively of sodium *D* line resonance radiation [7]. The most significant finding of our work is that the pulse width of SL flashes in the form of sodium emission is of the order of tens of nanoseconds (Fig. 2).

Sonoluminescence in the present work was generated in room temperature (24 °C) 100 ml samples contained in a 250 ml quartz beaker acoustically excited by a single hollow cylindrical piezoelectric crystal attached to the bottom. The transducer was driven at near 32 kHz, being one of the resonant frequencies of the system, using instrumentation described elsewhere [8]. The spectra was measured by allowing the emission to go directly into varying width entrance slit of a 0.25 m scanning monochromator and synchronous SL was detected and processed using a RCA (Burle) 4526 photomultiplier tube (PMT) connected to a lock-in amplifier (SRS530) and PC-based data acquisition system. During the spectra measurements with argon saturated salt solution, continuous sparging of Ar (purity better than 99.99%) was maintained. The wavelength calibration of the monochromator was done using a low pressure sodium lamp, a mercury lamp, and a He-Ne red laser. The acoustic pressure amplitude P_a quoted are the maximum values and were measured using a miniature PCB(105A03) pressure transducer in degassed samples to avoid cavitation on the transducer surface. The ambient pressure at the location where experiments have been carried out is $\simeq$0.9 bar.

There was definite concern whether the presently measured relatively long SL flash durations are related to the fact that our observations are in a multibubble environment, since under these conditions there is a possibility that the observed

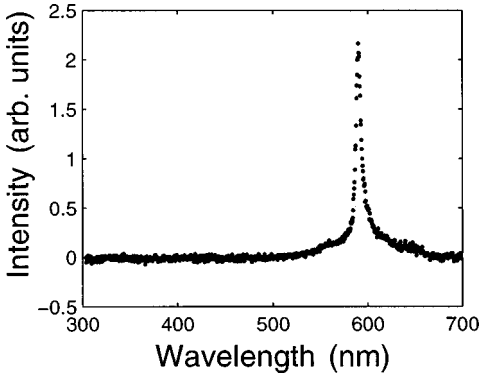

FIG. 1. Low resolution (3 nm full width at half maximum) spectra of SL from an argon saturated salt solution. The spectra shown is the average of ten 9600 s individual spectra obtained with a 1 mm slit width.

*Author to whom correspondence should be addressed. Electronic address: vijay@mecheng.iisc.ernet.in

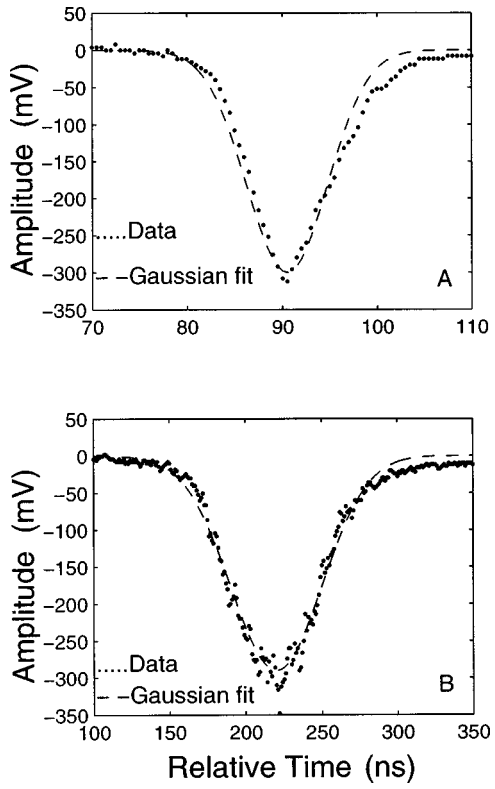

FIG. 2. Real time single shot SL pulse data recorded using a conventional PMT with a rated rise time of 1.7 ns and a 2 GHz real time digital sampling oscilloscope (Tektronix TDS 744A). (a) High drive level, pulse width=10 ns; (b) low drive level ($P_a \simeq 0.93$ bar), pulse width=70.6 ns. The pulse widths are estimated from a Gaussian fit to the recorded data.

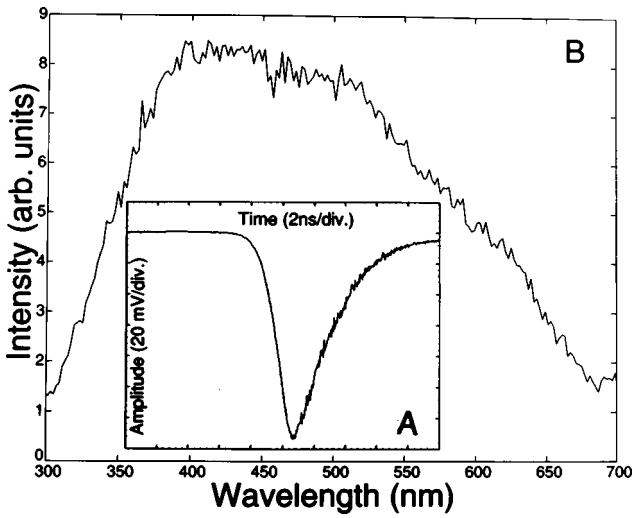

FIG. 3. (a) Pulse shape and (b) low resolution spectra of SL from air saturated ethylene glycol. The SL pulse shape shown is recorded using optical instrumentation indicated in the caption of Fig. 2 but with scope operated in repetitive sampling mode. The measured rise time is 1.92 ns and is indicative of PMT response to SBSL flash. The spectra shown is average of eight 9600 s individual spectra obtained with slit width of 2 mm and is corrected only for the PMT response.

pulse shapes are as a result of convolution or superposition of optical signal from a large number of SL events. Matula, Roy, and Mourad [2] have discussed this at length and have suggested certain techniques to ensure that the measured optical pulse width corresponds to a single flash even under a multibubble SL (MBSL) environment. We followed the precautions suggested by them and in addition carried out SL pulse width measurements in the ''few-bubble'' regime of MBSL, where the above mentioned difficulties should not arise. In an earlier study, Matula *et al.* [9] have suggested ''few-bubble sonoluminescence'' as an interesting area for examination, and our present results shown in Fig. 2(b) and later, on spectra measurement do support this. Following Matula, Roy, and Mourad [2], we first calibrated our optical instrumentation with SBSL in degassed water and the observed SBSL pulse rise time was 1.89 ns. Further, as an additional calibration in the MBSL environment we measured the rise time of the SL flash from air saturated ethylene glycol, and as shown in Fig. 3(a), it is indicative of the PMT response to SBSL flash. SL in air saturated ethylene glycol had bluish appearance and its spectra was broadband [Fig. 3(b)]. With the sodium salt solution, the SL pulse width, at high drive level, was measured by the use of a 50-mm focal length lens along with 1-mm-diameter aperture in front of the PMT; this was done to isolate a small region of SL activity [2]. At low drive level there was no need of lens-aperture combination, since the cavitation activity itself was limited to a small region near the acoustic pressure maximum. It is significant to point out that the measured pulse width in the ''few-bubble'' regime is substantially higher [10] than that with a high drive level [compare results in Figs. 2(a) and 2(b)]. This fact is a clear indication that our pulse width measurements do correspond to single flash events, since if the multiflash environment did influence the measurements, then we would have expected the pulse width to decrease as the bubble population density is reduced. We would like to further point out that there is no basis for suggesting here that the few bubble SL regime studied by us closely resembles the classical SBSL, since the methodology required for establishing the latter [9] is quite different from the conditions used by us.

We have, in addition, carried out observations of sodium line emission with helium gas instead of argon and we find comparable results in all aspects, including pulse timing characteristics. Further, we find that SL flash widths of potassium line emission (obtained using potassium salt) are also comparable to those with sodium line emission. Therefore, nanosecond pulse duration seems to be characteristic of SL in the form of alkali-metal line emission. These time scales are not a measure of radiative lifetime of excited alkali-metal atoms, since, as pointed out by Flint and Suslick [6] on the basis of observed line broadening ($\simeq$4 nm), the lifetime is expected to be of the order of 0.05 ps. Therefore, the only logical explanation for the measured long flash durations is that they are a measure of time span during which conditions are favorable inside a bubble for resonance radiation. This view is supported by the considerations indicated below.

What we have presently observed with argon saturated sodium salt solution is one of the ''cleanest'' forms of SL, where the physical source or species for light emission is identified. The regime of ''few'' bubbles is particularly interesting and should be amenable to theoretical modeling. It

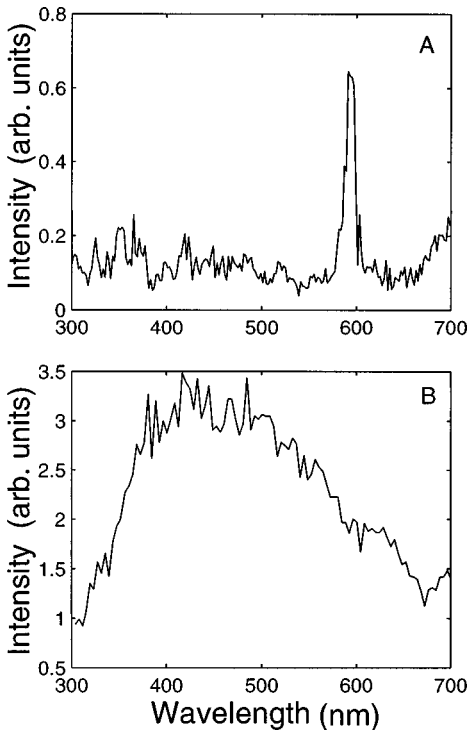

FIG. 4. Low resolution spectra of SL from air saturated 1*N* sodium chloride ethylene glycol solution at two different drive levels: (a) low drive level, $P_a \simeq 0.93$ bar; (b) high drive level. The spectra shown were obtained with a slit width of 3 mm, since the emission was not as bright as those for the conditions of spectra shown in Figs. 1 and 3(b).

is of significance to point out here that the estimated time of 100 ns for the persistence of ''high temperatures'' at the bubble center from a state-of-the-art model for SL by Kamath, Prosperetti, and Egolfopoulos (KPE) [11] compares well with the presently measured flash duration range of 60 to 150 ns in the few-bubble regime [typical result is shown in Fig. 2(b)]. Thus, the type of bubble motion analyzed by KPE seems to predict an important feature of presently observed SL in the form of resonance radiation; however, it cannot explain the timing characteristics of relatively short duration SL observed with air saturated ethylene glycol. The latter perhaps originates from a different type of bubble motion, involving harder collapse than that analyzed by KPE. Thus, on this basis we can propose two different types of bubble motion; one involving a ''soft'' collapse and another involving a ''hard'' collapse to explain SL with vastly different spectra and timing characteristics observed in the present work. Even with a ''soft'' collapse, temperatures of the order of 3000 K are predicted from the model of KPE and it is expected that much higher temperatures may be associated with a ''hard'' collapse.

In a multibubble environment from a resonant system, bubble motions with a soft collapse are likely to dominate at low acoustic drive level, whereas at a higher drive level both types of bubble motion may coexist. We, in fact, verified this by observing the PMT output from SL in air saturated 1*N* ethylene glycol sodium chloride solution. At low drive level mostly pulses with width in the nanosecond regime were observed, whereas at high drive level pulses with widths in nanosecond regime as well as those indicative of PMT response to SBSL were present. The primary difference noted above for SL characteristics at different drive levels is also reflected in the measured spectra as shown in Fig. 4. In Fig. 4(a) we see dominantly sodium resonance radiation (which we now associate with soft collapse) at low drive level and at high drive level the spectra is dominated by broadband emission [Fig. 4(b)]. However, as mentioned earlier on the basis of pulse width observations, even at high drive level resonance radiation is present and most likely originates from regions of the acoustic cell where pressure amplitudes are lower than the region seen by the monochromator. Therefore, we do see some variations in the SL characteristic at high drive level between argon and air saturated salt solution. The difference arises since with argon only one form of SL is seen, whereas with air saturated salt solution two forms of SL are possible. This clearly indicates that in addition to drive level, several other parameters are likely to influence the cavitation bubble dynamics [12] and the resulting form of SL. Thus, further work is required to formalize our above classification on ''soft'' and ''hard'' bubble collapse. Even though different types of bubble motion have been mentioned previously in connection with SL [13], we provide direct evidence for it. It is interesting to speculate on the basis of present observations whether the MBSL spectra measured by Matula *et al.* [9] does in fact originate from different types of bubble motion. It would be worthwhile to characterize the pressure amplitudes at which various transitions, for example from ''line'' emission to broadband emission takes place. In our present study with reference to Figs. 4(a) and 4(b), line emission was observed with $P_a \sim 0.9$ bar, whereas broadband emission only with an estimated $P_a > 3$ bar.

We thank M. S. Hegde for his valuable assistance in spectra measurement, K. S. Gandhi for useful discussions, and P. N. Shankar for his help in manuscript preparation. The oscilloscope was procured under a DSA grant from MHRD, Government of India.

[1] B. P. Barber and S. J. Putterman, Nature (London) **352**, 318 (1991); B. Gompf, R. Günther, G. Nick, R. Pecha, and W. Eisenmenger, Phys. Rev. Lett. **79**, 1405 (1997); R. A. Hiller, S. J. Putterman, and K. R. Weninger, *ibid.* **80**, 1090 (1998).

[2] T. J. Matula, R. A. Roy, and P. D. Mourad, J. Acoust. Soc. Am. **101**, 1994 (1997).

[3] K. R. Weninger, H. Cho, R. A. Hiller, S. J. Putterman, and G. A. Williams, Phys. Rev. E **56**, 6745 (1997).

[4] R. Hiller, S. J. Putterman, and B. P. Barber, Phys. Rev. Lett. **69**, 1182 (1992).

[5] Matula, Roy, and Mourad [2] have measured optical pulse widths from SL in glycerine-water mixture and the spectra of SL in this medium is found in D. F. Gaitan *et al.*, Phys. Rev. E **54**, 525 (1996).

[6] E. B. Flint and K. S. Suslick, J. Phys. Chem. **95**, 1484 (1991).

[7] From a study of the measured spectra we find that the sodium lines are red shifted by about 1 nm, the split is not resolved due to the unusual broadening estimated to be about 4 nm, and also there is pronounced asymmetry towards the red. These features are indicative of a high pressure environment at the instant of optical emission as pointed out, for example, in S. Chen and M. Takeo, Rev. Mod. Phys. **29**, 20 (1957).

[8] V. H. Arakeri, Curr. Sci. **66**, 213 (1994).

[9] T. J. Matula, R. A. Roy, P. P. Mourad, W. B. McNamara III, and K. S. Suslick, Phys. Rev. Lett. **75**, 2602 (1995).

[10] Results shown in Fig. 2 are representative; sample statistical study shows the estimated pulse widths to vary from 10 to 40 ns at high drive level and from 60 to 150 ns at low drive level when only few bubbles participated. Also the pulse heights varied from about -100 to as large as -1250 mV. These features are perhaps indicative of the sensitivity of the multibubble SL to parameters like initial nuclei size, acoustic pressure amplitude, and bubble interference effects.

[11] V. Kamath, A. Prosperetti, and F. N. Egolfopoulos, J. Acoust. Soc. Am. **94**, 248 (1993).

[12] See, for example, R. E. Apfel, Methods Exp. Phys. **19**, 355 (1981).

[13] A. J. Walton and G. T. Reynolds, Adv. Phys. **33**, 595 (1984).