Multibubble sonoluminescence pulse from Na atom in viscous liquid

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ABSTRACT

Multibubble sonoluminescence pulses of Na and continuum emissions were observed from NaCl solution in ethylene glycol saturated with xenon at ultrasonic frequency of 28 kHz using a system of a photomultiplier (0.78 ns rise time) and 4G sa/s oscilloscope. The continuum-emission pulse showed single peaks with the width being 1.4 ns that is nearly equal to an instrumental width. The Na-emission pulse showed multiple-peaks with each peak width being 1.4 ns and peaks interval being about 4 ns. This result disagrees with that by Arakeri et al. who reported a Gaussian shape pulse with the width ranging from 10 to 165 ns. High-speed photography of bubble dynamics observed in this solution suggests that the multiple-peaks are due to the superposition of single peaks which are resulted from many tiny bubbles fragmented from a very large bubble which phenomenon may be specific to a viscous liquid.

INTRODUCTION

Optical pulse width is one of the key parameters for understanding an origin of sonoluminescence. Pulse width of single-bubble sonoluminescence (SBSL) from water has been measured to be 100 - 300 ps. This result is in agreement with a bremssrahlung model of an ionized gas [1]. Recently, pulse width of Xe bubbles sonoluminescing brightly in sulfuric acid has been reported to be 2 - 10 ns [2-4]. Hopkins et al. [2] pointed out that this large value of flash duration is associated with a surface emitting blackbody. For multi-bubble sonoluminescence (MBSL), Matula et al. [5] measured the pulse width in an aqueous solution of glycerin at 28 kHz using a photomultiplier and a fast digitizer. They reported the value of less than 460 ps, which is comparable to that in SBSL.

Sonoluminescence from alkali-metal ion solutions has been investigated [6, 7] because the location of alkali-metal atom emission is of interest as well as how non-volatile ions are reduced and electronically excited. SL pulse width of Na emission was measured by Giri and Arakeri [8], and Arakeri and Giri [9]. They observed SL from NaCl solution in ethylene glycol at low acoustic amplitude near 1 atm, and reported the pulse width of 62 ns in argon saturated solution, which value depended on saturated rare gases. The purpose of the present report is to clarify the pulse width of Na emission from ethylene glycol solution in comparison with that from aqueous solution. We observed multiple-peaked pulses from ethylene glycol solution with each peak width being 1.4 ns and peak interval being about 4 ns, which has not been reported before. High-speed photograpy of bubbles and high-speed SL movies were also obtained for elucidating the multiple- peaked pulse.

EXPERIMENTAL

1 M NaCl solution in ethylene glycol was carefully degassed while stirring and saturated with Xe gas under 1 atm. The sample cell used was 300 mL cylindrical quartz flask. A 28 kHz sandwich transducer was bonded to the bottom of the flask. The experimental system used is shown in Fig.1. A signal from function generator was amplified and matched for impedance to the transducer. Applied acoustic power was 5.6 W corresponding to acoustic pressure of 2.3 atm. SL emission was filtered and detected using a photomultiplier (Hamamatsu H7422-01) having a rise-time of 750 ps and a
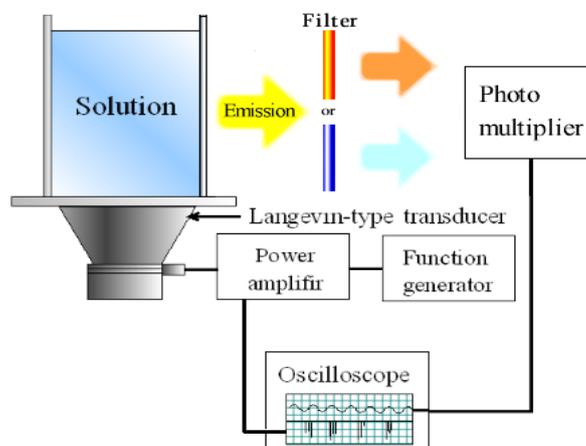
Figure 1. Experimental system for measuring SL pulse width from 1 M NaCl- ethylene glycol solution. SL emission was filtered with different filters to obtain Na emission pulse and a continuum emission pulse

digital oscilloscope (Agilent DSO5052A) with 4 G sampling/s. Single-shot waveforms of SL pulse were obtained with triggering by itself. Two optical filters were used: a sharp-cut filter transmitting wavelength over 580 nm for detecting Na emission, and a blue filter transmitting wavelength in the range of 300- 500 nm for detecting a continuum emission. A preliminary pulse-width measurement on SBSL from water showed a full width at half-maximum (FWHM) of 1.4 ns. This is nearly equal to an instrumental width of our system.

Shadowgraph movies of cavitating bubbles in ethylene glycol solution were captured using a high-speed video camera (Shimazu, HPV-2) with a maximum speed of 1,000,000 fps. Further, assembling the high-speed video camera with an image intensifier unit (Hamamatsu, C9546) enabled us to obtain a SL movie at the speed of 32,000 fps. A rectangular glass cell was used in this experiment.

RESULTS AND DISCUSSION

Pulse widths of Na and continuum emissions

Sonoluminescence from NaCl-ethylene glycol solution saturated with Xe is separated by two kinds of filters, which yields a continuum-emission pulse and Na-emission pulse as shown in Figs. 2 and 3, respectively. The pulse width (FWHM) of the continuum emission observed was 1.4 ns, which is equal to that for SBSL in water. Although the major contribution to the pulse width is an instrumental one, this result suggests that the pulse width of the continuum emission for MBSL is similar to that for SBSL.
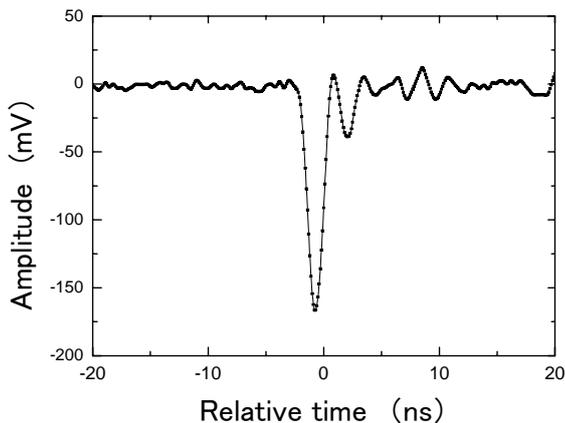
Figure 2. Single shot waveform of continuum-emission pulse obtained with a blue filter transmitting wavelength in the range of 300-500 nm.
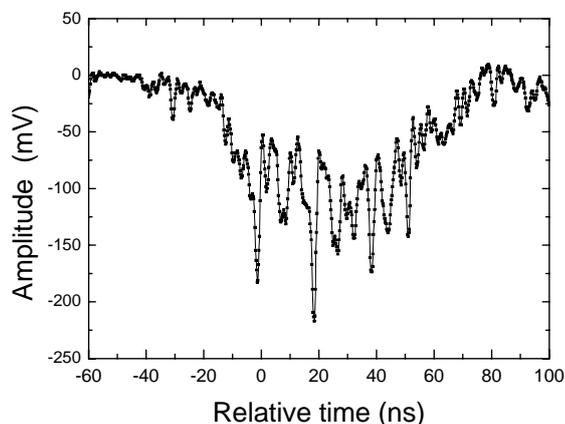
Figure 3. Single shot waveform of multiple-peaked pulse of Na-emission obtained with a sharp cut filter (>580 nm).

Unlike the single pulse of the continuum emission in Fig. 2, the Na-emission pulse in Fig. 3 shows a multiple-peaked shape. The width of individual peaks in Fig. 3 is nearly the same as that of the continuum emission and the peaks are separated by about 4 ns. The multiple-peaked pulse was observed at the frequency rate of 50% in the total events of 500. The other 50% are single pulses with the width of 1.4 ns. It is plausible that the multiple-peaked pulse is caused by the superposition of sonoluminescence from different bubbles generated at different locations. Figure 4 represents Na-emission pulses observed in the time span of 500 ns. One multiple-peaked pulse and three single pulses are detected. These pulses are separated by more than 100 ns. In most of the measurements, Na-emission pulses were separated widely and this separation time of 100 ns seems a minimum. We used no lens and only used a 5 mm-diameter aperture for limiting the observation region. It is, therefore, unlikely that the each peak in multiple-peaked pulse is emitted from un-correlated bubble, whereas the multiple-peaked pulse and the single pulses in Fig. 4 are probably resulted from different bubbles generated independently. This prediction is supported by the fact that no multiple-peaked pulse was observed in Na emission from NaCl aqueous solution. Only single pulses with the width of 1.4 ns were obtained. The multiple-peaked pulse is specific to ethylene glycol solution.
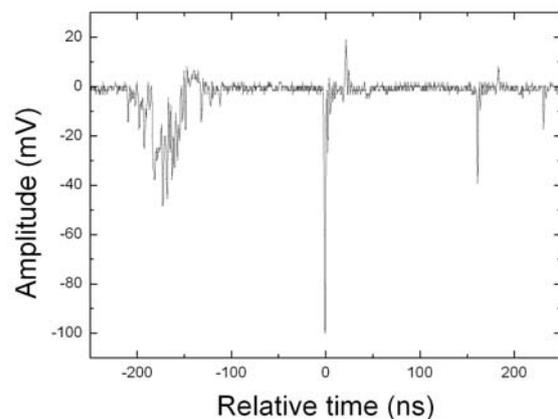
Figure 4. Na- emission pulses in the time span of 500 ns. One multiple-peaked pulse and three single pulses are obtained. These four pulses are emitted by different bubbles generated independently.

Observation of bubble dynamics

Shown in Fig. 5 is a photograph of sonoluminescence from NaCl solution in ethylene glycol at 28 kHz taken with a Nikon D90 camera. The exposure time was 10 s and the sensitivity was ASA 6400. The size of a rectangular cell in Fig. 5 is 70 mm in width. Orange colored region indicates Na emission and is located around antinode of standing waves established in the cell. Giri and Arakeri [8] reported that Na emission was observed at relatively low acoustic pressure, suggesting the mechanism of soft bubble collapse. We experienced a similar observation when air was partially contained in the solution. However, Na emission could be observed in all cases with the acoustic pressure of up to 2.3 atm under the condition that Xe gas is well dissolved.
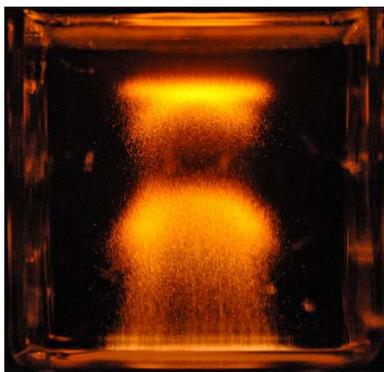
Figure 5. Photograph of SL from 1 M NaCl-ethylene glycol solution at 28 kHz. The size of a rectanglar cell is 70 mm in width. The exposure time was 10 s and the ASA number was 6400.

We obtained shadowgraph movies of bubble dynamics in 1 M NaCl-ethylene glycol solution. The salient feature of the bubble dynamics in the present solution is bubble fragmentation. Figure 6 is one shot of cavitating bubbles photographed at the speed of 2000 frames per second. A large bubble (about 250 μm in diameter at a maximum size) indicated as "A" in the figure is translating in the direction denoted as an arrow. Immedeately before this event, the bubble "A" coalesced with a bubble and changed its direction. After the coalescence the bubble "A" fragmented into many tiny daughter bubbles denoted as "B". A term "leakage" may be appropriate instead of "fragmentation" to describe this event. There are two characteristics which have not been observed in water. One is that large size of bubbles over 200 μm are frequenctly seen. This is associated with that noble gas can dissolve in ethyene glycol much more than in water. Rectified diffusion of gas into bubble is activated and this increases the bubble size. The large bubble stays a long time in the solution since viscosity is large in ethylene glycol. The other is the "leakage" of tiny bubbles. The maximum size of these bubbles is less than 10 μm. The smallness may be caused by the value of surface tension which is 66 % of that of water. We propose that these tiny bubbles characteristic to the present solution are responsible for the multiple-peaked pulse observed.

If a SL image corresponding to the bubble shadowgraph is obtained, it will help to prove our proposition. Though SL
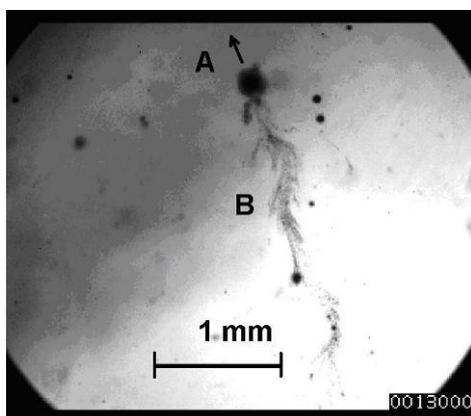
Figure 6. Shadowgraph of characteristic bubbles translating in the direction denoted by an arrow. A large bubble "A" leaks many tiny bubbles "B", which occurs immediately after coalescence with a bubble. The frame speed is 2000 fps.
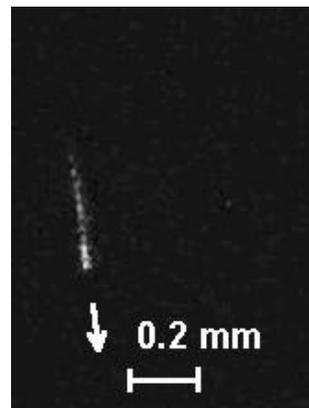
Figure 7. SL image of Na emission photographed at the speed of 32,000 fps. Each luminous point corresponds to a SL event from a bubble occurring in an ultrasonic period. This image suggests that the multiple-peaked pulse is resulted from many tiny bubbles shown in Fig. 6.

from the present solution is intense, as can be easily seen in dim room, it was difficult to take moving picture. In order to amplify the SL signal, we used an image intensifier unit using two-stage micro-channel plates in front of the high-speed video camera. With this system we succeeded in taking SL movie at the speed of 32,000 fps. Figure 7 shows a shot of SL of Na emission. It is noted that orange colour is converted to white. Eleven luminous points are propagating in the direction denoted by an arrow, while the number of luminous point increases from one to eleven in a time of 0.48 ms. The ultrasonic frequency of 28 kHz is nearly equal to the frame speed of 32,000 1/s, which means that an exposure time of one shot is nearly equal to an ultrasonic period. Since one event of SL occurrs every ultrasonic period, the luminous point in the shot correspods to one event of SL by one bubble considering the size and separation of bubbles shown in Fig. 6. Thus the shot of Fig. 7 represents the SL from tiny bubbles indicated by "B" in Fig. 6. Also the number of luminous points, eleven, roughly corresponds to the number of peaks of SL pulse in Fig. 2.

In conclusion, multiple-peaked SL pulse observed in NaCl – etylene glycol solution is caused by collapse of tiny bubbles which fragmented from a very large bubble after the instance of bubble coalescence. The intrinsic pulse width of Na emission may be similar to that of continuum emission, which disagreed with a previous study reporting that the pulse width is of the order of 10 ns. The present result strongly suggests that Na ion enter bubble via liquid droplet at the instant of bubble fragmentation.

REFERENCES

1 S.Hilgenfeld, S. Grossmann and D. Lohse, "A simple explanation of light emission in sonoluminescence" *Nature* 398, 402-405(1999).

2 S. D. Hopkins S. Putterman, B. A. Kappus. K. S. Suslick and C. G. Camara, "Dynamics of a sonoluminescing bubble in sulfuric acid" *Phys. Rev. Lett.* 95, 254301(2005)

3 J. Xu et al., "Composition and its evolution inside a sonoluminescing bubble by line spectra" *Phys. Rev. Lett.* 76, 026308(2007)

4 J-S. Jeon, C. Lim and H. Y. Kwak, "Measurement of pulse width of sonoluminescing gas bubble in sulfuric acid solution" J. Phys. Soc. Jpn. 77, 033703(2008)

5   T. J. Matula, R. A. Roy and P. D. Mourad, "Optical pulse width measurements of sonoluminescence in cavitation-bubble fields" *J. Acoust. Soc. Am.* 101, 1994-2002,(1997)

6   P.-K. Choi, S. Abe and Y. Hayashi, "Sonoluminescence of Na atom from NaCl solutions doped with ethanol" *J. Phys. Chem. B* **112**, 918-922 (2008)

7   Shogo Abe and Pak-Kon Choi, "Spatio-Temporal Separation of Na-atom Emission and Continuum in Sonoluminescence" *Jpn. J. Appl. Phys.* **48**(2009)07GH02

8   A. Giri and V. H. Arakeri, "Measured pulse width of sonoluminescence flashes in the form of resonance radiation" *Phys. Rev. E* 58, R2713-R2716, (1998)

9   V. H. Arakeri and A. Giri, "Optical pulse characteristics of sonoluminescence at low acoustic drive levels" *Phys. Rev. E* 63, 066303(2001)