Hydrodynamical perturbation effects in multibubble sonoluminescence

David W. Kuhns, Anatol M. Brodsky, and Lloyd W. Burgess
Department of Chemistry, University of Washington, Seattle, Washington 98115
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We describe the observation of new dynamical effects in multibubble sonoluminescence (MBSL). The experiments show a specific time-dependent nonmonotonic response of MBSL in aqueous solutions to hydrodynamical perturbations caused by sudden pressure changes. Such response as well as observations of the asymmetrical and nonmonotonic changes in the intensity and width of the sodium emission with hydrodynamic perturbations have been interpreted as the manifestations of the cooperative dynamical phase transition effect. [S1063-651X(98)07802-7]

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One of the most active fields in contemporary liquid state physics is the experimental and theoretical study of UV and visible light emission during sonoluminescence (SL) [1–7]. One of the most interesting results established in recent years is the very short time of the emission pulse during single bubble sonoluminescence (SBSL), equal to, according to different estimations, 10–300 ps [8]. It has been reported that the pulse duration of multibubble SL (MBSL) fits the same time interval [9], which indicates at least a degree of analogy in the mechanism of SBSL and MBSL.

The design of the experiments described in this article have been based on the presumption that analysis of the reaction of MBSL to hydrodynamical perturbations can substantially elucidate the mechanism of MBSL. Such analysis can in particular allow one to begin to test the hypothesis that acoustic and/or optical collective effects may substantially contribute to the SL phenomenon. The basis for such a hypothesis is the wide ranging analogy of SL emission to collective emission in amplifying gain media, of the type known in condensed state physics such as Dicke super-radiance [10]. The results of the described study of MBSL can lead to a more thorough understanding of SL in general, including both SBSL and MBSL.

We have measured the changes in sodium MBSL emission in aqueous sodium salt solutions after rapid change in external pressure. The idea to use alkali-metal emission characteristics for SL diagnostics was proposed in the pioneering work by Lewschin and Rschevkin [11] as early as 1937. The description of recent measurements of alkali-metal emission in SL made under static conditions can be found in the article by Flint and Suslick [12], in which a reasonable doubt was placed upon the usefulness of alkali-metal MBSL emission as an SL temperature probe. The experimental setup (Fig. 1) consists of an ultrasonic source (Sonics and Materials 400 W 20 kHz generator) connected to stainless steel mechanically focusing half inch diameter horn tuned to 20 kHz. The horn is secured and sealed into a stainless steel jacketed flow cell with a sample volume of approximately 80 ml. The cell is charged via an in-line pump, which operates at a constant rate of ~4 liters per min. The cell and sample are temperature controlled using a water bath that is circulated through the jacket of the cell and the sample is pumped through an immersed coil in the bath. Argon gas pressure has two entrances to the flow cell, one at the bottom to bubble gas through the sample and one at the top to immediately affect sample headspace pressure without a major disruption to the sample flow profile. The sample is sparged with argon gas for ~30 min prior to sonication. Pressure is measured via an Omega membrane transducer (part PX-203). For spectral evaluation the MBSL light is collected through the quartz window into a fiber optic bundle butted up against the window. The bundle consists of ~20 200 μm glass on quartz fibers distributed to fill a circular area. The other end of the fiber reorganizes the fibers into a slit configuration, which is attached to a MacPherson Model 218 0.3 m monochromator equipped with a 600 groove grating blazed at 300 nm and slit set at 200 μm. The incident light falls upon a liquid-N_2 cooled Princeton Instruments charge-coupled device (CCD) array. The resolution is just under one-tenth of a nanometer, with a hot pixel background of less than $1e^-$ per pixel per hour. A drawback to the CCD detector is a ~500 ms pixel transfer rate to download data. Therefore, for fast temporal data the MBSL light is collected through a 600 μm glass on glass fiber into an American Holographic monochromator set

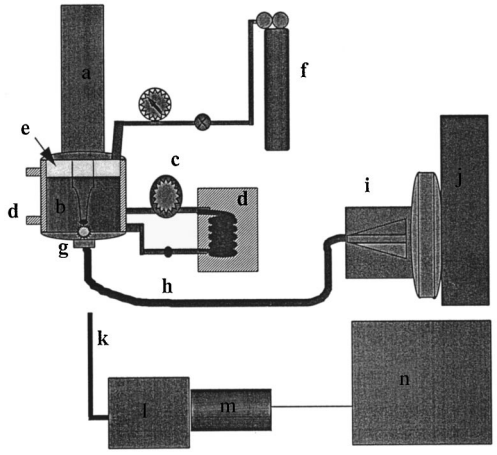

FIG. 1. MBSL instrument layout. (a) Ultrasonic horn, (b) jacketed sonocell, (c) sample recirculating-loading pump, (d) coolant waterbath and inlet or outlet to sonocell, (e) headspace above sample, (f) argon gas, (g) quartz window, (h) quartz fiber optic bundle, (i) MacPherson spectrometer, (j) Princeton CCD detector, (k) alternative 600 μm collection fiber, (l) American Holographic monochromator, (m) PMT, (n) computer with data acquisition hardware and software.

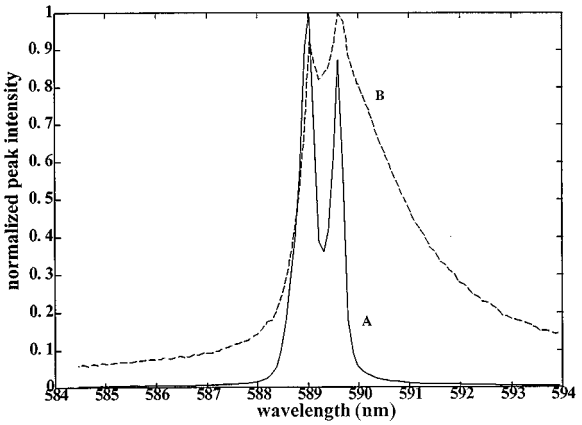

FIG. 2. The flame emission of a saturated sodium flame (A), (10 ms integration), and the MBSL of 1.0*M* $NaCl_{aq}$ emission (B) at 2.8 bars (15 s integration) were collected using the same optical system (200 μm slit). Note the asymmetrical broadening to the red in (B), not due to pressure broadening, and the alignment of the doublet transitions.

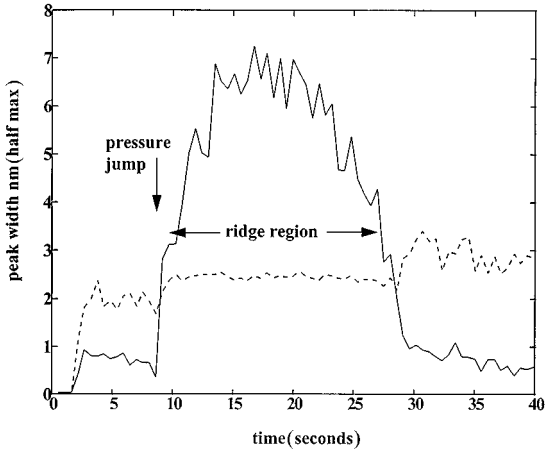

FIG. 3. Time profile of MBSL sodium peak max of 0.8*M* $NaCl_{aq}$ collected on a CCD system, during the fast pressure change from 1 bar to 3.3 bars. Relative peak intensity (solid line) was scaled to fit plot, calculated peak width at half maximum (dashed line) is overlaid to show difference before, during, and after ridge region.

at 589 nm with about a 1 nm bandwidth incident on a Hamamatsu Photomultiplier tube (PMT). PMT voltage, temperature, pressure, and hydrophonic (SEA model SPRH-0500 hydrophone) data are coincidentally collected on a Pentium powered PC running Labview data acquisition software. Pressure and temperature are digitized through a DASH16jr A/D and hydrophonic data are collected on a GaGe Compuscope running at 200 kHz. The temperature was held constant at 3.0 ± 0.5 °C. Acoustic ultrasonic input power remained constant at about 110 W into the horn. The concentration of the sodium sample remains at 1.0*M* sodium chloride in milli-*Q* water (18 MΩ/cm).

Two types of pressure experiments were performed, one in which the gas headspace pressure is incremented or decremented and held at that pressure for 60–100 s. This was done to reduce hysteresis effects, a possible result in stepping the pressure in just one direction, and to minimize total solvated gas uptake. The CCD detector was then used to integrate the spectral signal in these experiments. In a second type of experiment the gas pressure was changed suddenly from atmospheric pressure (1 bar) to some pressure p. Measurements in light output are made during the dynamic perturbation and subsequent relaxation of the bubble system using the PMT setup, previously described. Only the intensity at 589 ± 0.5 nm wavelength was measured. The typical shape of the measured sodium MBSL line is compared to the sodium doublet attained via flame emission in Fig. 2. It is seen in Fig. 2 that within the resolution of the doublet transitions that there is substantial peak width broadening on the red side. Such behavior cannot be attributed to gas phase pressure broadening, which would lead to the more symmetrical Lorentz type intensity distribution [13]. The far red tail of the sodium peak has the characteristic features of the Rayleigh wing effect [14] in nonequilibrium light emission, resulting from secondary nonelastic scattering of the emitted light in the surrounding water matrix. Correspondingly, this tail cannot provide information about the temperature of the source.

Figure 3 shows the changes of sodium MBSL peak intensity with time after a sudden pressure change. The solid line represents a cross section of data and gives a good view of what we term the stable ridge region between 9 and 30 s after the sudden pressure change. The peak width at half maximum for each peak (dashed line) is overlaid on the peak intensity data. Peak width data are taken from the stable ridge region. The dependence of the peak heights and the peak widths on the full range of external pressures used are shown on Figs. 4 and 5. The above experimental data can be mutually explained in the framework of the cooperative model of sonoluminescence with a dynamical phase transition (DPT) in the gain media, induced by acoustic wave pumping of the type described in [10,15]. Especially characteristic for DPT are the S-shape dependencies presented in Figs. 4 and 5. Typical for DPT is also the linear relationship in the critical region between the square root of the relative emission peak intensity and peak width, presented in Fig. 6. The observed sharp time and external parameter dependencies of MBSL cannot be attributed only to the smooth changes of the bubble resonance frequency and the bubble

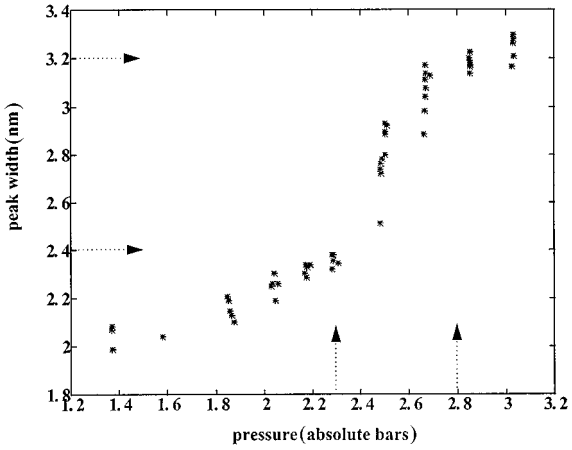

FIG. 4. Plot of incremental pressure changes vs the sodium MBSL peak width of 1.0*M* $NaCl_{aq}$. Arrows mark the bistability transition region in the pressure interval of 2.3 and 2.8 bars and corresponding peak width interval of 2.4 and 3.2 nm.

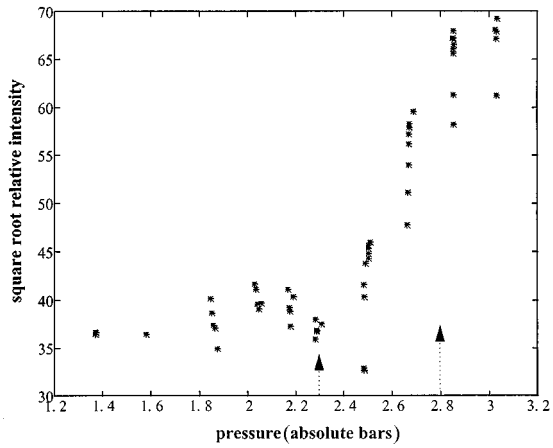

FIG. 5. Plot of incremental pressure changes vs the square root of sodium MBSL peak intensity of 1.0*M* $NaCl_{aq}$. Arrows mark bistability transition region consistent with Fig. 4.

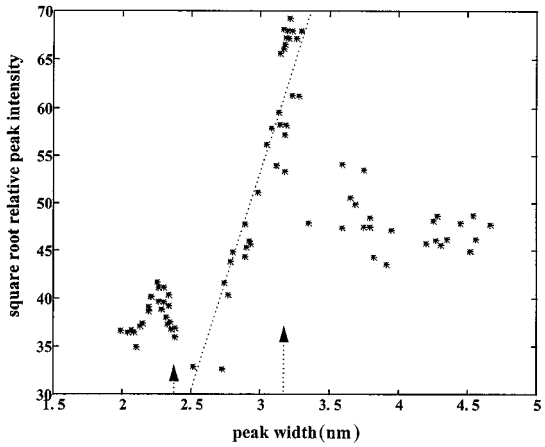

FIG. 6. Plot of peak width of sodium MBSL of 1.0*M* $NaCl_{aq}$ vs the square root of relative peak intensity. The dashed line represents the expected linear relation between ΔW and p in the phase transition interval. Arrows mark transition region consistent with Figs. 4 and 5.

radii and density. Analogous behavior has been observed in optical experiments in gain random media and have been considered as sufficient proof of the existence of DPT [10]. The transient oscillations in the MBSL output closely resemble a phenomenon that is known in regular laser theory as laser spiking [16]. The intensity of MBSL sharply increases after the pressure changes, plateaus, and then sharply decreases to a stationary level, lower or at the same intensity before the pressure change. Such behavior cannot be explained as a result of the simple gas transport effects to the bubble interior after a nearly instantaneous pressure change, since such effects could lead only to a monotonic change in intensity. The most detailed previous measurements of the influence of equilibrated static pressure on MBSL intensity can be found in the article by Chendke and Fogler [17] who observed that an increase in static pressure smoothly reduces the resonant bubble size and increases the overall bubble density in sharp contrast to our observations of the singular dependence of sodium emission on the dynamics of the pressure changes. At the same time such singular behavior is characteristic for DPT during Dicke superluminescence [10].

In conclusion, it can be said that the experiments described in this article show the substantial singular influence of hydrodynamics on MBSL and support the concept of the realization of DPT in MBSL. However, the final confirmation of the realization of DPT and especially the establishment of it's microscopic mechanism will require further experimental and theoretical efforts. We think that in an elucidation of such a mechanism, it is necessary to take into account specific mesoscopic effects in the bubble dynamics [18], including the dynamics of the adsorption desorption on the liquid-gas interface of the bubble [19].

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