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Comparisons of sonoluminescence from single-bubbles and cavitation fields: bridging the gap

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Abstract

Sonoluminescence (SL) refers to the generation of light through the energetic pulsations of acoustic cavitation bubbles in a liquid. For years, SL was observed primarily in cavitation fields. These bubbles are believed by many to undergo near-adiabatic compression, resulting in the heating of the bubble contents and the subsequent emission of light. Recently, researchers have discovered a 'new' form of sonoluminescence in which light is observed to emanate from a single bubble undergoing very large volume excursions. The mechanism for light production is unknown, but many believe it is due to a rapid heating of the central core by an imploding shock wave. Based in part on the emission time scales, there is a common belief that the two forms of SL are quite distinct. We address this issue by comparing the two phenomena with regards to their light-flash durations and emission spectra – leading to some surprising differences *and* similarities. © 1997 Elsevier Science B.V.

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1. Introduction

Sonoluminescence (SL) is an important tool for sonochemistry research and can be used for studying the extreme conditions attained within cavitation bubbles. Spectroscopic analysis of the light emission from SL can be used to infer maximum pressures and temperatures reached in cavitation bubbles [1]. Similarly, changes in the light intensity might be used to infer changes in chemical reaction rates. Unfortunately, the complexity associated with cavitation fields makes it difficult to perform strict scientific studies. A change in one parameter might effect changes in other parameters that cannot be precisely controlled. For example, varying the pressure amplitude might affect a change in the light intensity. Is this variation in light intensity due to a variation in the collapse dynamics of a bubble, or is the variation simply due to a change in the number density of light-emitting bubbles, or both? Similarly, are the effects from a change in drive frequency due to a change in the response of individual bubbles, or from the gross shift in the bubble size and population distribution, or more likely, a combination?

Questions like this are difficult to answer, since with cavitation fields, only macroscopic inferences can be made. Fortunately, the discovery of single-bubble sonoluminescence [2] has now made it possible to study the dynamics of cavitation bubbles in a highly controlled and repeatable fashion. For this case a bubble is acoustically levitated in a standing wave field by means of the primary Bjerknes force [3,4]. At driving pressures near 1 bar the bubble oscillates in a highly non-linear fashion, emitting a short flash of light at or near the minimum bubble radius. The bubble does not appear to disintegrate and coalesce during each acoustic cycle, but rather appears to remain intact throughout the cycle, repeating its behavior during subsequent cycles in a synchronous fashion [2,5]. Measurements of the dynamical response of single bubbles can be made using light scattering techniques [6,7], and changes in the dynamical behavior can similarly be measured, thus allowing unprecedented opportunities to study the effects of various parameters on cavitation.

Recent studies of single-bubble sonoluminescence

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(SBSL) have led to discrepancies with well-known properties of cavitation-field sonoluminescence (CFSL). For instance, The spectra from cavitation-field sonoluminescence typically contains molecular or atomic emission bands associated with the bulk fluid. In fact, specific C_2 emission bands from argon saturated silicon oil were used to infer the temperature of sonoluminescence from cavitation fields [1]. As yet, no emission bands have been observed from single-bubble sonoluminescence.

Another discrepancy between CFSL and SBSL is in the duration of the light flashes. Though no thorough investigation had been made on the pulse duration from CFSL, the few measurements that were made seem to indicate that CFSL pulses last on the order of nanoseconds [8–10]. This has led theorists to model the collapse from cavitation bubbles as near-adiabatic [11]. SBSL on the other hand, has an upper limit on the pulse duration of approximately 15 ps, [12] thus precluding the adiabatic collapse hypothesis.

On the surface it appears that CFSL and SBSL are completely different processes. However, it is also apparent that the commonality of cause (acoustic cavitation) and effect (light emission) for both types of SL suggests some association of the underlying physics. In the next two sections we take a closer look by making specific comparisons of the properties of CFSL and SBSL. In the first section, a comparison of the spectra from a sodium chloride solution is made. Then we investigate in a more thorough manner the pulse duration from CFSL and compare this with SBSL pulse durations. Though these comparisons are limited in scope, they indicate that there are both differences *and* similarities between CFSL and SBSL, and through more research these comparisons will lead to a more thorough understanding of sonoluminescence in general, and may thus lead to new insights into the physics of sonochemistry.

2. Comparisons of spectral properties

Unfortunately, while CFSL can occur in both aqueous and non-aqueous (and readily in gassy) fluids, SBSL appears to be stable and visible only in aqueous based (and highly degassed) systems. This makes it difficult to make specific comparisons of SL emission spectra from a host of different systems. The only direct comparison published to date involves sodium-chloride in water [13]. Fig. 1 illustrates the differences in the two spectra. The lack of a sodium line in the SBSL spectrum may indicate that the highly symmetrical pulsations from SBSL preclude the presence of sodium inside the bubble (because of sodium s low volatility). The presence of a sodium line in CFSL may be due to jetting [11] or other mechanisms whereby sodium can be entrained within the bubble [14]. Though it may be that SBSL has different emission mechanisms than CFSL, the spectral

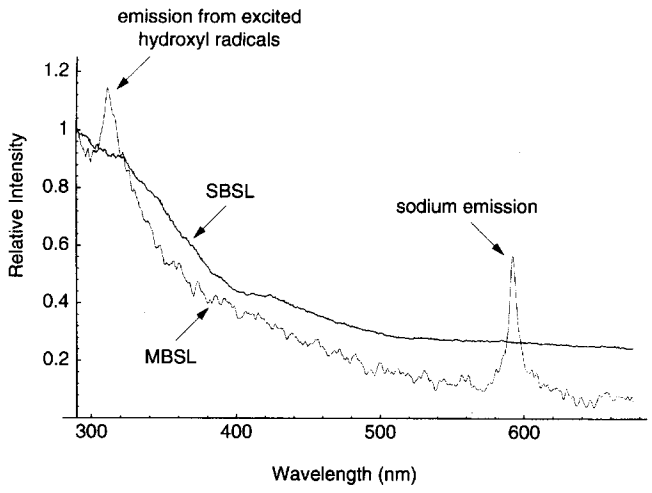

Fig. 1. Comparisons of the spectra of CFSL and SBSL from a 0.1 molar sodium chloride solution shows both the similarities (the underlying continuum) and differences (band and atomic emission) between these two types of SL. The lack of sodium emission in SBSL is attributed to the spherical symmetry of the bubble pulsations. Since sodium is non-volatile, there is no mechanism that drives sodium atoms into the bubble. With CFSL on the other hand, the extreme violence of the cavitation field is sufficient to generate highly asymmetric pulsations, with possible liquid jetting (containing sodium atoms) into the bubble interior, thus giving a mechanism for driving sodium into the hot bubble interior. Note that a liquid shell surrounding the bubbles does not heat sufficiently to generate sodium emission [11], since if this were the case, then one would see sodium emission from SBSL as well. (Reprinted by permission of the authors.)

differences may simply be due a difference in the symmetry of the bubble oscillations.

Another possibility is that sodium does enter the bubble, but the temperatures in SBSL become too hot for line emissions (only free–free and free–bound transitions would be permitted). However, though the core of the bubble would be hot indeed, this represents only a small fraction of the bubble volume. The surrounding cooler regions would presumably emit and this emission should be detectable *if* sodium penetrated the bubble wall and *if* the intensity of any subsequent line emission is not overwhelmed by the continuum emission from the very hot core of the bubble.

A more confusing picture arises when trying to describe the differences in the spectra near 310 nm. In CFSL, this emission is most probably due to excited hydroxyl radical recombination [15]. With SBSL, the continuum may simply overwhelm any OH band emission. We do not attempt at this point to explain the difference in this region.

Though there are marked differences in the emission band spectra, there is a commonality in the underlying continuum structure. Note that both systems show a strong continuum monotonically increasing towards higher energies. If SBSL is a much 'hotter' phenomenon than CFSL, as has been theorized, then the continuum associated with CFSL might simply be due to a few

bubbles within the cavitation field that are undergoing 'SBSL-like' behavior. That is, consider that the cavitation field is made up of vapor and gas bubbles of various sizes, undergoing volume mode pulsations to various degrees, depending of the local pressure field and their proximity to other bubbles and boundaries. It is probable that some subset of this cavitation field will undergo the volume growth and collapse dynamics that is associated with SBSL, if only for a single acoustic cycle. Thus, this subset of bubbles would contribute to the continuum while the rest of the light-emitting bubble population would contribute to band emission [16].

3. Comparisons of the pulse durations

Previous studies on the pulse duration from CFSL are limited, but seem to indicate that light pulses last less than 10 ns [8–10] though no thorough study has been done. From these early experiments, we believed that by using more contemporary instrumentation, a more well-defined pulse width, or range of pulse widths could be made. Thus, we attempted to measure the pulse duration of CFSL in order to make more quantitative comparisons with SBSL pulse durations [17].

Details of the experiment are given elsewhere [17]. We utilized a photomultiplier tube (PMT) detector with a 650 ps rise-time, and a 10 GS/s digitizer to collect the data. Fig. 2 illustrates a typical CFSL pulse received with this system. The response of our optical detector was estimated by utilizing SBSL as an impulse for our system; measuring the pulse duration from SBSL represents the minimum pulse width observable from our system. We then compared this impulse response measurement with that obtained with CFSL. Fig. 3a shows a histogram of pulse widths from SBSL. The peak near 1.1 ns indicates the limit of our detection system (since SBSL is already known to be less than 15 ps in duration). Fig. 3b shows a histogram of pulse width measurements from CFSL. Note that the pulse width for CFSL also peaks near 1.1 ns. Since this represents the minimum observable pulse duration, we conclude that the actual pulse width from CFSL is much less than 1.1 ns. In fact, by assuming that the pulses are Gaussian in shape, and that the detector system (PMT and digitizer) have independent Gaussian responses, we can place an upper bound on the pulse width of CFSL at 500 ps.

Fig. 4 shows the corresponding rise-times from SBSL and CFSL. Note that both systems show peaks near 650 ps, and corresponds to the impulse response of our system. Thus, not only are CFSL pulse durations shorter than 500 ps, the rise-times are much shorter than would be predicted based on a near-adiabatic collapse hypothesis. Indeed, the mechanisms governing CFSL and SBSL emission may have more in common than originally thought.

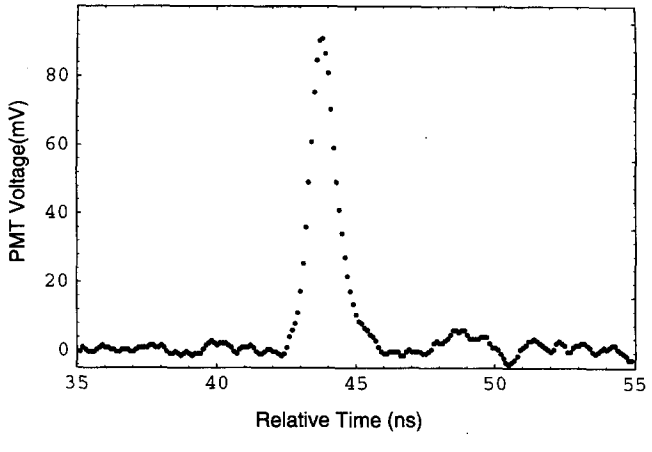

Fig. 2. Typical CFSL pulse acquired with our system. Measurements of the pulse height, rise-time, and pulse width can be made. The pulse height of approximately 90 mV is within the linear operating region of the PMT (the single photoelectron pulse height is near 20 mV). The slight ringing at the end of the pulse is typical of fast PMT's and is due to the external circuitry. The pulse width of this particular signal at FWHM is about 1.1 ns. (Reprinted by permission of the authors.)

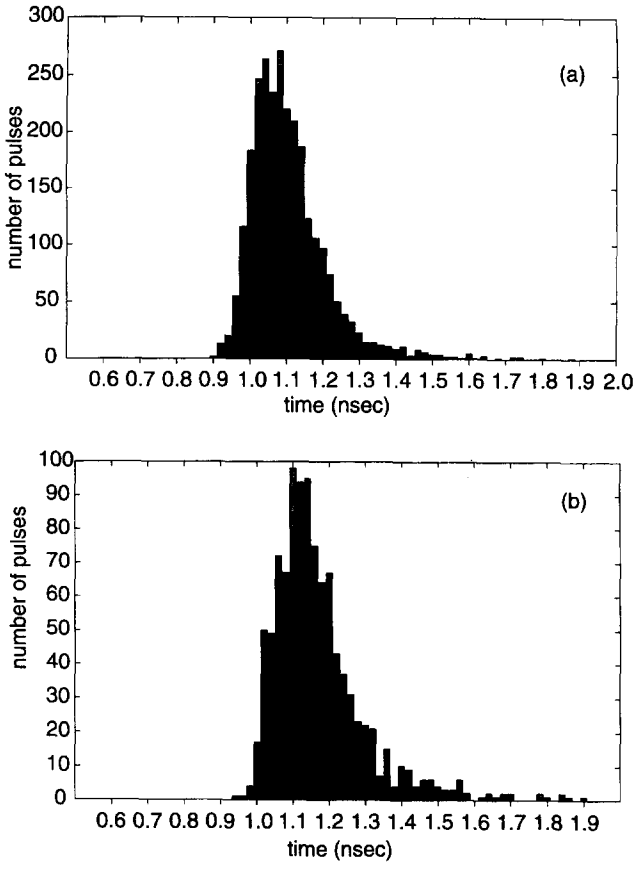

Fig. 3. Pulse width distribution for (a) SBSL and (b) CFSL. The pulse width is measured as the full width at half-max of the SL pulse. The spread in the distributions is due to the transit time spread of electrons in the PMT. The similarity of the two distributions implies that CFSL also generates an impulse with respect to our detection system. (Reprinted by permission of the authors.)

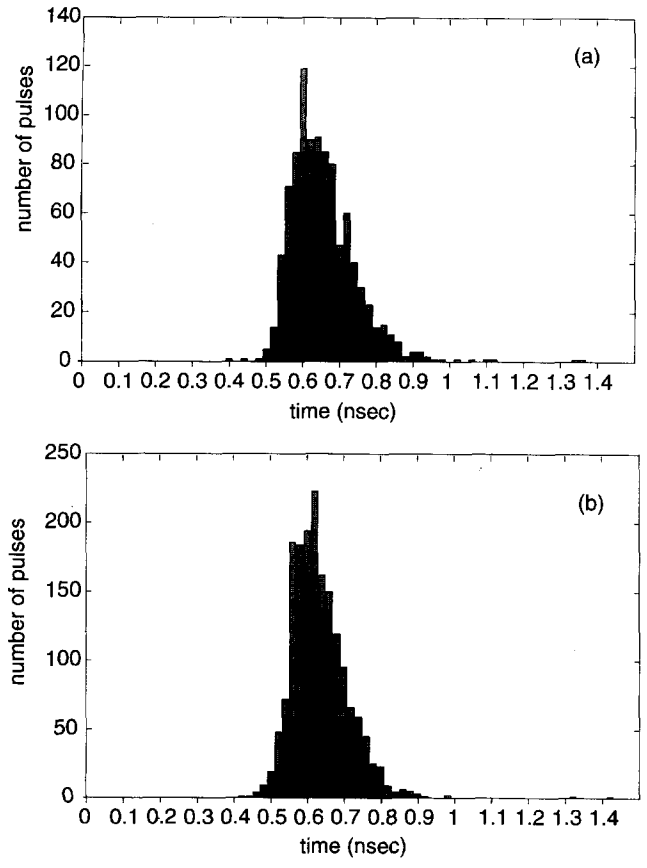

Fig. 4. Rise-time distribution for (a) SBSL and (b) CFSL. The rise-time is a measure of the time it takes for a signal to rise from 10% to 90% of its maximum height. The rated rise-time of the PMT is 650 ps. (Reprinted by permission of the authors.)

4. Conclusion

The similarities in the continuum structure from the corresponding emission spectra, together with the differences in band emission seem to indicate that CFSL is comprised of both 'hotter' and 'cooler' bubbles; the cooler bubbles generating bound–bound transitions, while the hotter bubbles generate both free–free and free–bound transitions. The implications for sonochemistry include maximizing sonochemical activity by optimizing the sound field and bubble size and population distribution.

From our pulse duration experiments, the results seem to suggest that sonoluminescence from cavitation fields is similar to that from single bubbles, in the sense that both types of SL have extremely short pulse durations. Since the pulse durations are too short to be caused by conventional adiabatic collapse, some other mechanism must be responsible for the light emission. Shock waves [18] and plasma discharge [19] are two of many possible interpretations.

Acknowledgement

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