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Synchronous Picosecond Sonoluminescence

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13. ABSTRACT *(Maximum 200 words)*

If acoustic cavitation is produced in a liquid, the implosion of cavities can heat the internal contents of the bubble to incandescent temperatures. The electromagnetic emissions associated with this energy concentration can often be seen with the naked eye. This phenomenon, in which light is generated by sound is called sonoluminescence. A particular form of this phenomenon is called single-bubble sonoluminescence. This form is much easier to study because the fundamental bubble dynamics that leads to bubble collapse and the associated electromagnetic emissions can be ascertained. Also, in single bubble sonoluminescence it is likely that the bubble collapse is spherically symmetric, resulting in an amplification of this already violent phenomenon. The intent of this project is to investigate the phenomenon of single bubble sonoluminescence, and from knowledge gained here, to be able to provide useful information concerning the more general phenomenon of multiple-bubble sonoluminescence.

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Project Title:

Synchronous Picosecond Sonoluminescence

Project Description:

If acoustic cavitation is produced in a liquid, the implosion of the cavities can heat the internal contents of the bubble to incandescent temperatures. The electromagnetic emissions associated with this energy concentration can often be seen with the naked eye. This phenomenon, in which light is generated by sound is called sonoluminescence. There are two kinds of sonoluminescence: one type is associated with the production of many cavitation bubbles by a sound field--this form is called multiple-bubble sonoluminescence; a second type is associated with the production of light from a single, stable, violently oscillating gas bubble--this form is called single bubble sonoluminescence. This latter form is much easier to study because the fundamental bubble dynamics that leads to bubble collapse and the associated electromagnetic emissions can be ascertained. Also, in single bubble sonoluminescence, it is likely that the bubble collapse is spherically symmetric, resulting in an amplification of this already violent phenomenon. The intent of this project is to investigate the phenomenon of single bubble sonoluminescence, and from knowledge gained here, to be able to provide useful information concerning the more general phenomenon of multiple-bubble sonoluminescence.

Research Approach:

If a gas bubble is positioned within an acoustic stationary wave, and driven at a frequency below it natural resonance frequency, it will experience radiation pressure forces, called Bjerknes forces [Crum, 1975], which will tend to force the bubble toward an acoustic antinode. Simultaneously, the bubble will also experience the buoyancy forces of gravitation which will normally be directed vertically upwards. Thus, under conditions that are not too difficult to obtain, it is possible to "acoustically levitate" a single bubble in the bulk of a liquid [Crum,1980; 1983]. Under conditions that ARE reasonably difficult to attain, it is possible to see SL from this single bubble. We are investigating single bubble sonoluminescence by this levitation technique.

The fact that this system is very "robust" and the luminescing bubble tends to remain in a stable configuration was initially very difficult to understand. A gas bubble is a very nonlinear mechanical oscillator, and one would not expect it to behave in such a stable pattern. At an acoustic pressure amplitude of 0.15 MPa, a typical value for a sonoluminescing bubble, there should be a considerable nonlinear response. Furthermore, one should expect considerable rectified diffusion for such a bubble. Obviously, the conditions that lead to single bubble sonoluminescence are both of interest and an important area of our research.

When one combines the effect of the nonlinear oscillations of a bubble and the concept of rectified diffusion, one can gain some insight into the probable origins of SBSL. Using the nonlinear rectified diffusion code developed by Church, [1988], we are investigating this phenomenon for bubbles under the set of parameters similar to those experiencing sonoluminescence. Consider Fig. 1, which shows the threshold for rectified diffusion under the conditions similar to those that would give rise to SL.

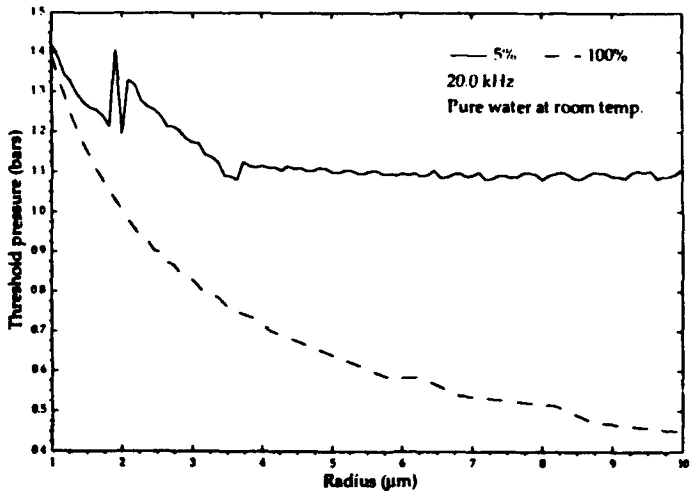

Fig. 1. Calculations of the threshold for rectified diffusion of gas bubbles in water for a driving frequency of 20.0 kHz and dissolved gas concentrations of 5% (solid line) and 100% (dashed line). Note that for this case, the threshold is so large that nonlinear behavior is observed.

Note that when one reduces the dissolved gas concentration to the level desirable for SBSL, a couple of "notches" appear in the threshold curve that could be very meaningful (these notches or depressions or valleys are due to the nonlinear response of the bubble and represent harmonic resonances). Consider the notch near 3.5 μm; this value of the radius is near that of the value measured by Barber and Putterman [1993] in their light scattering experiments. Suppose that a bubble were "positioned" within this notch (above the threshold) by selecting a bubble of about 3.5 μm in radius and driving it at a pressure amplitude of 0.115 MPa (1.15 bars) and at a frequency of 20 kHz. This particular bubble would then grow until it engages the threshold curve at about 3.7 μm. At this point, if it grew further, it would pass into a region for which the threshold is higher than 0.115 MPa, and it would start to dissolve. As it got smaller, it would cross the threshold line once again, and get larger, etc. Thus, a positive slope on the rectified diffusion threshold curve is a point of stable equilibrium for a bubble driven at a fixed acoustic pressure amplitude.

For this bubble to produce SL flashes each cycle, it would seem necessary that shape oscillations not occur, because that should lead to asymmetrical bubble collapse, which would in turn, tend to prevent SL. It is difficult to make measurements in this region, of course, but the extrapolations of our earlier measurements and calculations [Horsburg, et al., 1989] suggest that the threshold for shape oscillations is larger than 0.115 MPa in this radius range (2-5 μm). Thus, it is plausible that this general region of parameter space is the location for single-bubble sonoluminescence.

Currently, we are working to define the volume of parameter space in which this single-bubble sonoluminescence can occur. By measuring the maximum size of the bubble, the acoustic pressure amplitude, and the phase of the electromagnetic emissions, we can determine the equilibrium radius and thus the anticipated dynamic behavior of bubble. Furthermore, we are acquiring sonoluminescence intensity measurements for the transition to and out of stable behavior. Some very interesting transients are appearing that are currently anomalous, but may lead to some insight into the stability issue.

Project Accomplishments

Some recent accomplishments of the project are as follows:

- We have measured the light emissions from the bubble during the long-term transient stage--over several seconds (many thousand cycles); this information will probably be very useful in determining the origins of the remarkable stability of the bubble during single-bubble sonoluminescence. These results were presented at the 126th meeting of the Acoustical Society of America in Denver. A copy of the abstract for this presentation is appended.

- We have developed a method for determining the maximum radius of the bubble with a microscope and video camera. These results were presented at the 127th meeting of the acoustical Society of America in Cambridge, MA. A copy of the abstract for this presentation is appended.

- We have calculated the threshold for rectified diffusion for bubbles under single bubble sonoluminescence conditions. Our calculations suggest that a "kink" in the threshold curve may be a necessary condition for single bubble sonoluminescence to occur. These results were presented at a special IUTAM symposium on Bubbles and Cavitation held in Birmingham, England in September, 1993. A copy of the paper for this conference is appended.

- It has always been difficult to understand why the behavior of single bubble sonoluminescence is so different from multiple-bubble sonoluminescence. In a paper written for the Acoustical Society of America, I have suggested that they are two completely different phenomena, the former resulting from an imploding shock wave in the gas, the latter from adiabatic compression of an asymmetric bubble. This paper is also included in the appendix.

- The Principal Investigator was invited to present a lecture on sonoluminescence to the Western Pacific Regional Acoustics Conference. This meeting will occur in late August of 1994; a copy of the paper that will be delivered is also included in the appendix.

- The Principal Investigator was also invited to write a chapter on sonoluminescence for the Book entitled "Luminescence in Solids and Liquids", edited by Dr. D. R. Vij of Kurukshetra University, India, and to be published by John Wiley and Sons. Mr. Sean Cordry, who is supported by this grant, is the principal author on this work, and it is nearing completion.

- The Principal Investigator was invited to draft an article on Sonoluminescence for Physics Today. An initial draft has been submitted and is under review by the AIP staff.

References Cited:

Barber, B. P. and Putterman, S. J., "Light scattering measurements of the repetitive supersonic implosion of a sonoluminescing bubble", Phys. Rev. Lett. **69**, 3939 (1992).

Church, C. C., "Predictions of rectified diffusion during nonlinear bubble pulsations at biomedical frequencies", J. Acoust. Soc. Am. **83**, 2210 (1988).

Crum, L. A., "Bjerknes forces in a stationary sound field", J. Acoust. Soc. Am., **57**, 1363 (1975).

Crum, L. A., "Measurements of the growth of air bubbles by rectified diffusion", J. Acoust. Soc. Am. **68**, 203 (1980).

Crum, L. A., "The polytropic exponent of gas contained within air bubbles pulsating in a liquid", J. Acoust. Soc. Am. **73**, 116 (1983).

Horsburg, S., Holt, R. G. and Crum, L.A., "Thresholds for surface wave generation on acoustically levitated gas bubbles", J. Acoust. Soc. Am. **86**, S42 (1989).

List of Publications, Patents, Presentations, and Honors

This report is appended immediately following.

OFFICE OF NAVAL RESEARCH
PUBLICATION/PATENTS/PRESENTATION/HONORS REPORT
for
1 Oct 93 through 30 Sept 94

R&T Number: 4126966---01

Contract/Grant Number: N00014-93-1-0322

Contract/Grant Title: Bubble-Generated Picosecond Sonoluminescence

Principal Investigator: Dr. Lawrence A. Crum

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a. Number of Papers Submitted to Referred Journal but not yet published: 1

b. Number of Papers Published in Referred Journals: 1
(list attached)

c. Number of Books or Chapters Submitted but not yet Published: 0

d. Number of Books or Chapters Published: 0
(list attached)

e. Number of Printed Technical Report & Non-Referred Papers: 0
(list attached)

f. Number of Patents Filed: 0

g. Number of Patents Granted: 0
(list attached)

h. Number of Invited Presentations at Workshops or Prof. Society Meetings: 1

i. Number of Presentation at Workshop or Prof. Society Meetings: 2

j. Honors/Awards/Prizes for Contract/Grant Employees:
(list attached, this might include Scientific Soc. Awards/Offices, Promotions, Faculty Award/Offices etc.)

k. Total number of Graduate Students and Post-Docs Supported at least 25%, this year on this contract,grant:
Grad Students 1 and Post Docs 0

How many of each are females or minorities? (These 6 numbers are for ONR's EEO/Minority Reports; minorities include Blacks, Aleuts Amindians, etc and those of Hispanic or Asian extraction/nationality. This Asians are singled out to facilitate meeting the varying report semantics re "under-represented")

Grad Student Female	0
Grad Student Minority	0
Grad Student Asian e/n	0
Post-Doc Female	0
Post-Doc Minority	0
Post-Doc Asian e/n	0

Attachments to the P^3H Report.

b. Title of Paper published in referred journals:

L. A. Crum, "Sonoluminescence, sonochemistry, and sonophysics", J. Acoust. Soc. Am., **95**, 559-562 (1994).

j. Honors/Awards/Prizes

- L. A. Crum was elected Vice President of the Acoustical Society of America

- L. A. Crum was promoted to Chairman, Department of Acoustics and Electromagnetics, Applied Physics Laboratory

11:45

3aPA7. Some light emission features of single bubble sonoluminescence. Sean M. Cordry (Dept. of Phys., Univ. of Mississippi, University, MS 38677), Lawrence A. Crum, and Ronald A. Roy (University of Washington, Seattle, WA 98105)

Bubbles created via electrolysis were allowed to rise though water in a quiet acoustic levitation vessel. The sound field was then activated, forcing several bubbles to converge and coalesce near a pressure antinode. Light emission measurements were then taken with a Hamamatsu photomultiplier tube and single photon counter as a state of single bubble sonoluminescence (SBSL) evolved. The measurements reveal brief periods fluctuations in light emission intensity followed by long periods of stable (i.e., nonfluctuating) emission. The time scales for the fluctuations are on the order of half a second. Previous measurements of light emission have indicated that SBSL exhibits remarkably long-lived, stable behavior [Gaitan *et al.*, J. Acoust. Soc. Am. **91**, 3166 (1992); B. P. Barber and S. J. Putterman, Nature **352**, 318 (1991)]. These measurements, however, imply a transient regime that the bubble must pass through while seeking a final position of stability. The highest light emission intensities are seen in the transient regime. These transient light emission should provide important information concerning the mechanisms through which SBSL develops its remarkable stability. Further, acoustic emissions from SBSL were observed to evolve from

8:45

4aPA1. A novel technique for measuring the maximum radius of a sonoluminescing bubble. Sean M. Cordry (Dept. of Phys. and Astron., Univ. of Mississippi, University, MS 38677), Lawrence A. Crum, and Ronald A. Roy (Univ. of Washington, Seattle, WA 98105)

Single bubbles are acoustically levitated in a rectangular standing wave resonator filled with clean liquid that has been "degassed." The acoustic pressure of the sound field induces radial oscillations in the bubble of sufficient magnitude to cause them to sonoluminesce. Measurements of the maximum diameter are then made using a video imaging system composed of a microscope, CCD, and digitizing computer. These diameter measurements are compared to the light emission intensity as measured by a photomultiplier tube (PMT) and single photon counter (SPC). Both measurements of light intensity and diameter are made for pure water and several aqueous solutions. The results of this experiment as well as the accuracy and precision will be discussed. [Work supported by ONR.]

2938 J. Acoust. Soc. Am., Vol. 95, No. 5, Pt. 2, May 1994

SINGLE BUBBLE SONOLUMINESCENCE

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ABSTRACT If acoustic cavitation is produced in a liquid, the implosion of the cavities can heat the internal contents of the bubble to incandescent temperatures. The electromagnetic emissions associated with this energy concentration can often be seen with the naked eye. This phenomenon, in which light is generated by sound is called sonoluminescence. There are two kinds of sonoluminescence: one type is associated with the production of many cavitation bubbles by a sound field--this form is called multiple-bubble sonoluminescence; a second type is associated with the production of light from a single, stable, violently oscillating gas bubble--this form is called single bubble sonoluminescence. This latter form is much easier to study because the fundamental bubble dynamics that leads to bubble collapse and the associated electromagnetic emissions can be ascertained. Also, in single bubble sonoluminescence, it is likely that the bubble collapse is spherically symmetric, resulting in an amplification of this already violent phenomenon. This article presents a brief survey of single bubble sonoluminescence and describes some of more remarkable aspects of its behavior.

INTRODUCTION

When an acoustic wave propagates through a liquid, certain conditions can be attained in which the mechanical energy associated with the acoustic field is converted into electromagnetic energy. This process is called *sonoluminescence*, and is the principal focus of this article.

Sonoluminescence is the indirect consequence of a process called acoustic cavitation, in which the acoustic stress applied to the liquid causes the liquid to fail during the negative half cycle, producing vapor- and gas-filled voids within the liquid. The subsequent collapse of these voids during the positive half cycle can be sufficiently violent to produce sonoluminescence. When cavitation is generated in the bulk of a liquid, multiple cavitation "sites" usually appear with the result that the process is not localized at a particular point but spatially and temporally distributed over a relatively large parameter space. This cavitation takes on various forms, but at the moderate amplitudes of interest (0.05 - 0.50 MPa), one can observe many small bubbles; on an instantaneous basis, one sees random flashes of light from the SL zone that gradually build into a geometrical configuration representative of the acoustic field produced by the transducer and the constraining volume [Crum, et al., 1986; 1987].

SL was discovered nearly 60 years [Marinesco and Trillat, 1933; Frenzel and Schultes, 1935], and since then there have been a variety of explanations given for the origin of the electromagnetic emissions. Electrical discharge theories of various types were at first quite popular. As early as 1940, Frenkel [1940] suggested that electrical charges known to exist on the surfaces of bubbles (see for example, [Watmough, et al., 1992]) were somehow made to discharge. This model, though seriously challenged by the experiments of Suslick [Suslick, 1989;1990], even has it modern advocates [Margulis, 1992].

However, most modern researchers support the hot-spot model of Noltingk and Neppiras [1950] which posits SL to be the result of incandescence of the bubble's contents. Nonetheless, there are still many unanswered questions concerning the origin of this phenomenon.

One reason SL has intrigued investigators is its enormous capability of energy amplification. For example, an acoustic pressure amplitude of 0.1 MPa (1 bar) can generate sonoluminescence in water. This pressure corresponds to an acoustic energy density of about 2.2 j/cm^3 or, in a rather unconventional unit, to an energy density of about 4×10^{-10} ev/molecule. In contrast, there is recent evidence that the photons associated with SL can have energies in excess of 6 ev; thus, the generation of SL from an acoustic wave results in an energy amplification of approximately 1.5×10^{10}! To see how this compares with other phenomena, consider a thermal neutron that is absorbed by the fissionable isotope of Uranium. The neutron has about 0.025 ev of energy while the resulting fission releases about 200 Mev--an energy amplification of *only* 0.8×10^{10}.

It has been difficult to determine the basic physical processes that give rise to SL, partly because it is practically impossible to spatially and temporally control the production of cavitation, the origin of SL [Crum and Reynolds, 1985]. It occurs randomly and over a relatively large spatial area. However, the fortuitous discovery of SL from a single stable cavitation bubble by Gaitan, [1990] has now made it possible to study the phenomenon in much more detail than was previously possible. With this system the dynamics of a single cavitation bubble can be studied simultaneously with the physical processes that lead to SL, thus isolating the critical temporal and spatial parameters that give rise to SL [Gaitan and Crum, 1990; Gaitan, et al., 1992]. Furthermore, some recent discoveries associated with Single Bubble Sonoluminescence (SBSL) have been quite dramatic and totally unexpected; according, research into this phenomenon has potential for interesting new physics as well as important technology applications.

RESULTS

Single Bubble Sonoluminescence. In order to understand this phenomenon, it is first necessary to explain how one can generate the conditions under which it can be expressed.

If a gas bubble is positioned within an acoustic stationary wave, and driven at a frequency below it natural resonance frequency, it will experience radiation pressure forces, called Bjerknes forces [Crum, 1975], which will tend to force the bubble toward an acoustic antinode. Simultaneously, the bubble will also experience the buoyancy forces of gravitation which will normally be directed vertically upwards. Thus, under conditions that are not too difficult to obtain, it is possible to "acoustically levitate" a single bubble in the bulk of a liquid [Crum,1980; 1983]. Under conditions that ARE reasonably difficult to attain, it is possible to see SL from this single bubble. Shown in Fig. 1 is a diagram of an experimental arrangement that permits SBSL to be observed.

It is a rather dramatic sight to see a very small bubble that is a constant source of light emissions. When seen with the naked eye in a darkened background, it appears as a bright star, glowing brightly. When background lights are illuminated in the room, it is still possible to see the light emissions from the bubble. Even more remarkably, the bubble tends to remain firmly fixed in space, without any perceivable movement within the liquid. In our initial studies of this phenomenon, we wished to determine if the bubble remained a single entity during its entire oscillation cycle and thus conceived a light scattering technique for visualizing the bubble.

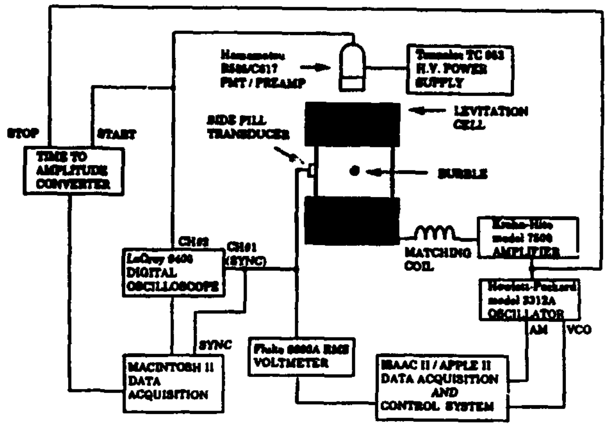

Fig. 1. A schematic diagram of the experimental apparatus used to acoustically levitate a gas bubble and to generate single bubble sonoluminescence. This figure also shows the apparatus used to record the electromagnetic emissions.

Shown in Fig. 2 is a diagram of the experimental system used to determine the radius-time curve of the single bubble. By using the apparatus shown in Fig. 2 and the techniques developed by Holt and Crum [1992], the data shown in Fig. 3. were obtained.

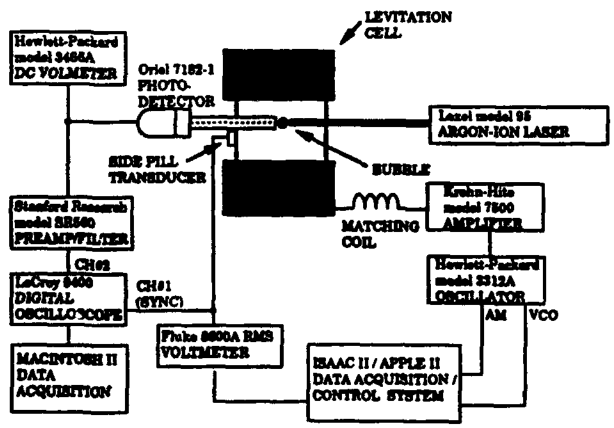

Fig. 2. A schematic diagram of the experimental apparatus used to scatter laser light from the sonoluminescing bubble and to obtain the radius-time curve.

In Fig. 3 the acoustic field is shown in the top trace, the experimentally-determined radius-time curve in the middle trace, and the SL emissions in the bottom trace. Note that the emissions are synchronous with the collapse of the bubble, and that they occur each and every acoustic cycle.

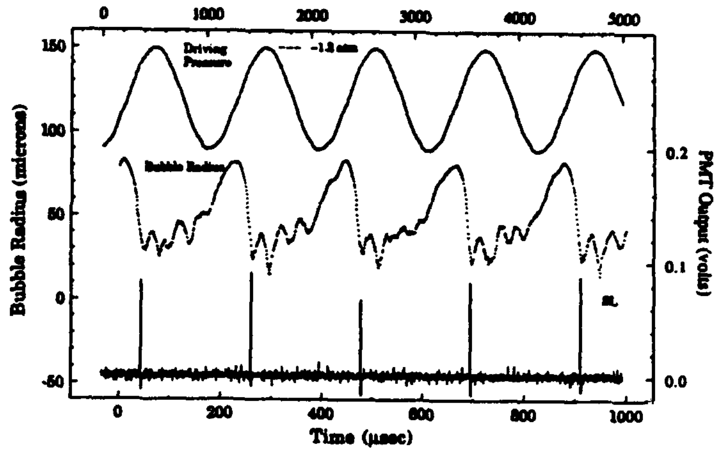

Fig. 3. Simultaneous plots of the sound field (top), the measured bubble radius (middle) and the sonoluminescence emissions (bottom). For this case the acoustic pressure amplitude was about 0.12 MPa, the driving frequency was 22.3 kHz, and the host liquid was a glycerin/water mixture.

Although only a few acoustic cycles are shown in this trace, we have observed the behavior shown here to be repeated for hours.

<u>Origins of Single Bubble Sonoluminescence.</u> The fact that this system is very "robust" and the luminescing bubble tends to remain in a stable configuration was initially very difficult to understand. A gas bubble is a very nonlinear mechanical oscillator, and one would not expect it to behave in such a stable pattern. For example, consider Fig. 4, which shows the calculated response curves for the bubble as a function of its size and its driving amplitude. Note that for the bottom trace, even for a driving acoustic pressure amplitude of 0.06 MPa (0.6 bar), there is a very nonlinear response. Since the threshold for SBSL is approximately 0.1 MPa, and extends to abut 0.15 MPa, the two top curves on this figure illustrate the expected bubble response. Obviously, the bubble must behave in a very nonlinear fashion.

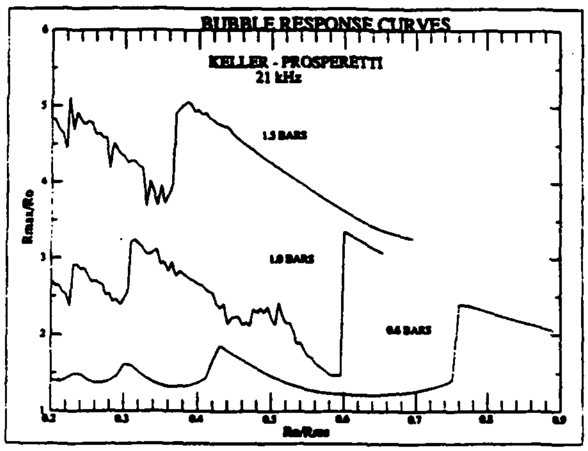

Fig. 4. Bubble response curves for various acoustic pressure amplitudes using the Keller-Prosperetti formulation of bubble dynamics [Prosperetti, et al., 1988]. For these curves, the liquid is assumed to be water, saturated with gas, and driven at a frequency of 21 kHz. R_o refers to the equilibrium radius; R_{max} to the maximum radius; R_{res} to the linear resonance radius of the bubble.

Another phenomenon associated with an oscillating bubble within a liquid that should affect the stability of this system is "rectified diffusion" [Hsieh and Plesset, 1961; Strasberg, 1961; Crum, 1980]. This effect also results from the nonlinear behavior of the bubble. Specifically, when the bubble is large, gas diffuses into the bubble; when it is small, gas diffuses out. Because the diffusion is proportional to the area, over a complete cycle, more gas diffuses in than out; thus, there is a "rectification" of mass into the bubble. The "area effect" is enhanced by the fact that a small shell of liquid surrounding the bubble is compressed during expansion, thus concentrating the dissolved gas near the bubble wall and enhancing the diffusion rate; just the opposite happens during compression. The combination of the "area effect" and the "shell effect" is to pump significant amounts of gas into the bubble each acoustic cycle.

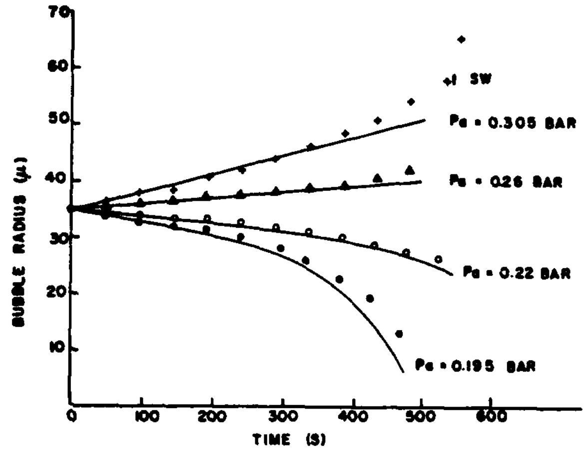

Fig. 5. Shown here are experimental measurements that demonstrate the phenomenon of rectified diffusion. One sees that for the range of conditions observed in this experiment, it is possible to use an analytical theory to accurately predict the threshold and the magnitude of the bubble growth rate.

When one combines the effect of the nonlinear oscillations of a bubble and the concept of rectified diffusion, one can gain some insight into the probable origins of SBSL. Using the nonlinear rectified diffusion code developed by Church, [1988], we have investigated this phenomenon for bubbles under the set of parameters similar to those experiencing sonoluminescence. Consider Fig. 6, which shows the threshold for rectified diffusion under the conditions similar to those that would give rise to SL.

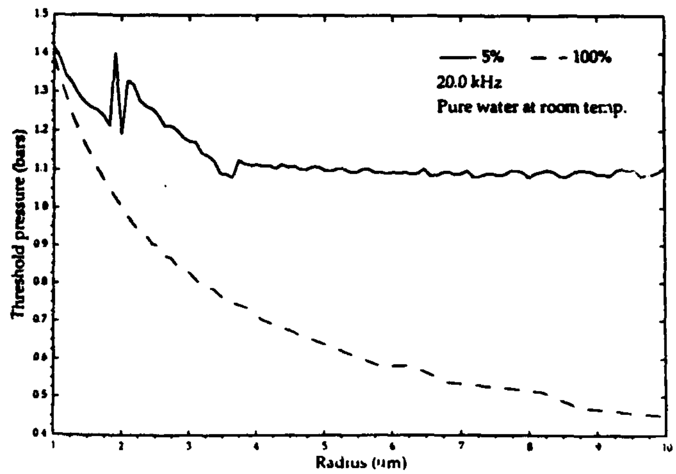

Fig. 6. Calculations of the threshold for rectified diffusion of gas bubbles in water for a driving frequency of 20.0 kHz and dissolved gas concentrations of 5% (solid line) and 100% (dashed line). Note that for this case, the threshold is so large that nonlinear behavior is observed.

Note that when one reduces the dissolved gas concentration to the level desirable for SBSL, a couple of "notches" appear in the threshold curve that could be very meaningful (these notches or depressions or valleys are due to the nonlinear response of the bubble and represent harmonic resonances). Consider the notch near 3.5 µm; this value of the radius is near that of the value measured by Barber and Putterman [1993] in their light scattering experiments. Suppose that a bubble were "positioned" within this notch (above the threshold) by selecting a bubble of about 3.5 µm in radius and driving it at a pressure amplitude of 0.115 MPa (1.15 bars) and at a frequency of 20 kHz. This particular bubble would then grow until it engages the threshold curve at about 3.7 µm. At this point, if it grew further, it would pass into a region for which the threshold is higher than 0.115 MPa, and it would start to dissolve. As it got smaller, it would cross the threshold line once again, and get larger, etc. Thus, a positive slope on the rectified diffusion thresh curve is a point of stable equilibrium for a bubble driven at a fixed acoustic pressure plitude.

For this bubble to produce SL flashes each cycle, it would seem necessary that shape oscillations not occur, because that should lead to asymmetrical bubble collapse, which would in turn, tend to prevent SL. It is difficult to make measurements in this region, of course, but the extrapolations of our earlier measurements and calculations [Horsburg, et al., 1989] suggest that the threshold for shape oscillations is larger than 0.115 MPa in this radius range (2-5 µm). Thus, it is plausible that this general region of parameter space is the location for single-bubble sonoluminescence.

Unique Aspects of SBSL. Putterman and his colleagues [Barber and Putterman, 1991; Barber, et al., 1992; 1994; Barber and Putterman, 1992; Hiller, et al., 1992] have examined various aspects of SBSL and determined that the phenomenon itself has some amazing and unique features. For example, the lifetime of the flash appears to be no longer than 50 ps. Since one would expect that this lifetime would be associated with the time for which the gas in the interior was heated to incandescent temperatures, it would seem possible to calculate the emission time. Although we have not performed these calculations specifically for the case of SBSL, we can learn from the computations of Kamath and Prosperetti [Kamath, et al., 1993] that elevated temperatures are expected to occur for times on the order of nanoseconds. The only explanation (at this time) for the short lifetime of SBSL is that an imploding shock wave is launched within the gas during the final stages of bubble collapse and this shock wave gives rise to these emissions [Barber, et al., 1994].

A second phenomenon discovered initially by Gaitan [Gaitan, et al., 1988] but refined in considered detail by Barber and Putterman [1991] is the remarkable stability of the luminescing bubble. If one measures the jitter in the SL emissions from cycle to cycle, under normal conditions, this jitter is itself on the order of 50 ps. This level of stability is much in excess of the electrical apparatus that drives the system. There is no current explanation for this behavior.

Finally, since the optical emissions from SBSL are sufficiently intense so that one can obtain a spectrum, it is of great interest to use this spectrum to obtain a measure of the effective temperature of the gas within the collapsed (imploded) bubble.

Shown in Fig. 7 is a spectrum obtained by Carlson, et al., [1993]. Because the spectrum, as shown in this figure, seems to be best-fit with a black body curve, one could estimate the effective temperature of the SL emissions by the best-fit-black-body temperature. As shown on this figure, this value can be as high as 16,000K--a rather remarkable value, considering the surface of the sun is only about 7,000K!

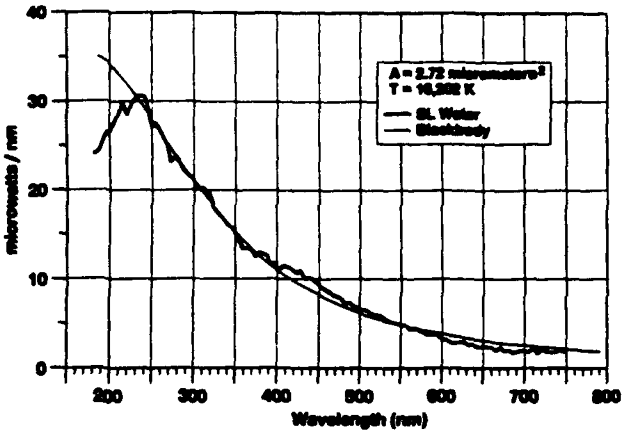

Fig. 7. Spectrum of light emitted by single bubble sonoluminescence. The heavy line is the measured spectrum; the light line is the best fit to the spectrum using a black body distribution function. [After Carlson, et al., 1993].

SUMMARY

Single bubble sonoluminescence is a remarkable phenomenon that is largely not understood. The bubble demonstrates a stability that is totally unexpected; the duration of the sonoluminescence emissions are so short as to be unmeasurable with currently available apparatus, and the emission spectrum shows indications of extraordinarily high temperatures. This phenomenon bears further study, both from an experimental and theoretical aspect.

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The author acknowledges many helpful discussions with Ron Roy, Sean Cordry, Tom Matula, and Pierre Mourad. This work was supported by the Office of Naval Research, Physics Programs.

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Sonoluminescence, sonochemistry, and sonophysics

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Recent measurements of sonoluminescence produced by a single, stable, pulsating gas bubble indicate that the spectrum can be modeled by that for a blackbody at a temperature of nearly 40 000 K [R. Hiller, S. J. Putterman, and B. P. Barber, "Spectrum of synchronous picosecond sonoluminescence," Phys. Rev. Lett. **69**, 1182 (1992)]. These results are in contrast with earlier measurements of the spectrum which is modeled by electronic transitions of rotational and vibrational bands within specific elements of the host liquid [E. B. Flint and K. S. Suslick, "The temperature of cavitation," Science **253**, 1397 (1991)]. It is suggested that the single-bubble SL observed by Hiller *et al.* is intrinsically different from the multiple-bubble SL observed by Flint and Suslick. In the former case, symmetric bubble collapse leads to shock wave formation in the gas; in the latter case, asymmetric bubble collapse leads to liquid jets that penetrate the hot bubble interior and result principally in incandescence of the host liquid.

PACS numbers: 43.35.Ei

INTRODUCTION

The phenomenon of sonoluminescence (SL) has been of considerable recent interest due to some exciting discoveries that have lately come to light! Although this phenomenon has been known for over 60 years (Marinesco and Trillat, 1933; Frenzel and Schultes, 1935), it is only recently that it is beginning to be understood. The standard paradigm for its existence is that the electromagnetic emissions are associated with the collapse of cavitation bubbles produced within the host liquid by an acoustic field: gas contained within microscopic nucleation sites is made to grow explosively during the negative portion of the acoustic cycle and then these gas and vapor-filled cavities are driven to an implosive collapse during the positive portion. Consequently, the cavity's contents are heated adiabatically to incandescence temperatures. However, as this phenomenon is examined in continuing depth, there appear to be intrinsic inconsistencies in the results of various investigators.

For example, Suslick and others have revitalized the discipline of sonoluminescence chemistry (sonochemistry) in which SL plays a major role (for example, see Suslick, 1989, 1990; Suslick *et al.*, 1990; Berlan and Mason, 1992; Luche, 1992). In some recent experiments, Flint and Suslick (1991) have measured the "temperature of cavitation" and discovered that the SL emissions correspond to a value of approximately 5000 K (surface of sun ~7000 K); furthermore, they have demonstrated that the spectrum can be characterized by spectral peaks that correspond principally to electronic transitions in the host liquid, rather than the gas contained within the bubble. Because the liquid, in addition to the gas, can be elevated to high temperatures, chemical effects of considerable magnitude can be attained: thus, the discipline of sonochemistry.

In contrast, recent measurements of the physical aspects of cavitation by Gaitan *et al.* (1992) and in greater depth and detail by Putterman and his colleagues (Barber and Putterman, 1991; Barber and Putterman, 1993; Barber *et al.*, 1992; Hiller *et al.*, 1992; Lofstedt *et al.*, 1993) have demonstrated that SL can occur in a single, stable, pulsating gas bubble and that the SL spectrum is devoid of major peaks and can be fitted only by a blackbody spectrum with temperatures as high as 40 000 K. These high temperatures, rapid SL pulse rise times, and remarkable synchrony imply the operation of some exciting new physical acoustics—sonophysics. Both the sonochemistry and the sonophysics groups have performed careful and repeatable experiments, with no inconsistencies within their own measurements. This brief communication purports to resolve these inconsistencies by proposing that there are two different forms of SL, in which the physical mechanisms that give rise to SL emissions are shown to be fundamentally different.

I. SONOCHEMISTRY

In the sonochemistry experiments of Suslick and his colleagues, cavitation is typically generated by an acoustic horn that is immersed within the host liquid and driven at relatively large amplitudes. As a consequence there is much cavitation at the tip of the horn, and throughout the bulk of the fluid (see, for example, Crum and Reynolds, 1985). The cavitation is seen to be "transient" in the normal use of the word, appearing and disappearing, often as rapidly as an acoustic cycle. In many cases the SL can be seen by the naked eye to be a faint glow that is distributed throughout the bulk liquid. Although measurements of the temporal duration of the SL flashes are difficult to obtain because of the transient nature of the SL, reported lifetime of several nanoseconds are typical (Jarman, 1960; Taylor and Jarman, 1970; Margulis, 1992). Individual flashes appear to be random and uncorrelated in time. Furthermore, the SL spectrum tends to be characteristic of the liquid rather than the gas. Figure 1 shows a typical spectrum obtained from an acoustic horn.

If the various atomic transitions are known, then synthetic spectra can be generated that can be matched to the

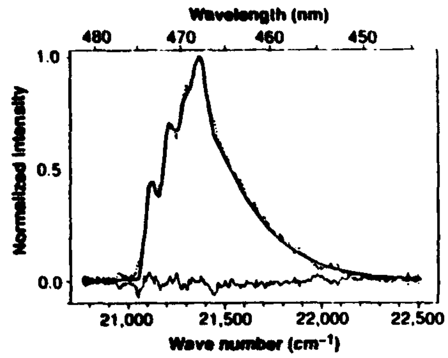

FIG. 1. Sonoluminescence spectrum of silicone oil under continuous Ar sparge at 0 °C as generated by an acoustic horn. The dotted line shows the experimental spectrum; the boldface line shows the best fit synthetic spectrum, assuming emissions from the Swan band of C_2, with $T=4900$ K; the thin line shows the difference spectrum [from Flint and Suslick (1991)].

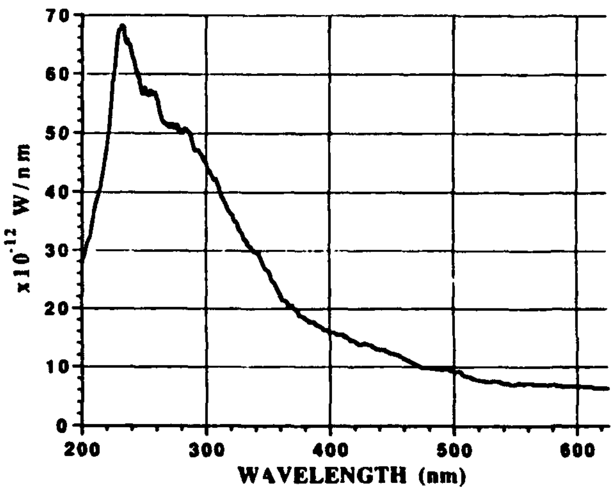

FIG. 2. Calibrated spectral density of the synchronous picosecond flashes of sonoluminescence at 22 °C for an air bubble in water. The fall-off below 240 nm is thought to be due to absorption in the liquid and the containing system. If this spectrum is assumed to be that of a blackbody, and a best fit made to the data, an "effective temperature" of approximately 40 000 K is obtained [from Hiller *et al.* (1992)].

measured spectra, with the single fitting parameter being the temperature. Flint and Suslick (1991) have obtained excellent fits to spectra such as these and have thus determined the "temperature" of sonoluminescence. With a fit similar to this one, a temperature of 4900 °C was obtained for an argon bubble in silicone oil. It is important to note that this spectra is characteristic of the host liquid, not the gas. In particular, the spectrum in Fig. 1 represents the Swan band emissions from C_2 in the $d^3\Pi_g$ excited state. This "effective temperature" may not necessarily represent the temperature of the gas because this excited state may not be fully equilibrated with other species within the cavitation bubble. In particular, one should expect significant temporal and spatial variability for this system.

Thus, in general, we see that in sonochemistry, using an acoustic horn or a similar source to generate extended (in space and time) regions of acoustic cavitation, SL spectra can be obtained that have well-defined peaks that correspond to atomic transitions in the host liquid, and can be associated with a temperature of roughly 5000 K.

II. SONOPHYSICS

In the recent sonophysics experiments of Putterman and his colleagues, significantly different results are obtained for the behavior of SL. In this latter case, a single gas bubble is acoustically levitated in a liquid contained within a resonant chamber. The gas bubble is driven in a volume mode at relatively large radial excursions and is observed to be remarkably stable, remaining at a fixed position and oscillation about an equilibrium size that remains unchanged over millions of cycles. Under relatively easily attainable but considerably specialized conditions, the bubble is observed to emit a steady light which can be determined by photometry to be reproduced repetitively every acoustic cycle.

One of the most remarkable discoveries by Putterman's group was that the lifetime of the SL flash was not measurable by standard techniques, i.e., currently available photodetectors were unable to resolve the rapid rise time of the flash. A conservative value of 50 ps was assigned as a best estimate of an upper bound. Furthermore, the synchrony of the flashes was also found to be remarkably stable, with an intrinsic "jitter" of less than 50 ps.

Shown in Fig. 2 is a spectrum obtained by Hiller *et al.* (1992) for a stable air bubble in water. Note the absence of any major spectral peaks and also the apparent extension of the spectrum into the ultraviolet. When Fig. 2 is compared with Fig. 1, it appears as if these two phenomena are fundamentally different.

Thus, we see that for sonoluminescence physics (sonophysics), in which a single, stable, gas bubble is made to emit electromagnetic emissions, the spectrum is representative of a blackbody, there are no spectral peaks, the lifetime of the pulse is very short (<50 ps), and the effective temperature is on the order of 40 000 K.

III. RESOLUTION OF THE PROBLEM

These intrinsic inconsistencies can be resolved only if the physical mechanisms that give rise to the respective SL emissions are fundamentally different. It is well known that when cavitation bubbles are permitted to collapse near a boundary, either soft or hard, the collapse is asymmetric and instabilities develop that grow without bound. Consider Fig. 3 which shows high-speed photographs of a cavitation collapse near a rigid boundary.

Note that for an asymmetrical collapse, portions of the host liquid are delivered to the center of the bubble. Because the liquid is an immense heat reservoir, a typical temperature profile within the bubble indicates that although the temperature at the center of the bubble may be several thousands of degrees, the temperature near the bubble wall must be near that of the liquid [Kamath *et al.*, 1993]. For the case shown in Fig. 3, it is seen that a liquid jet develops that penetrates into the interior of the gas bubble where the temperature is elevated. In this rather atypical case, it is also seen that small droplets of liquid can

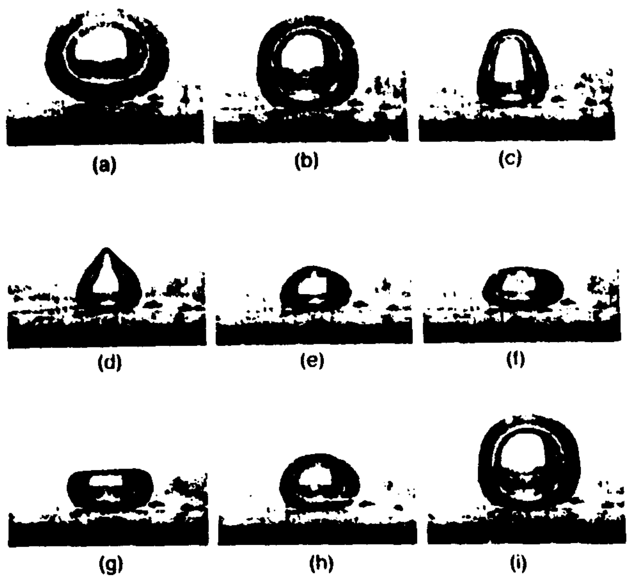

FIG. 3. Cavitation bubble collapse near a rigid boundary. In this case, the liquid was an aqueous solution of glycerin (30% by volume), the ambient pressure was near that of the vapor pressure of water, and the driving frequency was approximately 60 Hz. The maximum size of the bubble was approximately 2 mm [from Crum (1979)].

be deposited within the interior of the bubble, which would then be heated much more effectively during a subsequent collapse than would the liquid near the bubble interface. Thus, when asymmetrical bubble collapse occurs, the liquid can be elevated to high temperatures and spectra characteristic of the liquid rather than the gas can be observed.

The conditions under which asymmetrical bubble collapse would be more common are those in which an acoustic horn or a similar source is used to generate extended areas of cavitation throughout the bulk of the liquid. In this case, the presence of many bubbles triggers the collapse asymmetry by providing many pressure-release boundaries within the liquid itself. If a bubble is caused to collapse in the vicinity of another bubble, then the presence of this pressure release surface itself is an effective boundary. Asymmetrical bubble collapse due to the presence of other bubbles has been demonstrated by Tomita and Shima (1990). Thus, when "multiple-bubble" sonoluminescence occurs, the following behavior is observed.

(1) The cavitation bubbles tend to collapse asymmetrically, thus introducing liquid into the interior of the bubble, which is heated by adiabatic compression.

(2) The spectrum of multiple-bubble SL is dominated by the characteristics of the liquid rather than the gas.

(3) Because the symmetry of the collapse is destroyed, the final temperatures achieved in this case are relatively low.

Consider next the case in which a single, stable gas bubble is driven at sufficiently large volume oscillations to produce SL. In "single-bubble" sonoluminescence, symmetric bubble collapse is much more likely to occur, and conditions can develop that are unachievable in the multiple-bubble case. In fact, considerable evidence has been presented (Barber and Putterman, 1993) that a shock wave develops within the gas and is responsible for the enormous temperatures achieved. Thus, when "single-bubble" SL occurs, the following behavior is observed.

(1) The cavitation bubbles tend to collapse symmetrically, thus developing an imploding shock wave within the gas.

(2) The spectrum of single-bubble SL is dominated by the characteristics of the gas (and vapor) and tends to approach that of a blackbody.

(3) Because the symmetry of the collapse is preserved, the final temperatures achieved for this case are relatively high.

Finally, if the scenarios proposed here are correct, there are several experimental tests that could demonstrate the differences between these two phenomena. For example, in single-bubble SL, the shock wave that develops within the gas should also exist within the liquid, even at these low driving amplitudes. Similarly, these shock waves would probably not exist in *low-amplitude* multiple-bubble SL. (Of course, when driving amplitudes become large, shock waves appear to exist even in cavitation fields involving many bubbles.) A second test would be the duration of the SL flash. In single-bubble SL, it has been determined to be very short—as a consequence of the shock wave; in multiple-bubble SL, it should be much longer—because the temperature elevation comes from adiabatic heating. Finally, the rapidity of the collapse in single-bubble SL should eliminate the effects of heat conduction, while the longer collapse times in multiple-bubble SL should enable it to be expressed. Thus, in single-bubble SL, the presence of high thermal conductivity gases such as xenon should not have much effect on the final temperature, while in multiple-bubble SL, it should have a considerable effect.

IV. CONCLUSIONS

There are two types of sonoluminescence: multiple-bubble SL and single bubble SL. The former is important in sonochemistry and cases in which the cavitation is extended throughout the liquid. In multiple-bubble SL, cavitation bubble collapse is asymmetrical, which results in lower SL temperatures, but elevates the temperature of the liquid to incandescence temperatures. In single-bubble SL, in which a single, stable, pulsating gas bubble is driven at large radial excursions, cavitation bubble collapse is symmetrical, which results in higher SL temperatures. These effects occur principally within the gas and are of interest for their fundamental sonophysics.

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Origins of Single-bubble Sonoluminescence

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Abstract

The discovery of single-bubble sonoluminescence [Gaitan and Crum, 1988] has lead to several interesting and remarkable observations [Barber and Putterman, 1991]. Among these are picosecond-length light flashes and a level of syncronicity several orders of magnitude greater than the period of the applied acoustic field. Although new and unique observations concerning this phenomenon are being rapidly reported, an adequate explanation for the physical mechanisms that give rise to single-bubble sonoluminescence has never been given. We present here evidence that this phenomenon arises from nonlinear aspects of bubble dynamics coupled with the process of rectified diffusion. Our results suggest the presence of multiple stability locations that depend upon the driving acoustic pressure and the equilibrium size of the bubble.

Introduction

When an acoustic wave propagates through a liquid, certain conditions can be attained in which the mechanical energy associated with the acoustic field is converted into electromagnetic energy. This process, typically intermediated by acoustic cavitation, is called *sonoluminescence*, and is the principal focus of this article.

When cavitation is generated in a liquid, multiple cavitation sites appear with the result that the process is not localized but spatially and temporally distributed over a relatively large parameter space. Fig. 1 below shows a photograph of sonoluminescence (SL) generated by a therapeutic ultrasound device [Crum et al, 1987]. Note the localization of SL into bands associated with standing waves in the liquid, but also the distributed nature of the process throughout the bulk of the fluid. Further, this photograph is a time-exposure (about 15 sec.); on an instantaneous basis, one sees random flashes of light from the SL zone that gradually build into the geometrical configuration presented here [Crum et al., 1986].

Fig. 1. Photograph of sonoluminescence produced by a therapeutic ultrasound transducer, shown at the top; here the driving frequency was about 1 MHz and the acoustic pressure amplitude was about 0.15 MPa. The width of the transducer is approximately 2.5 cm.

It has been difficult to determine the basic physical processes that give rise to SL, partly because it has been practically impossible to spatially and temporally control the production of cavitation, the origin of SL. It appears to occur randomly over a relatively large spatial area, as shown in Fig. 1 above. However, the fortuitous discovery of SL from a single stable cavitation bubble by Gaitan et al., [1992] has now made it possible to study the phenomenon in much more detail than was previously possible. With this system the dynamics of a single cavitation bubble can be studied simultaneously with the physical processes that lead to SL, thus isolating the critical temporal and spatial parameters that give rise to SL. We shall first present some background material on SL in general, and then describe the process of single-bubble SL. Finally, we shall present some preliminary evidence that suggests the reason for the existence of this phenomenon.

Background

SL was discovered nearly 60 years ago [Marinesco and Trillat., 1933; Frenzel and Schultes, 1935], and since then there have been a variety of explanations given for the origin of the electromagnetic emissions. Electrical discharge theories of various types were at first quite popular. As early as 1940, Frenkel [1940] suggested that electrical charges known to exist on the surfaces of bubbles (see for example, [Watmough, et al., 1992]) were somehow made to discharge. This model, though seriously challenged by the experiments of Suslick [Suslick, 1989;1990], also has its modern advocates [Margulis, 1992; Lepoint and Mullie, 1993]. However, the results summarized below strongly support the hot-spot model of Noltingk and Neppiras [1950] which posits SL to be the result of incandescence of the bubble's contents. Nonetheless, there are still many unanswered questions concerning the origin of this phenomenon.

In 1959, Strasberg [1959] discovered that a stationary sound field could be used to levitate a gas bubble in a liquid. Since then, this technique has been used by a number of researchers to determine various aspects of bubble dynamics [Crum, 1980; 1983; Crum and Prosperetti, 1983]. Recently, Holt and Crum [1987; 1992] developed a technique that enabled real-time measurements of the radius-time curve for an oscillating gas bubble to be obtained; they used this technique to investigate the behavior of bubbles that were driven into nonlinear volume oscillations--a major aspect of acoustic cavitation and SL.

In 1988, when Gaitan was studying the conditions necessary for sonoluminescence (SL) during stable cavitation, he discovered that under certain fairly restrictive conditions, a single, stable gas bubble could produce SL emissions each cycle [Gaitan et al., 1988]. Using the light-scattering technique developed earlier [Holt and Crum, 1987; 1992], Gaitan was able to demonstrate the SL emissions were indeed coming from a single gas bubble and that they were being emitted at the final stages of gas bubble collapse. Figs. 2 and 3 below show the light scattering technique used to obtain the radius-time (R-t) curve and the phase of the SL emissions. (These experimental systems are described in considerable detail in previous publications, [Gaitan and Crum, 1990; Gaitan, et al., 1992] and will not be described again here.)

Fig. 2. Experimental arrangement used to levitate a gas bubble and to obtain SL emissions.

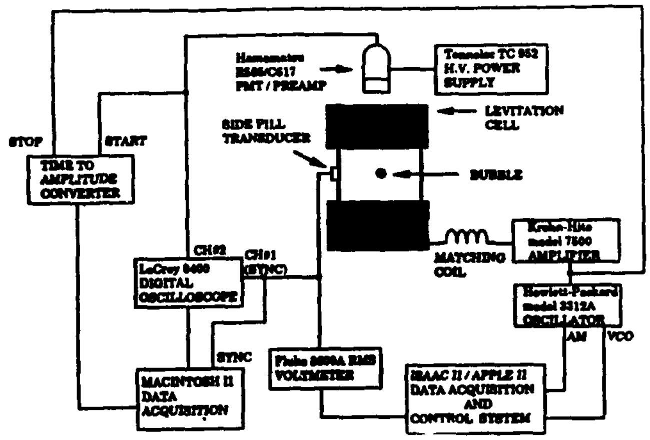
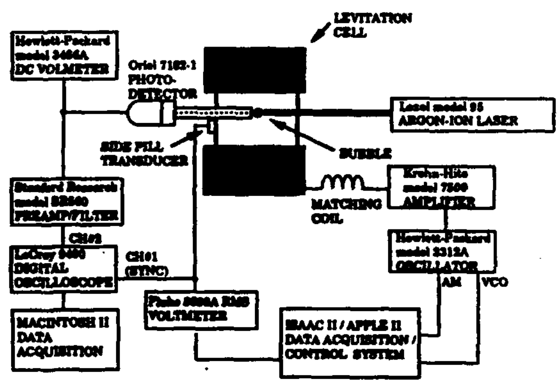
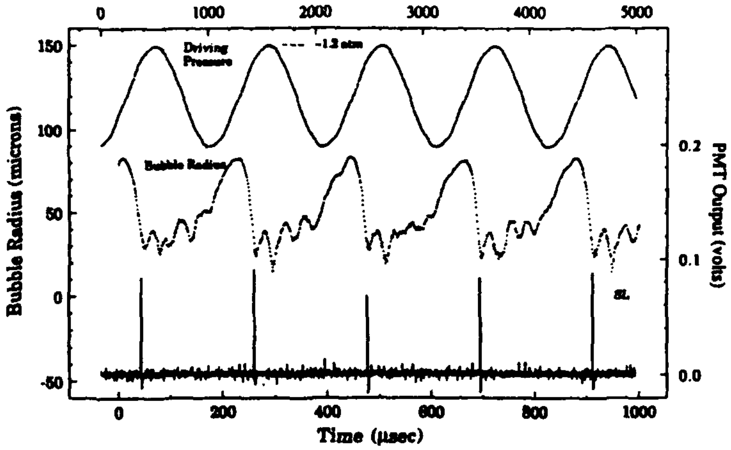


Fig. 3. Experimental arrangement used to obtain the radius-time curve for a luminescing bubble.



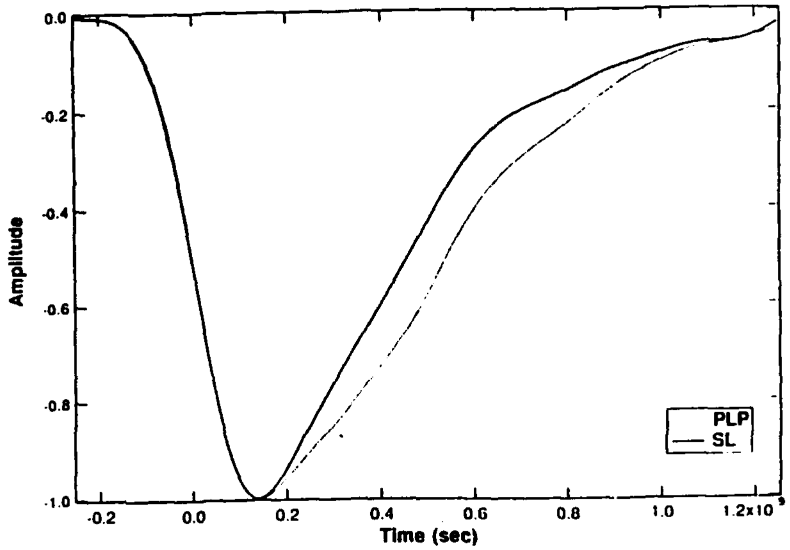
These two sets of apparatus were combined to obtain the following figure which shows "simultaneous" measurements of the R-t curve and the SL emissions. These are not truly simultaneous, but by synchronizing both the R-t curve and the SL emissions with the acoustic pressure, all three of these variables could be plotted as shown in Fig. 4 below. This technique was observed by Putterman in our laboratory and then duplicated in his own. A series of remarkable discoveries [Barber and Putterman, 1991; Barber, et al., 1992; Hiller, et al., 1992] were then made that has caused this phenomenon to attract international attention [Levi, 1991].

Fig. 4.. Synchronous relationship between the driving acoustic pressure (top trace), the bubble radius-time curve (middle trace) and the sonoluminescence emissions (bottom trace) for a gas bubble of approximately 25 μm in radius driven at an acoustic pressure amplitude of approximately 0.12 MPa and at a frequency of 22.3 kHz. Here the liquid was an aqueous solution of glycerin.



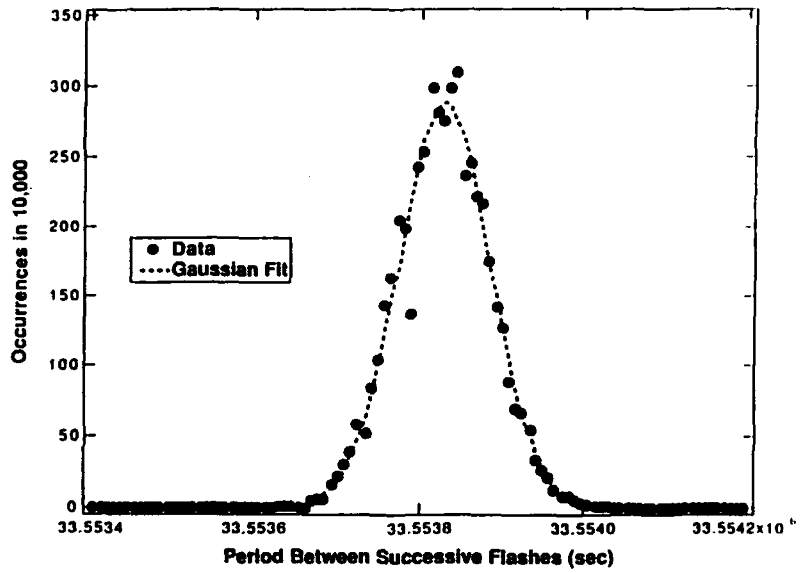
Barber, Putterman and their coworkers, working in clean, degassed water, have discovered that these light emissions are extremely short and remarkably repetitive. Shown in Fig. 5 below is the response of a state-of-the-art photodetection system. It is seen that the SL pulse is no more easily resolved than that of a 35 ps pulsed laser. It should be noted, however, that the PMT was not fast enough to resolve either of these events. From this data, Barber and Putterman estimated that the maximum duration of the SL pulse was 50 ps.

Fig. 5. Response curve of the PMT for SL and a pulsed laser (PLP). The tail in the response of the PLP compared to SL is thought to be due to ringing in the laser. {From Barber et al. [1992]}



An equally intriguing result is the phenomenon shown in the following figure, which shows the temporal "jitter" in the SL pulse from cycle to cycle.

Fig. 6. Distribution of events versus the period between flashes for SL emissions. {From Barber et al. [1992}



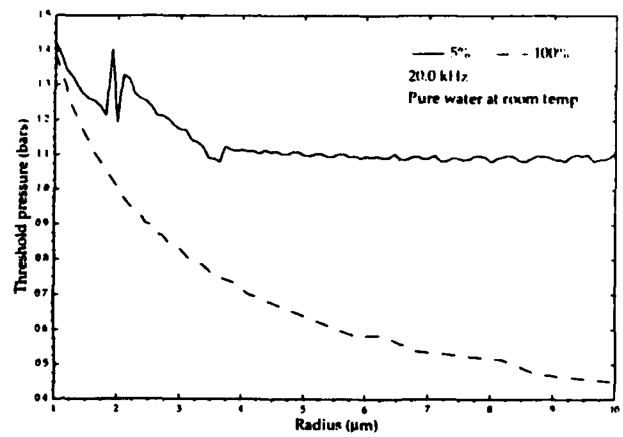
This result is remarkable because the half-width of the jitter is estimated to be on the order of 50 ps. Since the electrical voltage that drives the transducer is operated at a frequency of 20 kHz, this jitter represents an instability of only one part in 10^6 of the acoustic cycle! Surely the electronics of this system has an intrinsic jitter larger than this value. For unknown reasons, the bubble has "mode-locked" to an incredible precision; moreover, the jitter seems to be independent of the stability of the oscillator. Single-bubble SL is like a light source emitting 1.5 cm long light bursts, with a separation between the bursts of 15 km, and with an uncertainty in the position of the bursts of only 1.5 cm. Truly, this is an amazing natural phenomenon!

Results and Discussion

If one were to ask, *a priori*, if stable, single-bubble SL could exist--as it does now--most of us would be quite skeptical, because of rectified diffusion. It is difficult to imagine that a bubble of 5 microns, say, could remain at a fixed size for essentially an infinite amount of time. It should either grow or dissolve, but it seems unlikely that it would remain at a fixed size.

Using the nonlinear rectified diffusion code developed by Church, [1988], we have investigated this phenomenon for bubbles under the set of parameters similar to those experiencing sonoluminescence. Figure 7 below shows what we believe is an important result.

Fig. 7. Calculations of the threshold for rectified diffusion of gas bubbles in water for a driving frequency of 20.0 kHz and dissolved gas concentration of 5 % (solid line) and 100 % (dashed line).

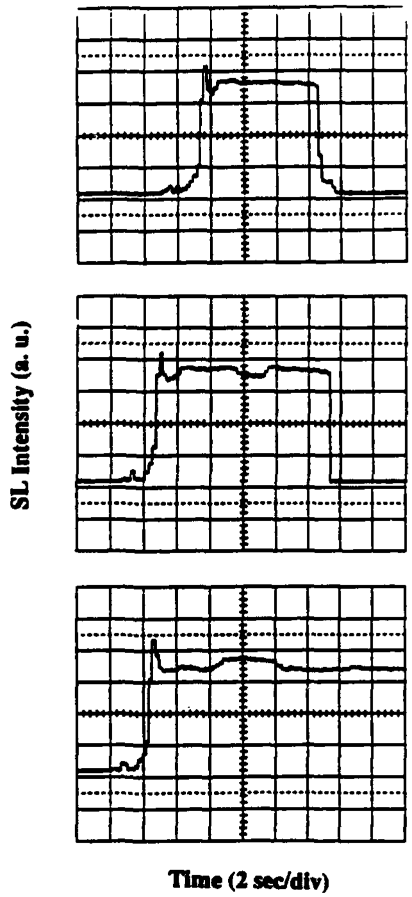


Note that when one reduces the dissolved gas concentration to the level desirable for single-bubble SL, a couple of "notches" appear in the threshold curve that could be very meaningful (these notches or depressions or valleys are due to the nonlinear response of the bubble and represent harmonic resonances). Consider the notch near 3.5 μm; this value of the radius is near that of the value measured by Barber and Putterman [1993] in their light scattering experiments. Suppose that a bubble were "positioned" within this notch (above the threshold), by selecting a bubble of about 3.5μm in radius and driving it at a pressure amplitude of 0.115 MPa (1.15 bars) and at a frequency of 20 kHz. This particular bubble would then grow until it engages the threshold curve at about 3.7 μm. At this point, if it grew further, it would pass into a region for which the threshold is higher than 0.115 MPa, and it would start to dissolve. As it got smaller, it would cross the threshold line once again, and get larger, etc. Thus, a positive slope on the rectified diffusion threshold curve is a point of stable equilibrium for a bubble driven at a fixed acoustic pressure amplitude.

For this bubble to produce SL flashes each cycle, it would seem necessary that shape oscillations not occur, because that should lead to asymmetrical bubble collapse, which would in turn, tend to prevent SL. It is difficult to make measurements in this region, of course, but the extrapolations of our earlier measurements and calculations [Horsburg, et al., 1990] suggest that the threshold for shape oscillations is larger than 0.115 MPa in this radius range (2-5 μm). Thus, it is plausible that this general region of parameter space is the location for single-bubble sonoluminescence.

Consider some further support for this contention. Figure 8 shows the response of a PMT to the "initiation" of single-bubble SL. For this case, a bubble was generated by electrolysis and allowed to rise into the antinodal region of a standing wave field (with the field inactivated); at this point, the field was activated and the time-averaged SL intensity observed as single-bubble SL was initiated.

Fig. 8. PMT outputs for the initiation of single-bubble SL. In this case a bubble was introduced into a standing wave field and the time-averaged SL intensity measured as a function of time. These traces are for three different events under approximately the same conditions. Note the repeatability in the structure of the initiation phase. In the top figure, the bubble went unstable, in the middle trace the field was deactivated, and in the bottom trace the bubble remained stable for the duration of the trace.



This figure shows three separate traces taken at different times and under slightly different conditions. Note that similar behavior was observed in each case: A small burst of light occurred for a second or so, followed by a rapid rise to a value in excess of the equilibrium value, and then a relaxation to a steady output. This behavior seems consistent with the proposed idea that the bubble will adjust its radius by rectified diffusion to reach some stable state. The time scales for bubble growth by rectified diffusion are on the order of seconds for this range of acoustic parameters [Crum and Hansen, 1983].

A second region of notches is also observed in Fig. 7 for a range of bubble radii from 1.5-2.5 μm . This lower-radius region occurs at a higher value of the acoustic pressure amplitude, and could represent a second (and even third, because there are two separate notches here) region of rectified diffusion stability. We believe we have seen evidence of this region.

Consider Fig. 9, which represents a long-term time series for the initiation and subsequent deactivation of single-bubble SL.

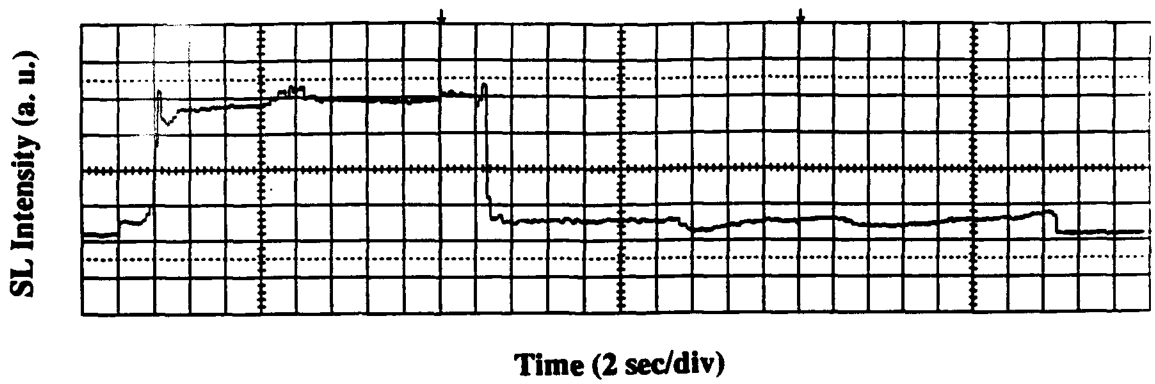


Fig. 9. PMT output for the initiation and stabilization of single bubble SL. In this case the bubble was initiated and stabilized, remaining in this state until the position of the arrow at the top. At this point the system was perturbed, and a transition occurred from a high SL output to a low SL output. The bubble stabilized again in this low output state and remained until the time indicated by the second arrow at the top. At this point, the system was again perturbed, and a transition from this particular state to the noise floor soon occurred.

In this instance, a bubble was activated; it followed the general behavior for initiation as seen in Fig. 8, and then was stabilized. However, in this particular case, the local conditions were slightly perturbed to the extent that after a few minutes, the bubble's intensity dropped to about 15% of its stable value. It remained at this level, with some slight drifts, for a few more minutes, and then dropped to the noise floor.

We wish to suggest that the bubble could have made a transition from the larger-radius location for single-bubble SL to the smaller-radius one. Keep in mind that it is difficult to maintain equilibrium in the liquid parameters. For example, a slight increase (or decrease) in temperature will significantly changes the level of the dissolved gas concentration. If the temperature of the liquid changes, then the threshold curve moves down (or up) with a result shift in the location of the stability regions. Furthermore, slight drifts in the temperature result in resultant changes in the modal frequency and antinodal position. Accordingly, although it may have been fortuitous that a transition from one stability location to another occurred, it's not an unlikely possibility. Finally, seems likely that a smaller radius bubble would have a lower SL intensity output, which would account for the observed behavior shown in Fig. 9.

These suggestions for the behavior of single-bubble SL are admittedly speculative. However, there seems to be some justification for the general arguments presented here. Related arguments have been presented previously by Kamath, et al., [1993]. A consistent and detailed explanation for this general phenomenon, however, awaits further research.

Summary

We have presented some preliminary evidence that single-bubble SL is the result of nonlinear bubble dynamics in which regions of positive slope occur on the rectified diffusion threshold curve. It appears that bubbles can be entrapped in these regions and, provided liquid and acoustics parameters remain unchanged, thus provide locations where individual bubbles can radiate SL emissions each cycle for indefinite periods of time.

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