The Origin of Sonoluminescence
(or measuring the size of the light-emitting region in a sonoluminescencing bubble)

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Introduction to sonoluminescence

The scope of the thesis
As the title (measuring the size of the light-emitting region in a sonoluminescing bubble) suggests I have had an experimental approach to the problem of determining what goes on inside a sonoluminescing bubble. The theoretical and numerical work in this thesis has been done to explain measurements, and not the other way around. In the search of effects that might be used to explain the size of the light-emitting region, we (Mogens Levinsen and I) have of course found other interesting phenomena and deviated from the set course to examine them. However, a deeper understanding of sonoluminescence has always been in the scope.

In my time as a master student and PhD student I have published and submitted the following papers [1,2,3,4,5,6,7].

History of sonoluminescence
In 1934 Frenzel and Schultes [8] discovered that photographic plates in an ultrasonic bath are darkened by weak luminescence in the water. This luminescence caused by sound is called sonoluminescence. In an ultrasonic bath many bubbles are created and collapse a few times, each emitting tiny amounts of light. The phenomenon is difficult to study due to its transient nature, and weak emission. Frenzel and Schultes quickly “moved on to more interesting subjects”. One could say that, for many years sonoluminescence lived quietly in the dark. However, in 1990 Gaitan [9] succeeded in capturing a single bubble and made it glow much stronger than usual multi bubble sonoluminescing bubbles. The discovery is known as single bubble sonoluminescence (SBSL). SBSL is much easier to study, since the light-emission is much more stable and strong, and the bubbles can be captured in a fixed position. This set off a lot more research on the phenomenon (according to google scholar ~1830 papers containing the term sonoluminescence have been published since 1989).
**General introduction to sonoluminescence**

SBSL is an intriguing phenomenon with surprising powers. Just the simple fact, that you can produce light, which is a highly concentrated form of energy, from vastly more diffuse energy (sound) is interesting on its own. That the bubble oscillations can exhibit accelerations of about $10^{11}$ G is none other than extreme. To compare accelerations a billion times smaller are lethal to humans. The speed of the bubble wall collapsing can be $\sim 3000$ m/s. That the bubble is heated to temperatures much warmer than the suns surface is also extreme. The gas inside the bubble is compressed close to a van der Waals hard core, with extreme pressures and temperatures as a result. The frequency of the sound field can be anywhere from $\sim 20$ Hz [10] to more than 1 MHz [11], although most experiments are done in the 20 to 50 kHz regime.

All this can be set off by just adding simple sound to slightly degassed water\(^1\). Amazing!

**Bubble dynamics**

The existence of a bright stable sonoluminescing bubble requires a multitude of conditions to be fulfilled. The pressure oscillations driving the bubble are typically around 1.4 bar at light emission. If too much gas is dissolved in the liquid the bubble will grow too large over time and become unstable. If too little gas is in the liquid the bubble will dissolve. Also a bubble has to be seeded in the water to begin with, as bubbles (usually) don’t spontaneously form. Furthermore, chemical reactions inside the bubble are important for the gas composition.

The bubble dynamics can be calculated numerically using the Rayleigh Plesset equation. The oscillations are extremely non-linear at the time of bubble collapse, so great care in the numerical calculations around bubble collapse must be taken. To make a good model many considerations has to be taken into account including chemical reactions, diffusion and liquid vapor, so a complete model is in no way easily accomplished. A good account of how to make a model is shown in a review by Brenner, Hilgenfeldt, and Lohse [12].

The Rayleigh-Plesset equation:

$$R\dddot{R} + \frac{3}{2}R\dot{R}^2 = \frac{1}{\rho} \left( p_g - p_0 - P(t) - 4\eta \frac{R}{R} \frac{\dot{R}}{R} + 2\sigma R \right)$$  \hspace{1cm} (1.1)

\(^1\) Light can actually be produced in air-saturated water as demonstrated by Krefting et al [64].
\( R \) is the radius of the bubble, \( \rho \) is the density of the fluid, \( p_g \) is the pressure in the gas, \( \eta \) is the shear viscosity, \( \sigma \) is the surface tension of the gas-liquid interface, \( P_o \) is the ambient pressure, and \( P(t) \) is the applied acoustic pressure.

To obtain good agreement to experimental observations of bubble oscillations, the Rayleigh-Plesset equation has to be extended with heat transfer, dampening of bubble dynamics by sound irradiated from the bubble, chemical reactions, and many other effects outside the scope of this thesis.

**Shape stability**

The shape stability of sonoluminescing bubbles has been investigated numerically [13,14,15]. The most complete analysis is done by Augsdörfer et al [16] (see also elaborating comments in [12]), who calculated the shape instability boundaries for \( n=2 \) and \( n=3 \) spherical harmonic deformations (take a look on Fig. 6 in [16]) Augsdörfer predict \( n=2 \) instability for bubbles larger than 4.6 \( \mu \)m and \( n=3 \) instability for bubbles larger than 6.6 \( \mu \)m. Following the notation of Augsdörfer et al a shape deformation can be written as:

\[
R_{\text{distorted}}(t) = R(t) + \sum_{n=2}^{\infty} \sum_{m=-n}^{n} a_n^m(t) Y_n^m(\theta, \phi)
\]

\( Y_n^m(\theta, \phi) \) is a spherical harmonic and \( a_n^m(t) \) is the time-dependent amplitude thereof.\(^2\)

Augsdörfer et al use a linear stability analysis and thus cannot predict whether the deformations result in stable shape distorted bubbles, or if it eventually results in the bubble breaking up/micro bubbles pinching off. With experimental evidence of stable shape distorted bubbles (see eg. [17,1,2,3]), their model should be improved by non-linear factors, which must provide stabilizing factors. Augsdörfer et al show the spherical harmonic instabilities only depend on \( n \) (and not \( m \)).

The model presented by Augsdörfer [16] predicts where in the size/acoustic pressure space period doublings of \( n=2 \) and \( n=3 \) types are observed. The paper by Augsdörfer [16] combined with Hilgenfeldt et al [14] in a way predict shape deformed period doubling. On page 6 (below Fig. 4) in their paper Augsdörfer write: “If strong surface waves develop during the after bounces and restoring forces are weak, distortions may not be entirely stretched out during the next

\(^2\) One could argue that the sum over \( n \) should be from 1 to infinity. But for this thesis \( n=2 \) to infinity is fine.
expansion phase. As a consequence the deformation developed in the first cycle will experience a second and subsequent amplification in the following cycles. A third instability mechanism, referred to as parametric instability is responsible for the build-up of the shape instabilities over more than one cycle…”

Shape distorted period doubling could be considered predicted when combined with a quote from Hilgenfeldt et al: “It is also seen that the dynamics of the distortion $a_x(t)$ has half the frequency of its forcing bubble dynamics $R(t)$ as typical for an instability of the Mathieu type.”

**Argon rectification and chemical reactions**

A good liquid in which to make single bubble sonoluminescence is water. A good gas to have dissolved in the water is air (typically around 200 hPa). But what happens to the air bubble as it collapses and is heated to extreme temperatures?

Dry air is composed of approximately 78% Nitrogen, 21% Oxygen, and 0.934% Argon [18]. The bubble gets hot enough to make N₂, O₂ and H₂O react and form nitrous oxides and other reaction products. This was shown experimentally by Didenko and Suslick [19], the idea was proposed by Lohse and co-workers [20] and supported by numerical calculations by Storey and Szeri [21]. A thorough investigation of the possible chemical reactions in an air-bubble in water has been done by Yasui in [22].

The result is a bubble comprised of almost pure Argon (dubbed Argon rectification). This is especially important when estimating the collapse temperatures, because a noble gas gets warmer from compression than air. The $\gamma$-coefficient $\left(\frac{C_p}{C_v}\right)$ for a noble gas is 1.67, whereas it is only 1.4 for air.

Furthermore chemical reactions in air (e.g. splitting of a Nitrogen molecule) cost energy, inhibiting extremely high temperatures. The recognition that the stable light-emitting bubbles are in effect Argon bubbles is of vital importance in understanding SBSL.

For multi-bubble sonoluminescence the bubbles do not survive for a long enough time to rectify Argon, thus multi-bubble sonoluminescence with air bubbles cannot reach the higher temperatures (and light-emission) of SBSL bubbles.
Theories of sonoluminescence

How come that the bubble emits light? What physical mechanisms are responsible? I really would like to give a definitive answer to those questions, but let’s discuss the different main contenders to explain the light emission, and in the process discuss how uniform the interior of the bubble is at light emission.

**Blackbody radiation**
For a body to emit blackbody radiation, it must be black to the relevant wavelengths, which mean the mean free path of photons should be low compared to the size of the bubble. Measurements on a hydrogen bubble in water have been done by Vazquez et al in [23]. Those measurements of the light spectrum closely match that of a blackbody at a certain size and temperature. The size of the light-emitting body appears to be smaller than the bubble itself. More often than not blackbody radiation is a good fit to measured spectra, and thus the conclusion that the mechanism behind light emission is blackbody is tempting.

The apparent temperature of a blackbody can be much lower than the core temperature of the blackbody. The apparent temperature will be from the region where the mean free path of photons gets sufficiently high to allow the photons to escape the blackbody. E.g. the sun emits 5-6000 K blackbody radiation, but the core temperatures are orders of magnitude higher. This could also be the case for our “star in a jar”.

**Bremsstrahlung**
The rapid acceleration of charges results in photons being emitted. A fast electron passing close to a nucleus is accelerated by the coulomb forces, resulting in photons being emitted.
In a paper by C. Camara et al [24] a small bubble driven at 1 MHz is examined and the spectrum is fitted much better by bremsstrahlung than blackbody radiation. Their proposed explanation for this is that since the bubble is much smaller, the light-emitting region gets so small that the mean free path for photons now is larger. They fit their measured spectrum to 1,000,000 K bremsstrahlung. If this
fitted temperature in the paper is to be taken literally, the small bubbles do reach extremely high core temperatures.

**Hotspot or uniformity?**

Whether the bubble has more or less uniform temperature and pressure inside, or there is significant pressure and temperature gradients inside the bubble is not agreed upon by researchers in the field.

One idea proposed to explain increased core temperatures has been some sort of shock wave converging on the central point resulting in extremely high pressures and temperatures, like the model calculations presented in [25]. If not a shock wave, a compression wave could have similar effects, as simulations by Cheng *et al* [26] show.

One try at giving a final description of Single Bubble Sonoluminescence was done in a paper by Hilgenfeldt, Grossmann, and Lohse [27], where there is assumed equal temperatures and pressures throughout the bubble. This paper has been criticized by Putterman *et al* [28], and again defended [29]. In the model by Moss *et al* [30] there is a small light-emitting region inside the bubble, whereas other models like one from Yasui [31] assumes equal temperatures and pressures except from a thin boundary layer.

The controversy can, as I see it, be boiled down to whether or not there is a significant temperature and pressure gradient. If it can be experimentally established whether the light originates from a large region inside the bubble or rather from a smaller region, then good progress in the understanding is made. This has been my goal to find out, and many of the measurements I have done and models I have proposed, have at least initially revolved around this issue.

**Hanbury-Brown Twiss intensity interference**

Astronomers measure the size of a star by a method developed by Hanbury-Brown and Twiss [32]. The same method can be applied on a much smaller scale, namely our star in a jar. Theoretical estimates for sonoluminescence have been made by Hama *et al* [33] and Slotta *et al* [34]. Estimates similar to the latter has been done by Putterman [35] and in my master thesis [36]. However, all estimates predict small correlations, which make it quite difficult to measure, firstly because the dataset has to be large, and furthermore because even small systematic errors and disturbances can cause much larger effects.

Using 1 nm filters for 400 nm wavelengths, the theoretical HBT-correlation for a 100 ps flash (according to [34]) is about 0.0002. To measure this value with some std. dev. certainty requires correlating about $10^9$ photon events. If the two channels
you correlate measure a photon 10% of the time, then you have to measure for $10^{11}$ flashes 2 months non-stop. In other words it easily takes a week or a month to acquire enough data to realize something is wrong. This makes it a very cumbersome and time consuming measurement, if not bordering on impossible. I spent a lot of time during the work for my master thesis trying to measure this effect (without success as you can guess).

A shape distorted bubble can easily produce correlation of order 1.5, so no shape distortion can be tolerated. Thus one has to do the measurements at low argon pressures and small bubbles, and continuously monitor for shape distortions. It is difficult, but it can be done.

I have shown (as you will see later) that particles near the bubble can create small angle shades and constructive interference. Particles can shade the light almost completely. This creates small angle correlations in the light-measurements. Thus the experiment cannot succeed if particles are near the bubble. As the bubble has a tendency to catch and hold on to particles so they stick around for extended periods. This is a big problem for HBT measurements.

As destiny turns out, the two major sources of problems for HBT correlations, namely shape distortions and particles, give insight to both the size and nature of the light-emitting region. In a way, one could say that the problem turned out to be the solution.

**Sonofusion**

A Science paper by Taleyarkhan et al [37] published in 2002 claims evidence of nuclear fusion in sonoluminescing bubbles in deuterated acetone. The claim is later repeated in [38,39]. In the history of tabletop fusion one must mention cold fusion by Pons and Fleischman [40]. The cold fusion story sparked an intensive research effort, and a lot of time and effort was wasted trying to reproduce the irreproducible. The claim of bubble fusion has not sparked the same amount of research, but nonetheless it has to be checked if the claim is erroneous. Often bubble fusion is referred to as cold fusion, but it is not claimed to be cold. The extreme temperatures and pressures in the bubble are claimed to be responsible. But unfortunately there seems to be many similarities to cold fusion. Several groups around the world have tried to reproduce the observed fusion, but none other than Taleyarkhan and his students [41] seem to be successful. Recently Taleyarkhan published his measured neutron spectrum [39]. However, the measured neutron spectrum, at least according to B. Naranjo [42], who has some experience with real (read undisputed) tabletop fusion [43], the spectrum does not fit that of neutrons from fusion, but rather that of a common laboratory neutron source, Californium.
Apparently Rusi Taleyarkhan has sabotaged some colleagues attempts at reproducing his experiments, and an investigation [44] on the matter has been conducted by the Purdue University, furthermore the US patent office has rejected Taleyarkhans patent application [45]. The investigation by Purdue University is now completed, but the results of the investigation are kept internally [46] at Purdue although they were originally promised to be publicly available.

On the other hand, we must remember that failing to produce bubble fusion in deuterated acetone, does not exclude the possibility of making acoustically driven fusion in other systems. The bubbles undeniably do produce high temperatures and pressures, so I think it is worth investigating.

One problem arising from these apparently false claims (not being reproduced after many years of trying is not a good sign), is that many people (including funding agencies) now will run away screaming by the mere mention of bubble fusion. On the other hand it has spurred some research effort, that may be able to produce bubble fusion.

Calculations predicting fusion such as the one presented by Nigmatulin et al in [47] rely heavily on non-isotropic conditions inside the bubble, or rather, an extremely hot and dense core ($10^8$ K and 10 g/cm$^3$) in the center of the bubble. Their model attains these temperatures and densities by shockwaves focusing on the bubble center.

One interesting paper in that regard is the paper by Camara et al [11] where the light-emitting region in their model calculations does appear to be small and very hot ($10^6$ K).
Experimental setup

I have built and used three experimental setups. I will briefly describe each and point out their strengths and weaknesses.

**Eight channel multifunction shape, size and spectrum analyzer**

This setup is designed to examine the angular dependence of period doubling and also to make time resolved crude spectra of bubbles.

![Diagram of light-triggered data acquisition setup](image)

**Figure 1:** Light-triggered data acquisition setup. Eight PMT’s provide intensity measurements, and one PMT is positioned to collect as much light as possible to provide a timing signal for the data acquisition.

The setup is based on nine PMT’s positioned around a cylindrical cell all inside a refrigerator usually cooled to around 9°C. Up to eight PMT’s can be equipped with optical filters to form a crude spectrometer. The PMT’s used are R2076 from Hamamatsu in case of a filter below 300 nm, otherwise we use R3676 from
Hamamatsu. The gain on the PMT's can be adjusted by varying the high voltage supplied to them. The outputs from the PMT’s are amplified by amplifiers (Hamamatsu C7319) with variable gain in steps of 10. However the amplifiers give out slightly different shapes depending on amplification, so we prefer having all amplifiers set to the same amplification. The outputs of the amplifiers are fed into two ADlink 9812 data acquisition cards. The amplifiers are best suited for measurements at frequencies lower than 25 kHz, since at higher frequencies the amplifiers have not come to rest, so some signal will be carried over from the previous flash, slightly altering measured period doublings and other fast changes in intensity. This is only a problem (although small) when we drive the cells at overtones, since the fundamental frequency of the cells used is typically around 23 kHz.

The signal from the 9th PMT is used as a clock signal for the data acquisition. The ADlink cards require two clock signals to make one acquisition. Thus the clock signal from the 9th PMT has to be first put through a discriminator (Ortec 583) followed by a delay and pulse doubling circuit.

The ADlink cards have digital inputs as well, and I use one of those bits to check if the clocking system clocks perfectly or not. I have built a small digital circuit that flips between 0 and 1 once per acoustic cycle. Thus the digital signal should ideally alternate between 0 and 1 for successive data points. This is almost always the case for stable bubbles, however for weakly light-emitting bubbles (including recycling events) some clocks are missed and hence deviations from the usual alternation between 0 and 1’s are observed. This information of imperfect clocking is shown on the data acquisition programs user interface, and also can be dug out in later data analysis.
The cells used are cylindrical, 6 cm diameter and usually between 5 cm and 7 cm high. The cells equipped with a large piezoelectric crystal on top, are driven by a computer-controlled function generator (Agilent 33220a) whose signal is put through an amplifier (ILP HY2005). The Q of the acoustic system should neither be too large nor too low. A large Q (~1000) results in frequency instability, where a small temperature change results in a large difference in resulting amplitude. Also if the Q is too low we cannot produce enough amplitude in the sound field. A suitable Q for our setup is around 100. Some experiments were done with a cell with smaller piezoelectric crystals similar to the ones used in the next section.
Figure 3: Side view and top view of cylinder. A 5 or 6 cm tall quartz cylinder (6 cm diameter) is mounted with aluminum end caps. The caps are held in place by metal rods which are securely fastened. Two studs are used to fill in water, and let gas out respectively.

The computer can turn the output from the frequency generator off, and is able to control the frequency and amplitude with a user friendly interface. Since increasing the amplitude too rapid can in certain cases result in bubble extinction, the computer is programmed to be able to spread out a user selected amplitude increase in many smaller steps. The amplitude and frequency can be adjusted in user selectable step sizes for convenience.

The computer acquires data in double buffered mode, and presents the data on screen as they are acquired. This allows the user to immediately see how the bubble responds to a changed frequency or amplitude. The amplitude and frequency is controlled directly from the program and automatically logged along with time stamps and saved data to be used in later data analysis. Another computer controlled phase locked function generator outputs a same frequency saw tooth function, which usually is put in to one of the eight AD channels to

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3 Double buffered data acquisition is where data is acquired to a relatively small piece of memory with periodic boundary-conditions so to speak. When half the memory has been filled, a flag is raised and the memory portion can be read, processed, displayed and saved to file. This happens about five times per second in my program.
monitor how the relative timing between flash and acoustic sound field changes over time. This information can be used to assess the size of the bubble, since a larger bubble collapse later in the acoustic cycle [48]. New bubbles are seeded by a computer controlled relay connected to a resistor inside the cell or by dissociation of H₂O to O₂ and H₂ bubbles by the same relay putting a DC current through the water via electrodes.

**Degassing system**

Our degassing system is used to mix gasses, degas water and finally add the gas mixture to the degassed water.

![Degassing system diagram](Image)

**Figure 4: Degassing system. Arrows indicate valves or clamps on tubes. The arrow near the gas mixing system indicates a needle valve, so gas can be let in slowly.**

Our gas mixing system is built with a gas reservoir connected to its own vacuum pump (not water tolerant). Two gas pressure bottles can be connected to the system. A pressure gauge monitors the pressure in the tubes leading to the reservoir. When mixing gas, one must first pump the reservoir and tubes to vacuum, and then let in the desired amount of the first gas species. The reservoir must then be sealed, and the tubes pumped to a vacuum again. The second gas species must then be filled into the tubes to a pressure larger than the pressure in the reservoir. Finally the valve to the reservoir must be opened and additional gas be added to the system, until the desired total pressure has been reached. Finally the tubes must be pumped to vacuum, since they only contain the second gas species. The gas should be left in the reservoir to mix for a time. If this procedure is not followed, one can easily end up with one gas in the tubes, and hence only this gas will be let in to the water. Furthermore, as soon the desired total pressure is reached, it is important to close the valve to the reservoir as quickly as possible, to prevent the first gas species escaping the mixing reservoir.
Gasses can also be mixed directly in the water chamber. This is appropriate if mixing a small amount of gas with a large amount of another gas. E.g. when mixing 1% argon in nitrogen. When making completely argon-free measurements flushing the water with nitrogen gas helps removing all the argon. This is done by adding and removing pure nitrogen gas in ~10 minute intervals.

**Data acquisition and data analysis programs**

When examining a bubbles reaction to changing applied acoustic amplitude and frequency, it is convenient to have data acquisition with real-time data analysis on screen. To that end, the data acquisition program will show time-averaged data from two selectable channels, updating on screen about 5 times per second. This allows the user to immediately observe the results of modifying parameters.

The data acquisition program logs the applied frequency and amplitude, as well as a time-stamp for each data set collected. The intensity of the individual light flashes in all channels are saved to disk for later detailed analysis. Data is segmented into files containing 3,000,000 flashes for convenience. The data acquisition program saves the data in a compressed format [49] to conserve disk space. When a new data acquisition is started, the program first acquires a full speed sample of the four input channels on each data acquisition card and saves it to disk. The program thus acquires 8000 sample points at 20 MHz on each channel (referenced as “shapetest” in the program). This corresponds to about 9 acoustic cycles, where one later can see the typical shapes of the signals acquired. This is valuable for later data analysis, where one can check that all raw signals look nice with low noise levels etc. It can also be used to determine voltage offsets for the different channels. It can be used for regular data acquisition as well, if one does not need long time-series, but rather high temporal resolution (e.g. to measure if the time between flashes is constant). For very high temporal resolution a fast oscilloscope is still better with its higher sampling frequency.

In the following figures I show screenshots of the program, where the first one is from real-time measurements appearing on screen, and the latter is a sample of “shapetests”.
Figure 5: Data acquisition program. This program has too many features to describe here, but the most convenient of them include continuous logging of time of data acquisition and the frequency and amplitude settings of the frequency generator. The effect of changes to the sound field is immediately visible on screen (e.g. an intensity increase). The program also does a quick analysis for the period doubling and fits a \( \sin(2\theta) \) function and displays the fit in a polar plot. 2 (user selectable) of 8 channels are displayed with user selected temporal averaging. Screen updates with \(~5\) Hz.
Figure 6: Data acquisition program with a “shapetest” measurement. The signal shapes of the 8 channels put in to the program is measured with 20 MHz sample rate. This happens when the user pushes “ShapeTest” and each time a new measurement is started as well. The black “sawtooth” signal is used to determine the relative timing of the bubble collapses.

The program for data analysis reads in both the regular data files produced by the data acquisition program, as well as “shapetests”. All 8 channels are simultaneously displayed on screen. In the program the user can easily adjust the temporal averaging applied, as well as zoom in on interesting time-pieces in the data file. The program can average even and odd data points separately if one is looking for period doublings. For period doubled data, the program fits a sine function to show how good the data points lie in correspondence to the expectations, and can do statistical analysis like calculating the standard deviation on the average value, and reduced chi-squared tests on the fits - all done with a single click on the mouse. The program can also display the intensities of the odd and even data points (at a user selected point). Data shown on screen can be exported to a text file for processing in other programs. Samples at specific time-points can be marked and exported to a file.
Figure 7: Program for data analysis. The voltages measured on 8 channels are shown as a function of time, in this case odd and even flashes are shown with separate colors clearly portraying the period doubling. The shape distorted nature of the period doubling can be seen from the fact that for some channels the odd flashes are brighter and vice versa. As can be seen from this screenshot, this program has a lot of functions built in. A click on the screen gives you all the information you can ask for, in regards to the amplitude of period doubling, as well as a fit to a $\sin(2\theta)$ (or $\sin(3\theta)$) function. In this screenshot, channel 6 (from the top) looks different, as it measures the size of the saw tooth signal, effectively giving the timing of the flash relative to the acoustic field.

**Particle interference setup**

Historically this is the oldest one of the three setups I have built and used. The setup was actually built in my time as a master student in collaboration with Martin Skogstad [50].

This setup is based on optical fibers allowing for positioning different channels at small relative angles. This is optimal for measuring small angle effects like
Hanbury-Brown Twiss correlations and (it turns out) interference patterns from small particles.

Figure 8: Setup with 4 fibers mounted in diamond formation to get 2D information of interference pattern. Fibers lead the light to PMT’s, and the signal is processed by preamplifiers followed by an electronic filter removing 337 kHz noise from the internal power supply in PMT’s. The signal is finally put through a pulse shaping amplifier and put in to a PC equipped with an ADlink 9810 data acquisition card. The data acquisition is clocked by a PMT collecting light from the bubble. The clock signal is generated by a discriminator and finally delayed and doubled into two pulses.

The cell used in this setup is a spherical quartz cell with piezoelectric crystals glued to the sides of it. A microphone is glued to the bottom of the cell. The cell is put inside a refrigerator optimizing light emission as well as functioning as a dark chamber. An Agilent 33120a function generator is connected through an amplifier (ILP HY2005), a transformer and impedance matching circuit to the piezoelectric crystals. The function generator can be remote controlled by computer. The computer is programmed to be able to reseed a bubble and adjust amplitude and frequency automatically when needed.

Four 1 mm Ø optical fibers are directed at the bubble from a distance of 3 to 6 cm (depending on the situation). The optical fibers lead the light out of the refrigerator and into a separate box in which four PMT’s (Hamamatsu 5783P optimized for single photon acquisition) are placed. The PMT’s can be fitted with optical filters.
if desired. The output from the PMT’s is led into preamplifiers (MAX4005) followed by an electronic filter to remove unwanted 337 kHz noise originating from the PMT’s internal high voltage supply. The signals are then further processed by four time shaping amplifiers (Ortec 571/572). A separate clock PMT is situated inside the refrigerator. The clock PMT function is to provide a timing signal, so the peak voltage output from the time shaping amplifiers can be measured. The clock PMT is connected to an amplifier and a constant fraction discriminator. The signal is then delayed in a delay box (Ortec 551). Our 4-channel data acquisition card (ADLink 9810) requires two clock signals to acquire one datapoint, thus our delayed timing signal is put through a pulse doubling box outputting two short peaks into the external clock input of the AD card. 3,000,000 data samples from each channel is acquired and saved to disk.

Further details of this setup can be found in my master thesis [36].

Clean water is prepared from a clean water supply (EASYpure RF from Barnstead) equipped with a 200 nm Millipore filter and the glass cell is carefully cleaned and filled with this clean water filtered through 3 serially connected 200 nm filters. This should remove nearly all particles from the water, and we do see a definite decrease in the occurrence of refraction patterns.

**UCLA setup (particle interference and imaging)**

The setup shown in Figure 9 I built at UCLA to simultaneously measure the distance from a dirt particle to the sonoluminescing bubble and the refraction pattern from the dirt particle. This information can be used to give an estimate of the bubble size as we have published in [4] (see also later chapter).

The principle of the setup is to use red light to illuminate the bubble and give a picture on a video camera through a microscope. The blue light from the bubble is refracted in the dirt particle resulting in a refractive pattern measured by photo multiplier tubes (PMT). By applying optical filters the two regions of wavelengths can be used for these different tasks without affecting each other.
Figure 9: Setup built at UCLA to simultaneously capture video of dirt particles and corresponding refraction pattern. Long wavelengths (600-700 nm) are used for video imaging and short wavelength (below 500 nm) are used to measure refraction pattern.

**Basic setup**
A piezoelectric crystal is attached to a cylindrical glass cell. The cell is filled with water containing a desired amount of various gasses like Xenon and Nitrogen. Most experiments were run with 1% Xenon in Nitrogen gas mixture, to facilitate easy bubble generation as well as brightly emitting bubbles. Typical total air pressure in the water is around 200 mbar. The cell is driven at an overtone around 37 kHz, where the bubble was positioned about 10 mm from the top of the cell,
making high magnification video imaging possible. The top (and bottom) of the
cell is very flat. At the fundamental frequency (29.3 kHz) the bubble is caught at
the center of the cylinder, but too far from the glass top to make a good clear and
highly magnified image.

The sound is generated by a function generator (Stanford Research System
DS345) amplified by a model L2 power amplifier from Instruments Inc. coupled
to an impedance matching circuit. The sound field is monitored by a microphone
with a signal amplified by a low noise differential preamplifier (SR560 from
Stanford Research Systems). Bubbles can be seeded by a boiler wire as needed.

**Bubble imaging**

The microscope used (Zeiss Stemi SV 11) is a stereo microscope where one of the
optical pathways can be led to a video camera. This was nice, since one eyepiece
with low magnification could be used to find the bubble and do rough alignment
of the microscope, while the other pathway goes directly to the video camera.

However, being a stereo microscope (intended for 3D viewing) there is significant
chromatic aberration in the optical pathway making the blue light from the bubble
and the red backlighting appear at different positions in the video image. For
normal applications it is usually not an issue, but for the very different colors I
work with, the position is clearly separated as you will see later. It does not affect
the determination of relative positions of particle and bubble, only the blue/UV
light emitted from the bubble appears at a different position.

Furthermore being a stereo microscope it collects light to the two eyepieces from
two different angles. However, as the light is refracted in the flat water/glass/air
interface, the optical pathway towards the right eyepiece/camera has to be
perpendicular to the surface. To obtain that the microscope has to be tilted by half
the stereo angle (the stereo angle is 35° meaning the microscope must tilt 17.5°).
In practice it is not easy, and it takes a lot of aligning to produce sharp images,
especially at high magnification (which I need). To adjust the position of the
bubble relative to the camera, the cell is mounted on a 3D stage, where the X, Y
and Z position of the bubble/cell can be fine tuned by variable speed electrical
motors. The Z-position is adjusted to reach the focal plane. X- and Y-positions are
adjusted to center the image.

A 3 ns pulsed laser (Coherent Infinity Nd:YAG ) is clocked at the same rate as the
video camera records (30 Hz) to image the position of the bubble and a dirt
particle at one moment. The laser is timed to flash at the precise moment the
bubble emits light. The laser emits 1064 nm infrared light, which is frequency
doubled in a nonlinear crystal producing 532 nm light. The 532 nm light excites a
dye (Nile Blue dissolved in methanol), which emits wavelengths between 600 and
700 nm. The laser beam and dye is shielded from the rest of the setup by black paper, rubber and cardboard, except for a window made from a dyed glass filter allowing only wavelengths over 600 nm to pass. In effect, no laser light can escape the box, allowing only the red light to pass through the dyed glass window to be focused by a lens onto the bubble and particle. The bubble and nearby particles can then be seen as dark shades by the video camera. The light emitted from the bubble is seen as a white spot on the video image. However, due to chromatic aberration in the microscope the white spot is displaced slightly from the shade of the bubble as you will see in later figures. The video signal is recorded on a VCR, and later transferred to a computer for later frame by frame analysis.

As the laser is fired continuously one must limit the power per flash to prevent heating and potentially boiling the dye solution. Also the dye has a limited lifetime especially when subjected to laser-light and must be changed after some usage time. Firing the laser in burst series make higher intensities possible. This turns out to be a good idea anyway in regard to data collection of the refraction pattern. One could choose to only fire the laser in e.g. every other video frame, but that has the cost of less accurate tracking of the particle paths.

The light emitted from the dye is focused by a lens to hit the bubble. The lens is necessary because the bubble is far away from the dye. To further increase the amount of light collected by the camera the aperture in the microscope can be opened further. This comes with a loss of image sharpness thus an optimal position must be found. Adjusting the gain on the Cohu camera also produces better signals at the cost of added noise. The best possible gain setting is found by further examining the captured frames.

In addition to the imaging of bubble and dirt particles we need simultaneous measurements of the refraction patterns produced by refraction in the dirt particles.
Figure 10: Experimental setup acquiring observed intensity traces, while at the same time recording the timing of laser flashes and sound field sync signals. The delay generator inhibiting the laser from firing, triggers data acquisition simultaneously with turning on the laser.

**Refraction pattern**

Two PMT’s are aligned to look directly at the bubble. To reduce light from reflections and outside light, a cone of black paper is mounted as a shield. A piece of black paper with a small hole is mounted on the individual PMT’s. The holes make sure the detector area is smaller than the interference pattern they are supposed to detect. On the other hand there has to be enough area to collect enough photons to be able to distinguish the interference pattern from noise. Therefore an angular size of $1^\circ$ (radius of holes as seen from the bubble/particle) was chosen. The PMT’s are also put behind a dyed glass filter allowing only wavelengths below 500 nm to pass, cutting off potentially escaped laser light, as well as red light from the dye.

The only source of light below 500 nm is the light emitted from the sonoluminescing bubble. When a particle slowly passes in between the PMT’s and the bubble, refraction or shading should be visible in the observed intensity traces.
The PMT’s (Hamamatsu 5783) are supplied with 12 V DC and controlled by a variable resistor to adjust the internal gain. The signal from the PMT’s is amplified by fast preamplifiers (Ortec VT120C) and further amplified by Gaussian amplifiers (CR-200-4µs) from Cremat Inc. The signal is collected by a digitizer (Acqiris CC103 with an Acqiris DC270 board), with a sampling time of 2 µs allowing a total sampling length of 4 s. The acquired data is transferred to a computer and later analyzed.

The data analysis is done to get the intensity as a function of time, with some time averaging. Since the data collected has timing information (from the sync output of the function generator into a channel on the digitizer), we know that the flashes of light come synchronized with the sound field. By doing the data averaging in a clever way, the influence of false light (whether dark signal from the PMT or photons from outside the dark box) can be significantly reduced. A typical sample of the output from the cremat amplifiers is shown in Figure 11. In stead of just calculating the area above and below the zero-line for each individual flash, I average the output synced to the acoustic field. An average over 100 acoustic cycles is shown in Figure 12. This technique removes noise out of phase with the acoustic field much more efficiently.

Figure 11: Typical output from Cremat amplifier (voltage as function of time). The frequency of the acoustic field is marked by the black vertical lines. Substantial more or less random noise is apparent on this signal.

Figure 12: Averaged output of signals above. Running average of 100 acoustic cycles is shown.

Summing over ~500 waveforms makes the data virtually immune to random noise, since random noise will be averaged out to a very high degree. In principle the noise of X “false” photons should be reduced by order $\sqrt{X}$ on top of the usual gain from averaging. In my setup this method reduces the noise to more acceptable...
levels - especially for longer averaging. The averaged (or summed if you like) signal is finally found by taking the absolute value of the individual samples in the averaged waveform and adding those absolute values, or if you like, summing the area above and below the zero-line⁴. Before all this is done the signal can automatically be corrected for a DC offset.

The laser is only fired for four seconds followed by a 16 second break. The sync signal from the sound field function generator is used as a timing trigger of a digital delay generator (Stanford Research Systems DG535). This delay box is used to adjust the timing of laser flashes. The laser is fired by a second digital delay generator (also DG535) producing the required pulses to fire the laser every 33.3 ms in sync with the frame rate of the Cohu LCD camera.

Along with the two channels for intensity measurements, the third channel is fed with the sync output from the sound field function generator. The last and fourth channel is fed with the sync signal that fires the laser pulses. This signal also triggers the start of data acquisition, so when the laser begins firing, data acquisition starts.

The video is captured on a VCR-tape for later analysis. As the laser fires for 4 seconds followed by 16 seconds darkness, it is easy to make a correspondence video frame by video frame to the acquired light intensity data. The data analysis can then later be gone through, looking for interesting peaks and valleys in the measured intensity. Once the videotape is digitized, finding the corresponding video frame to a segment of intensity data is fairly easy.

For digitizing the video I used a TV-card⁵ in my own personal computer. The video was compressed to the highest bitrate possible MPEG2-stream [51] with minimal loss in quality. Furthermore the contrast and brightness of the images was adjusted for improved perceived video quality. For analyzing the video frame-by-frame I use an open source program called VirtualDub (actually a modification of it (VirtualDubMod) that supports MPEG2-streams). In VirtualDubMod fast forwarding and rewinding to a specific video frame number is easy. Further video processing (sharpening/brightness/contrast etc.) can be performed in VirtualDubMod if desired.

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⁴ This is essentially how a lock-in amplifier works, but this way I can vary the averaging time as needed in the data analysis. That would not be possible with the use of a conventional lock-in amplifier.

⁵ ATI All-in-wonder 9000Pro
In my time studying sonoluminescence, first as a master student and later as a graduate student, I have done some measurements, with some very interesting results. My list of published and submitted papers include:


Jeppe Seidelin Dam and Mogens T. Levinsen (paper on period doubling with n=3 symmetry based on data similar to what is presented in this thesis, not finished by deadline of this thesis.) (2006?).
Mogens T. Levinsen and Jeppe Seidelin Dam, “Period doubling in Single Bubble Sonoluminescence”, Proceedings of 9th Experimental Chaos Conference 2006 (to be printed). The two PRL papers, the preprint, and the conference paper are appended to this thesis.

The first three papers were done in my time as a master student and all revolve around shape distorted period doubling.

The next paper [4] published in PRL presents a method to measure the size of the light-emitting region in SBSL producing the first direct measurements of the size of the light-emitting region. Small particles in the water is caught by the bubble and kept at a close distance. When the small particle is positioned directly between the bubble and light detector, there will be some influence on the observed intensity. At first look one might expect that the particle will create some sort of shadow on the detector. This does happen, but under the right conditions the particle does not create a shadow, but rather a sharp peak. A detector smaller than the peak will then observe a dramatic increase in intensity, as the particle passes by. In the paper we present some estimates on the distance between bubble and particle, but the entire conclusion of the paper relies on those estimates being correct. To back up the paper, I made a setup at UCLA, where I could simultaneously measure the position of the particle and the created interference pattern. Those measurements are not published, but I present them in the following chapter.

In the following paper also published in PRL we present a new type of unstable single bubble sonoluminescence, being a kind of mix between multibubble SL and single bubble SL. Stable SBSL with air in water quickly results in argon rectification, meaning the bubbles contents of Nitrogen and Oxygen reacts to form water soluble reaction products. In the paper we show the known bubble recycling with a timescale of typically ~10 s and the type 2 recycling with a 100 times shorter timescale. The difference in timescales can be found in the ratio between Argon and Nitrogen plus Oxygen in air. Since we have a 100 times more air than argon the diffusion timescale to reach an unstable sized bubble is approx a hundred times shorter. We observe that (n=2) period doubling occur for bubbles collapsing at the same relative phase, thus having the same size, no matter if they are argon-rectified ,only partial argon rectified, or only very slightly argon-rectified.

To further investigate the type 2 recycling we tried to produce the type 2 recycling with a gas mixture without any argon. However, much to our surprise we found that stable light emission could be obtained over a surprisingly large range of pressures resulting in the next paper mentioned above. As the bubble grows in size it will collapse harder and burn the incoming gasses faster, so the bubble
diffusion/burn rate reaches a steady state. The conclusion drawn from this stability, is, that even though the diffusion rate into the bubble is larger for larger bubble sizes, the burn rate increases even more with larger bubble sizes. This is something that could be put in to models like the one recently presented by Puente et al [52]. Much to our surprise we found stable light emission from both pure oxygen bubbles, nitrogen bubbles, as well as mixed nitrogen/oxygen bubbles. This may seem disconnected from the size of the light-emitting region, but the burn-rate of nitrogen and oxygen depend on the temperature profile inside the bubble. The light emitted from the bubbles fits reasonably well to blackbody radiation. Nitrogen bubbles fit to a temperature of around 8000 K whereas Oxygen bubbles fit to a much colder temperature around 6600 K. The Oxygen bubbles are so dim that we cannot see the light with our eyes. The light-emission intensity as well as the fitted blackbody temperature is also much lower. In Fig. 5 in the paper we show what we call a prefactor to the blackbody fits. For a light-emitting blackbody this prefactor can be interpreted as the area of the blackbody multiplied with the lifetime of it. So even though the paper is not meant to be about the size of the light-emitting region, it is still somewhat important for the paper. Unfortunately we do not have the equipment to measure the duration of the light-flashes. If we could do it, then the prefactor could be reduced to the average size of the light-emitting region (in a blackbody interpretation). Also the theoretical predicted burn rate of nitrogen and oxygen in the bubble are affected by whether there is a temperature gradient or not, so the stability over a large regime of acoustic pressures might suggest that there is some non-uniformity. However, this conclusion would rely heavily on the selected model for the complete description. Still, models claiming to explain all about sonoluminescence and argon rectification must be able to explain the stability of the nitrogen and oxygen bubbles.

Our next peer-reviewed paper is unfortunately not finished by the deadline of my thesis, but it will present pretty much the same conclusions I will present in the next chapter (on page 36) on n=3 spherical harmonic distorted bubble.

A conference paper on period doubling is also about to be printed (copy attached to this thesis).
Experimental results

This chapter is comprised of supplementary material to published/submitted papers, as well as unpublished results.

**Period doubling (shape)**

Most sonoluminescing bubbles can be driven to exhibit period doubling, when driven close to their extinction level. Water with 250 mbar air or more is likely to produce good stable period doubling. We observe that when a bubble is period doubled the light emitted in a given direction will be different for even and odd numbered flashes, and whether the odd or even flashes are stronger depends on the specific angle from which one observes the bubble. Thus there is no question that the bubble is emitting anisotropic, and furthermore when the relative amplitude of the period doubling is plotted against the measuring angle \( \theta \) along a cut, it fits very well to a \( Ampl(\theta) = A \cdot Sin(2\theta + \theta_0) + Offset \) function. As I will show later, this phenomenon can be explained by refraction in shape distorted bubbles. Other possible explanations I will argue in the following to be inadequate.

![Non-spherical bubble](image)

**Figure 13:** Non-spherical bubble. Bubble shape is distorted by a second order spherical harmonic function, resulting in some oval shape.

The bubble shown in Figure 13 is shape distorted so there is non-isotropic light refraction in the bubble/water interface. When the shape distortion is becomes a
critical size the bubble will break. An example of how the bubble could look just before breaking is shown in Figure 14.

![Bubble Image](image)

**Figure 14:** Bubble as it could look just before a pinch off event. Often when the bubble is in a recycling state, the bubble gets period double for a short time preceding the pinch off. This indicates that the pinch off is connected to shape distortions.

**Period doubling (timing)**

If the applied sound field contains the half harmonic, the bubble will period double in the timing and general intensity of flashes. This is understandable as the local pressure is different for consecutive collapses. In our experimental setup shown on page 14 we can in certain conditions hear a high-pitched sound from the cell. This sound is the half harmonic of the driving frequency, at the same time as the sound appears the timing of the flashes period doubles by up to about 2 µs difference. Also the measured intensity period doubles and the microphone picks up the half harmonic as well. Something like this, is probably what was observed in the earliest paper reporting period doubling and other time of flash peculiarities [53], although their half-harmonic may not have been audible. Whether a period doubling is in the timing or shape can of course be identified by monitoring the time between flashes, or by observing the bubble from multiple angles. In our experiments we have a synched saw tooth signal going into one of our eight channels, which, along with our clock signal being synched to light-emission, gives us the timing information we need.

The effect of half-harmonics could be further investigated by applying a sound field natively containing the half-harmonic. However, I find the effect rather uninteresting and thus the experiment not worth the effort. One should on the other hand still be aware of the possibility of it happening spontaneously.

In a stable sound field (read no sub-harmonics) we do not observe these time-of-flash phenomena. However, in other liquids the parameters ruling the bubble oscillations are different so one can have period doubling in the timing and
amplitude as well as other time-of-flash phenomena without any disharmonies in the applied sound field as shown experimentally by Hopkins et al [54].

**3-symmetry period doubling discovery and its implications**

By chance we discovered what we call $n=3$ shape distorted period doubling. The discovery came by leaving degassed water (with 275 hPa air) in a cell, not sealed, but with a thin ~6 cm plastic tube filled with water acting as a pressure release. Over a couple of months air diffuses through the tube and into the water in the cell. Small molecules (i.e. Argon) will diffuse faster through the tube than the larger nitrogen and oxygen molecules. To make easily repeatable measurements of this 3-symmetry we need to find a gas mixture that readily produces bubbles large enough to become $n=3$ shape distorted. We found that a mixture of 5 hPa argon and 300 hPa air (giving a total argon content of ~8 hPa) was a good mixture in which bubbles grow to a large enough size to observe $n=3$ period doubling.

In Figure 15 I show period doubling with first the usual 180° symmetry, and later a period doubled state with 120° symmetry. The observation that the larger bubble (higher phase) has a 120° symmetry is in good accordance with the shape stability analysis of Augsdörfer [16].

![Graph showing period doubling](image)

**Figure 15:** Top curve shows one of 7 channels monitoring the intensity as a function of time with a 800 flash temporal average of even and odd flashes separately. The lower curve shows the relative phase. A higher relative phase corresponds to a larger bubble. The first vertical line marks a point where a fit to a Sin(2\(\theta\)) function is made, and the second line marks the point where a fit to a Sin(3\(\theta\)) is done. The percentages indicate the amplitude of the period doubling and the standard
deviation (calculated from the individual 800 samples making up the average intensities and the statistical uncertainty of the averages). Because of the not constant intensity at the first mark, the uncertainty of the average is overestimated.

The period doublings are fitted to these functions:

$$\sin(n\theta + \theta_0) + k$$

(1.1)

Where n is 2 or 3 (depending on the shape deformation being from a n=2 or n=3 spherical harmonic). \(\theta_0\) indicates the spatial orientation of the fit, and k is an offset as explained in [3]. The fits produce Figure 16.

Figure 16: Relative period doubling as function of angle for the two marked cases in the previous figure. The upper curve shows a very good fit to a \(\sin(2\theta)\) function, whereas the latter shows a mediocre fit to a \(\sin(3\theta)\). The numbers written near the top of the curves shows how many standard deviations the points deviate from the fitted curve.
The reduced Chi-square test gives a value of 0.13 for the n=2 fit, and 5.8 for the 
n=3 fit. If one tries to make a n=2 fit for the lower points, one gets a Chi-square of 
40 indicating that although the n=3 fit is not perfect, it is much better than n=2. 
The failure of the n=2 fit can be seen from the data points at 0° and 180°, which 
are very dissimilar as opposed to the upper curve.

Now I give an explanation of how the n=3 symmetry comes about, and most 
importantly what implications it has on the light-emitting region.

A spherical bubble with a spherical blackbody inside will of course emit light with 
a spherical symmetry. A spherical bubble with an ellipsoidal blackbody will emit 
light in good accordance with the usual 180° (n=2) symmetry. However, a 
spherical bubble with a triangular blackbody cannot account for the 120° 
symmetry (it would rather have 60° symmetry, as the amount of light emitted from 
the blackbody is proportional to the size of the silhouette). This can be explained 
by refraction in a bubble shaped like the one shown in Figure 17.

![Bubble shape explaining the observed 120° symmetry. Bubble is overly distorted for clarity.](image)

Linear combinations of n=3 shape distortions produce a variety of bubble shapes. 
The basic shapes for n=3 shape distortions are shown in the following figure.
Figure 18: 3D renderings of n=3 shape distortions. m=0, m=1, m=2 are shown. m=3 results in the same shape as m=1, only with a different orientation in space.

Whereas an n=2 distorted bubble will give a near perfect $\sin(2\theta)$ fit, the n=3 bubbles cannot be expected to do the same, due to the fundamental different shapes with less symmetry.

We know (as mentioned under bubble dynamics) that theoretically shape distorted bubbles are predicted. The question is whether the shape deformation is most prominent in the bubble wall, or whether it is a very slight shape deformation in the bubble wall that produces a more distorted center. Logically, if the light-emitting region has a long mean free path for photons, only refraction in the bubble wall can account for the observed shape distortion. However, if the light-emitting region is of blackbody nature, the case is more complex. Here the n=3 shape distortion provides a conclusion, which cannot be drawn from the regular n=2 shape distortions. The different possibilities are sketched in the following figure.
Figure 19: Both the situation sketched in A and B can explain 180° symmetry in the light emission. However, only case C can explain the 120° symmetry.

The argument shown in Figure 19 tells us that the non-spherical light-emission is rooted in non-spherical bubbles, as opposed to slightly distorted bubbles with much distorted light-emitting regions. Figure 19 D would emit light equally much in two opposite directions, since it is the silhouette of a blackbody that determines the intensity observed in a given direction.

But what would the situation be like, if the light is originating from a blackbody filling (almost) the entire bubble?

Figure 20: The situation sketched in E can explain 180° symmetry. However, the situation sketched in F, cannot explain 120° symmetry, (as it would result in 60° symmetry). For example the silhouette of F is equal seen from two opposite directions, thus not obliging with a basic requirement of 120° symmetry.
Figure 20 presents an argument for the light not being emitted from a blackbody filling almost the entire bubble. The only element that could “save” such a situation is if the refractive index of water is affected with the same symmetry in a region large enough to refract the light emitted.

By these arguments, from the 3-symmetry period doubling being observable in the light emission, a light-emitting blackbody must be significantly smaller than the bubble radius, although a large volume light-emitting region cannot be ruled out.

**Mechanism responsible for memory in period doubling**

Driving the bubble with higher harmonics [55,56] has been shown as a method to increase stability and light output. Also higher harmonics have been shown to be produced by the bubble itself [57]. Maybe the higher harmonics are responsible for the shape distorted period doublings by creating sufficient non-uniformity in the sound field. Also the returning shockwave emitted from the previous flash returning shortly before a bubble collapse [58] could be the memory of orientation. However, the period doubling works equally well in a spherical cell and a cylinder, which would not be expected if the returning shockwave was important.

Another candidate for this memory is in the flow around the bubble [59]. My idea is that the distorted shape of the collapsed bubble causes a small perturbation in the flow near the bubble. This flow is then thought to seed the direction of the inherent shape instability.

A third possibility is the shape distortion lives on throughout the bubbles oscillation, as I have described in the section “Bubble dynamics” on page 7.

**Particle interference measurements**

I have done more experiments on the particle interference setup described on page 22 and in our paper [4]. In this section I will present some of the more interesting interference patterns obtained. In this setup the 4 channels are set up in the following configuration, the angles indicate the distance between channels as observed from the bubble:
Figure 21: Relative position of fibers in measurements described here. As a help for the reader I mark the position of the individual channels in the following figures.

Figure 22: Beautiful interference pattern. Particle has very good trajectory with peak going through channel 0 and 1. The relative intensity in channel 0 and 1 reaches up to about 3, and the valleys go down to about 1/3. The peak in channel 1 is simultaneous with the valley in channel 0. This proves that the angular size of the interference pattern (distance from peak to valley) is 5°.
Figure 23: Here is a particle casting a shade on all channels. The shade is about 5° in diameter. Larger shades have been observed. The shade first passes through channel 0 and last through channel 1. This means the particle is moving upwards, according to the sketch of the channels positions.
Figure 24: In this measurement the fibers are very far apart (spread out over 180°), so the spatial shape of the interference peak in channel 3 is unknown. But clearly the amplitude of the peak is spectacular. The intensity increases by a factor of \( \sim 6 \). The fibers in this case are further away from the bubble (twice the distance), so the spatial averaging due to the fibers size is only half when compared to the previous curves. This turns out to be important when estimating the size of the light-emitting region. Intensity data is plotted with a running average of 5 flashes.

**Particle position measurements (performed at UCLA)**

In the paper on refraction of light in small particles, we presented measurements of what we argued was interference from nearby particles. At UCLA I made some measurements proving that interpretation to be correct. I will present some of the evidence in this section. The measurement setup is described in a previous chapter, but to recapitulate I make simultaneous measurements of the intensity in small-angle detectors while taking high resolution pictures of the bubble and any particles near to it.

In Figure 25 I present pictures of two video frames collected by the setup described in a previous chapter. The frames show a bubble emitting light, with two particles in the vicinity. In Figure 26 I present simultaneous measurements of the
intensity of light measured in PMT’s in the direction indicated by the lines in Figure 25. Figure 27 shows the entire 4 s of acquired data. The effect of the dirt particle can clearly be seen as an intensity peak.

Figure 25: Captured frames of light-emitting bubble with two particles nearby. Frames are 33 ms apart. Frame 71 and 70 are shown here (see next figure). Arrow is pointing at bubble. Particle in front of PMT is marked by square. The direction to PMT is indicated with a line (between bubble and particle). Note that chromatic aberration in the stereo microscope causes the (blue/UV) light from the bubble to appear to the right of the bubble. In frame 70 (the one to the right) the shockwave produced by the bubble collapse can be seen (indicated by arrow). The circles mark artifacts in the video picture (or rather dust particles on the CCD camera). The artifacts are seen stationary on all frames, and can be disregarded.

<table>
<thead>
<tr>
<th>Time (frames)</th>
<th>Intensity (arb. units)</th>
</tr>
</thead>
<tbody>
<tr>
<td>70</td>
<td>1</td>
</tr>
<tr>
<td>72</td>
<td>1</td>
</tr>
<tr>
<td>74</td>
<td>72</td>
</tr>
<tr>
<td>76</td>
<td>1</td>
</tr>
</tbody>
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Figure 26: Interference pattern (intensity as a function of time) as measured by the two PMT's. Note the position of the interference pattern between the markers for frame 70 and 72. From the interference pattern it is clear, that the particle passes in front of the upper PMT almost simultaneously with frame 71. Running average is done over 500 acoustic cycles (corresponding to 0.4 frames or 13 ms).
Figure 27: The complete four second data set. Running average is done over 500 acoustic cycles. The interference peak near frame 71 is clearly above the noise level. Note the intensity increases by almost a factor of 3 in the top channel.

Figure 28 shows pictures of a large particle (the higher contrast compared to other pictures suggests the particle blocks more light, i.e. larger) in front of the PMT’s. This large particle has more of a shading effect than constructive interference, as can be seen in the intensity trace in Figure 29.

Figure 28: Frame 22 and 24. Large dirt particle is casting a shade on the PMT’s. The bubble is positioned just left of the bright spot (as previously explained). Two particles are caught close to the bubble, the upper left one creates the shade measured in the figure below.

Figure 29: Intensity trace of entire file with frame 22 and 24 shown in previous figure. Running average is done over 500 acoustic cycles.
The effect of particles passing by is illustrated in Figure 30 and Figure 31, where a particle is shown in front of the PMT’s and a little later as well. The interference pattern is synchronous with the position of the particle – just as expected. The interference pattern coincides with the timing of a laser flash, which is why I have chosen to show this specific example. Figure 31 shows the interference pattern, where a decreased intensity on the sides of the peaks is also seen. The depth of these valleys can under some assumptions predict a minimum of the size of the light-emitting region.

Figure 30: Frame 29 and 32 from the data file shown below. Direction towards PMT’s is indicated with line. It is clearly seen that the particle has passed by a couple of frames later. Note that the lines used in this figure is exactly the same (direction and length) as the lines used in previous figures, proving that the interference patterns and shading effects are undoubtedly correlated with the position of a particle.
Figure 31: Run2 file 98. Interference pattern near frame 29 and nothing in frame 32, exactly as expected from the frames in previous figure. The slightly delayed peak in the lower channel corresponds well with the particles path indicated in Figure 30. Running average is done over 500 acoustic cycles.

Using only two PMT’s to obtain the intensity-trace does not give enough spatial knowledge of the angular size of the interference pattern to accurately determine the size. This problem originates in the data acquisition only having four channels available, where I need to use two of them for timing information. However, in the experiments done in Copenhagen the size of the interference pattern (as measured in angle from peak to valley) was always about 5°. The experimental data shown here do agree with the size being 5°. The ideal way to measure the interference pattern would be to have many channels close together, providing a time-resolved 2D picture.
Refraction Calculations

Refraction in shape-distorted bubbles was first proposed by Weninger et al [17] to explain observations. Madrazo et al [62] did numerical calculations on the subject. However, they did the calculations with only a single, centered light-emitting point. I will examine numerically if the size of the light-emitting region plays a role in the resulting refraction patterns. Also refraction in nearby particles gives clues to the size of the light-emitting region. I will elaborate on the calculations done in our paper [4].

At light-emission the bubble is of a size similar to the wavelength of light. Because of this one cannot expect all wavelengths to be refracted equally. Therefore a model for the light-refraction must be used. The resulting spatial distribution of light may also depend on the size (and nature) of the light-emitting region. Light from a blackbody filling almost the entire bubble is expected to give refraction different to that of a more point-like light-emitting region as well as a volume light-emitting bubble. Calculations on this matter, along with actual measurements may provide valuable insight regarding the size and nature of the light-emitting region. Stable shape distorted period doubled bubbles are the perfect candidate for such measurements.

I do not have a working model that takes a blackbody into account. My model assumes undamped spherical waves emitted in all directions from a light-emitting point. The light-emitting points can, however, be freely distributed inside the bubble. Thus, I cannot make final conclusion of the nature of the light-emitting region, but others with more time and advanced numerical models may be able to develop this idea to give this most valuable insight. With this in mind, I present what I do have developed (volume emitting bubbles).

**Refraction in shape distorted bubbles**

In this thesis I argue, that only a shape distorted bubble can produce the observed anisotropic light-emission. Calculations on the refraction in shape distorted bubbles were first done by Madrazo et al [62]. Their model is based on a point light-source in differently shaped bubbles including ellipsoids and homogenous refraction indices in water and bubble respectively. The model I developed also
assumes homogenous refractive indices, but allows for larger light-emitting regions in the bubble. My model is based on ellipsoidal bubble shapes with rotational symmetry. My model does not take reflections into account, nor differences between polarizations. Ideally one would start out from Maxwells equations and integrate those. However, my model is more computationally simple and does produce quite similar results when compared to Madrazo’s model. The basics of my model is also presented in my master thesis, but is improved in this thesis to allow for larger light-emitting regions than just the central point as well as integration over a distribution of wavelength. I start with Kirchoff’s diffraction formula as found in [60].

\[
\psi = \frac{1}{4\pi} \int \left\{ \left( \frac{e^{-ik_1 r}}{r} \cos (\overrightarrow{Ne}, \overrightarrow{r}) \frac{\partial}{\partial r_1} \left[ \frac{A}{r_1} e^{-ik_1 r_1} \right] \right) + \left( \frac{A}{r_1} e^{-ik_1 r_1} \cos (\overrightarrow{Ne}, r) \frac{\partial}{\partial r} \left[ \frac{1}{r} e^{-ik_1 r} \right] \right) \right\} dS \quad (1.1)
\]

\( r \) is the distance from integration point to the observer \((\gg \lambda)\). \( r_1 \) is the distance from light source to integration point \((\sim \lambda)\). \( A \) is the amplitude. If the coordinates for the source point is \((Ox, Oy, Oz)\) then the distance \( r_1 \) to a point on the surface \((Rx, Ry, Rz)\) can be expressed as:

\[
|r_1| = \sqrt{(Ox - Rx)^2 + (Oy - Ry)^2 + (Oz - Rz)^2} \quad (1.2)
\]

\( k_1 \left( = \frac{2\pi \cdot n_1}{\lambda} \right) \) is the wave number inside the bubble, and \( k_2 \) is the wave number in water. \( \cos (\overrightarrow{Ne}, \overrightarrow{r}) \) is cosine to the angle between the vector normal to the integration surface and the vector pointing towards the observer. \( \vec{e}_{oobs} \) is a unit vector point towards the observer.

---

\(^6\) According to the theory for shape deformations one should not use ellipsoids, but rather n=2 spherical harmonic shape deformed bubbles. For small shape deformations, however, the difference is not really noticeable. Ellipsoidal bubbles are more computationally simple, which is the reason I (and others before me) choose to use them.
Figure 32: Integration surface (and bubble surface) with indication of vectors etc. The ellipsoid has rotational symmetry around the x-axis.

Replacing the cosine functions with dot products of unit vectors, and performing the differentiation gives the following function (see Figure 32 for explanation of names).

\[
\psi = \frac{a}{4\pi r} \left[ \frac{1}{r_1} e^{-ik_1 - k_2 (\overline{N_e \cdot eobs})} \right] \left[ ik_z (\overline{N_e \cdot eobs}) + (\overline{N_e \cdot r_i}) \left( \frac{1}{r_1} + ik_1 \right) \right] dS \quad (1.3)
\]

For an ellipsoid \( dS \) can be written as:

\[
dS = \frac{r_z^2 \sin(\theta)}{\overline{N_e \cdot R}} d\theta d\phi \quad (1.4)
\]

\( \theta_n \) (see Figure 32) can be calculated:

\[
\theta_n = \text{ArcTan} \left( \frac{a^2}{b^2} \tan(\theta) \right) \quad (1.5)
\]

From \( \theta_n \) one easily calculates \( \overline{N_e} \):
\[
\bar{N}_e = \begin{pmatrix}
\cos(\theta_n) \\
\sin(\theta_n)\cos(\varphi) \\
\sin(\theta_n)\sin(\varphi)
\end{pmatrix}
\] (1.6)

The direction to the observer (by definition in the x,y plane) is \( \text{eobs} \):

\[
\text{eobs} = \begin{pmatrix}
\cos(\theta_{obs}) \\
\sin(\theta_{obs}) \\
0
\end{pmatrix}
\] (1.7)

With equation (1.1) to (1.7) the light emission from one point within the ellipsoid in one direction can be found. To approximate real physical situations, one must add intensities from many different source points and wavelengths. This I do by using a quasi Monte Carlo approach [61], “quasi” meaning that I do not choose points and wavelengths randomly, but in a systematic manner. The calculation continues until the result has converged. For a complicated calculation, this can take up to a couple of days in Mathematica, since the integration has to be done for thousands of points evenly distributed inside the light-emitting region.

Figure 33 shows a calculation done with my program, but with the same parameters as in the paper by Madrazo. The oscillations present there do not correspond with our measurements. These oscillations are averaged out both when increasing the size of the light-emitting region, and when summing over a distribution of wavelengths.
Figure 33: Calculation of refraction with same parameters as Fig. 1 in [62] (250 nm in 0.75 µm / 1 µm ellipsoidal bubble). Qualitatively the results are reasonably similar.

So let us investigate what happens when integrating over a wavelength distribution.

Figure 34: Light-emitting point as in previous figure, but now the wavelength is a Gaussian distribution around 250 nm with 30 nm std. dev. This reduces the oscillations quite much, as you can see.
Now, let us see what happens when increasing the size of the light-emitting region:

![Graph showing intensity variation with angle]

**Figure 35:** Madrazos parameters, except for a 0.2 μm light-emitting region and Gaussian distribution of wavelength around 250 nm with 30 nm std. dev.

A further increase of the size of the light-emitting region smoothes the calculated intensity profile even more, as can be seen in the next figure.
From the previous figures it is clear that both increasing the size of the light-emitting region, and summing over a region of wavelengths results in smoothing of the intensity function. This explains why we do not experimentally observe the oscillations apparent in Madrazo et al [62].

This effect could be utilized as a way to determine the size of the light-emitting region. If distorted period doubled bubbles are observed with narrow band short wavelength optical filters from multiple angles, one can check if there are any of these oscillations present, and from the amplitude of the apparent oscillations one could make numerical calculations fitting the observations, and thus get a better grasp on the size of the light-emitting region.

I have not done such a measurement, because of the inherent difficulties connected with it. To do the measurement in our lab, we would need 8 UV-optimized PMT’s. Furthermore we would need 8 short wavelength optical filters. In addition to that we would need a very stable period doubled bubble, and a method to determine the stability of it (using ~8 ordinary PMT’s without filters and observing a stable light-emission would be a good idea.) The alternative to a stable period doubled bubble is a tumbling period doubled bubble, where the spatial orientation of the period doubling tumbles around. One large dataset of this kind could be investigated with a correlation analysis (similar to a Hanbury-Brown Twiss experiment), where the oscillations would show up as small angle correlations.
**Period doubling and the size of the light-emitting region**

In this section I will investigate whether different sizes of the light-emitting region have measurable effects with parameters typical of our experiments. This I do to check if the experimental data we have can only be explained by some sizes of the light-emitting region.

![Diagram](image)

**Figure 37:** A volume-emitting region inside an ellipsoidal bubble. An inner sphere is filled with individual light-emitting points.

The model sketched in Figure 37 is used to calculate the resulting light-refraction in the following figures, where only the size of the light-emitting region is varied. The calculations are done in an ellipsoid with one axis 1.1 µm and the 2 other axis’s are 1 µm. The refractive index inside the bubble is chosen to be 1.125 and 1.33 outside. The calculation is done with a Gaussian distribution of wavelengths around 350 nm with a std. dev. of 30 nm.

In the following figures I plot the intensity as a function of angle for various sizes of the light-emitting region.
Figure 38: $R = 0.1 \, \mu m$.

Figure 39: $R = 0.3 \, \mu m$.

Figure 40: $R = 0.4 \, \mu m$. 
In Figure 38, Figure 39, Figure 40 and Figure 41 it is seen that the distribution of the intensity is in all cases roughly similar to a sine distribution, and the curves are too similar for one to experimentally tell them apart, thus the observation of period doubling nicely fitting a $\sin(2\theta)$ function does on its own not provide any insight to the size of the light-emitting region. We have not measured any large deviations from a sine distribution of light in our period doubling measurements. The wavelength dependence (product of PMT wavelength sensitivity and SL spectrum) of our measurement setup can be found in our paper [2]. The actual wavelength dependence is not very dissimilar from the distribution chosen in the calculations shown above.

Refraction by particles

The model presented in our paper [4] is basically derived from the model for refraction in shape distorted bubbles. The major difference to the bubble wall refraction is that the integration surface is chosen as a sphere with the particles distance as the radius (rather than the bubble-water interface). The phase of the light waves (originating from a single point in the calculation) is then shifted according to the distance traveled inside the particle and the refractive index difference. Note that the only modification the particle does (in my model) to the integration, is to shift the phase by the delay it causes by its different refractive index. The refractive index of common dust is 1.5 [63] compared to waters refractive index of 1.33. The difference in refractive index is 0.17 and the delay of the phase can be now be deduced:

$$\theta_{\text{delay}} = \frac{2\pi(1.5-1.33)d}{\lambda}$$ (1.8)
Where $\theta_{\text{delay}}$ is the phase delay, $d$ is the distance traveled through the particle, and $\lambda$ is the wavelength in vacuum.

Figure 42: Sketch of integration surface used to calculate the field far away from the bubble and particle.

This model gives a very large integration sphere with very long calculation times. However the calculation can be simplified greatly, by remembering that the total integration of the total sphere with no particle present must give a regular $\frac{1}{r}$ dependence ($\frac{1}{r^2}$ in the intensity), and the relative phase (as function of distance) is known as well. This can be used to simplify the integration. A prefactor and resulting phase can be put outside the integral, and then the integral over the integration area (sphere in this case) can be chosen to be 1.

The total spherical area I call $S$. The area covered by the particle I call $P$. The area not covered by the particle is called $A$. The function to be integrated for no particles present I call $f$. Since we know that the integral with no particle present is 1, we can use this information to only integrate over the area $P$ as I show in the following.

$$ S = P + A \quad (1.9) $$

$$ \int_S f dS = \int_{P+A} f dS = \int_P f dS + \int_A f dS = 1 \quad (1.10) $$
\[ \int_{A} f dS = 1 - \int_{P} f dS \] (1.11)

The integral with the particle present, I call the function to be integrated \( g \). Outside the area covered by the particle the two functions are equal.

\[ \int_{A} f dS = \int_{A} g dS \] (1.12)

Using the above relations we can now decrease the area over which we need to integrate by some orders of magnitude. Rather than integrating over the entire sphere (S), we only need to integrate over the much smaller area covered by the particle (P).

\[ \int_{S} g dS = \int_{A} g dS + \int_{P} g dS = \int_{A} f dS + \int_{P} g dS = 1 - \int_{P} f dS + \int_{P} g dS = 1 - \int_{P} (g - f) dS \] (1.13)

When calculating an interference pattern, one must remember that the bubble emits a broad spectrum of wavelengths, and the PMT also has a sensitivity profile. The product of the spectral profile of the light and PMT sensitivity gives the curve we show in Fig. 2 in [2]. Thus a calculated interference pattern should be a weighted sum of multiple wavelengths to resemble the measured interference patterns. Using equations (1.3) through (1.13) I have made a program in Mathematica to calculate the appearance of the interference patterns. One numerical calculation of the interference pattern is shown in the following figures:
Figure 43: Interference pattern from a 2.2 µm (radius) particle positioned 20 µm from the bubble. The interference pattern is of course rotationally symmetric. Wavelengths are chosen in a Gaussian distribution around 350 nm with 30 nm std. dev.
Figure 44: 3D plot of previous figure, just to illustrate the rotational symmetry. Relative intensity is plotted as a function of angle.

However, if only calculating for one wavelength, many fringes are observed. The next figure is for the same parameters, except for the wavelength only being 300 nm.
Figure 45: Same parameters as Figure 43, except for the singular wavelength (300 nm). Clearly there are many more oscillations in the intensity.

Figure 46: Same as previous figure except for the wavelength which is now 350 nm. Notice the position of the minimum intensity is shifting. Also the peak intensity is different.

**Effect of particles in a Hanbury-Brown Twiss experiment**

The much more pronounced peaking seen in Figure 45 and Figure 46 when compared to Figure 43 has important consequences for the Hanbury-Brown Twiss experiment. The peaking for a singular wavelength (as is the case for Hanbury-
Brown Twiss measurements) will result high, small angle correlations, obscuring any real effect in the HBT measurement. For HBT measurements one would use approx 1 nm wide filters. To investigate if this width dampens the oscillations I make a numerical calculation for a Gaussian distribution around 350 nm with a 0.5 nm std. dev. This results in the following figure:

![Relative intensity vs Angle in degrees graph](image)

**Figure 47:** The peaking is just as pronounced compared to Figure 46. A 1 nm filter helps nothing in averaging out peaks.

Calculating the effect of the particle on the measured correlation is not trivial [1,34]. A worst case estimate can be found assuming random position of the particle for each flash. For a 350 nm correlation measurement this results in a correlation effect plotted in Figure 48.
The correlations expected to be measured in a HBT correlation measurement are of the order 0.001. This means, that if a particle is near the bubble for more than a few minutes for every hour of measurements, the experiment could be ruined - or worse, resulting in one believing to have measured HBT correlations, rather than just interference from particles.

**Averaging induced by the size of the light-emitting region**

The spatial averaging of the intensity arises from both the angular size of the detector and from the angular size of the light-emitting region (as seen from the particle.) Remember that all the previous calculations concern a point light-source and a point light-detector.

**Figure 49: Sketch indicating the angular size of the light-emitting region and detector.**
Figure 50: Sketch illustrating how a large light source results in overlapping interference patterns, or a spatial smearing in the intensity in other words. The size of the light-detector (PMT) also adds significant smearing.

To take the size of light-emitting region and detector into account a convolution must be done. The peak intensity is thus lowered, and the intensity does no longer go as far down in the valleys. The larger the light-emitting region, the more smearing will occur. For functions to smear I will use my experimental determined parameters. The distance of dirt particles has been measured in the UCLA setup to be about 20 µm. The size of the particles is unknown, but only a limited regime of sizes produce constructive interference patterns (heightened intensity peaks). Those sizes, do, by the way, match the sizes of controlled particle sizes we added to the water in the experiments presented in [4]. The particles added in the experiment performed in Copenhagen had a size distribution with a mean radius of 2.5 µm with 0.5 µm std. dev.

From the following 6 figures it can be seen that our requirements of significantly increased peak intensity (at least a factor of two) combined with the minimal intensity being for angles close to 5° from the peak cannot be fulfilled for all the simulations. Interference patterns are plotted for particles with radii ranging from 2.0 to 3.5 µm.
Figure 51: Original and 1, 2, and 3° (2.0 μm particle 20 μm from bubble). The black curve is the point source, point detector calculated intensity distribution. The blue, green, and red curves are all with 1° detector and 1°, 2°, and 3° size of light emitting region respectively.

Figure 52: Same as previous figure, but now with a 2.3 μm particle.
Figure 53: Same as previous figure, but now with a 2.6 µm particle.

Figure 54: Same as previous figure, but now with a 2.9 µm particle.

Figure 55: Same as previous figure, but now with a 3.2 µm particle.
The 2.0 µm particle cannot produce the required peak intensities. The 2.9, 3.2, and 3.5 µm particles produce high peaks, but their minima are too far from their peaks to fit our experimental data. The distance from peak to valley should only be 5°, where it is closer to 7° or 8° for those particle sizes.

Remaining are the 2.3 µm and 2.6 µm particle sizes, where the 3° light-emitting region do not produce high enough peaks. This leaves us to the conclusion that the patterns observed come from light-emitting regions no more than 2° in size. This corresponds to a light-emitting region with a radius:

\[ R_{\text{light-emitting}} \leq \sin(2°) \cdot 20 \mu m = 0.7 \mu m \]  

The fits with a light-emitting region of only 1° in size (corresponding to a light-emitting region of ~0.35 µm), do seem to be a better fit to the experiment, but up to 2° radius cannot be ruled out with the evidence at hand.

To give a better view of the variation in parameters I have compiled the following table containing peak intensities, depth of valleys (minimal intensity) and the angle of minimum intensity. I have color coded the table, red meaning parameters not agreeing with experimental data, and green meaning acceptable values for all parameters.
<table>
<thead>
<tr>
<th>Particle radius</th>
<th>Detector size</th>
<th>Peak Intensity</th>
<th>Minimal Int.</th>
<th>Angle of Min. Int.</th>
</tr>
</thead>
<tbody>
<tr>
<td>2.0 µm</td>
<td>1°</td>
<td>2.0</td>
<td>0.30</td>
<td>4.1°</td>
</tr>
<tr>
<td>2.0 µm</td>
<td>2°</td>
<td>1.5</td>
<td>0.40</td>
<td>4.3°</td>
</tr>
<tr>
<td>2.0 µm</td>
<td>3°</td>
<td>1.1</td>
<td>0.49</td>
<td>4.8°</td>
</tr>
<tr>
<td>2.3 µm</td>
<td>1°</td>
<td>2.8</td>
<td>0.45</td>
<td>6.6°</td>
</tr>
<tr>
<td>2.3 µm</td>
<td>2°</td>
<td>2.0</td>
<td>0.47</td>
<td>5.5°</td>
</tr>
<tr>
<td>2.3 µm</td>
<td>3°</td>
<td>1.3</td>
<td>0.49</td>
<td>5.3°</td>
</tr>
<tr>
<td>2.6 µm</td>
<td>1°</td>
<td>3.3</td>
<td>0.39</td>
<td>6.2°</td>
</tr>
<tr>
<td>2.6 µm</td>
<td>2°</td>
<td>2.3</td>
<td>0.44</td>
<td>6.4°</td>
</tr>
<tr>
<td>2.6 µm</td>
<td>3°</td>
<td>1.6</td>
<td>0.50</td>
<td>6.5°</td>
</tr>
<tr>
<td>2.9 µm</td>
<td>1°</td>
<td>3.6</td>
<td>0.40</td>
<td>7.4°</td>
</tr>
<tr>
<td>2.9 µm</td>
<td>2°</td>
<td>2.7</td>
<td>0.41</td>
<td>6.9°</td>
</tr>
<tr>
<td>2.9 µm</td>
<td>3°</td>
<td>2.1</td>
<td>0.46</td>
<td>7.0°</td>
</tr>
<tr>
<td>3.2 µm</td>
<td>1°</td>
<td>4.3</td>
<td>0.36</td>
<td>7.5°</td>
</tr>
<tr>
<td>3.2 µm</td>
<td>2°</td>
<td>3.5</td>
<td>0.39</td>
<td>7.7°</td>
</tr>
<tr>
<td>3.2 µm</td>
<td>3°</td>
<td>2.9</td>
<td>0.43</td>
<td>7.6°</td>
</tr>
</tbody>
</table>

The radii of the particles producing the interference patterns we observe must be between 2.0 and 2.6 µm. This experiment deserves to be done with particles with a specific size in this regime and (just as important) a refractive index close to 1.50.

The larger particles can also be responsible for some interference patterns. We have observed peaks with a relative intensity of up to 6. However, those patterns were obtained with the fibers positioned at twice the usual distance from the bubble, meaning that the opening angle in the fiber was only 0.5°. This is very important when estimating the size of the light-emitting region, because such a dramatic change in observed intensity will only be possible, if the light-emitting region has a similar angular size. From this argument the size of the light-emitting region should be no more than the same size, meaning a maximum of the light-emitting region of 0.5° to 1° corresponding to 0.17 µm to 0.35 µm in radius.\footnote{Unfortunately I only became aware of the relation between distance to detector and the magnitude of the relative intensity shortly before my deadline, so I had no time for further investigations.}

**Effect of refraction in bubble surface**

An issue not mentioned in the calculations here, is that the refraction of light in the bubble wall may make the light-emitting region appear to be of a different size, than it is in reality. I have sketched the situation in the following figure.
Figure 57: Drawing of the effect of refraction on the apparent size of the light-emitting region.

Figure 58: A blow-up of previous figure - intended to illustrate the following calculation determining the relation between apparent and real size of light-emitting region (in a simple classical case).

In Figure 58 the sketch from Figure 57 is blown up, and angles of refraction are drawn to help figure out the relation between apparent and real size of the light-emitting region in the classical case.
From Snell’s law we have:

\[
\frac{\sin(\theta_1)}{\sin(\theta_2)} = \frac{n_2}{n_1}
\]  

(1.15)

From the geometry sketched in Figure 58 we find the following relations:

\[
\sin(\theta_1) = \frac{R_{\text{app}}}{R_{\text{bubble}}}
\]  

(1.16)

\[
\sin(\theta_2) = \frac{R_{\text{real}}}{R_{\text{bubble}}}
\]  

(1.17)

From (1.15), (1.16) and (1.17) one easily deduces a surprisingly simple relation between apparent and real size of the light emitting region:

\[
R_{\text{real}} = R_{\text{app}} \frac{n_1}{n_2}
\]  

(1.18)

Thus, in the classical calculation, one has to modify the found apparent size by multiplying with the relative refractive index. In the not classical case, there may be a similar effect.

To conclude, this method is not far off in determining the size of the light-emitting region – with some additional work on the model calculations, there can be set a lower limit on the size of the light-emitting region from the depth of valleys argument presented above.

Furthermore, more work should be done with detectors of varying sizes further investigating the effect as discussed on page 70.
Conclusion

Summary and Discussion
In my time as a PhD student I have built the setups described in this thesis, and I have done experiments to uncover some of the unknowns of sonoluminescence. Measurements of interference patterns from particles captured near a bubble have turned out to be quite successful phenomena. By measuring the angular size of the interference patterns, and the distance from bubble to dirt particle, I found out that this observation is a way to estimate the size of the light-emitting region. The numerical calculations of the interference patterns, as well as the observation, that using detectors with a smaller angular size results in increased maximum intensity of the interference patterns – all points to the light-emitting region being significantly smaller than the minimum collapse radius of the bubble. Thus, models assuming equal pressure and temperature throughout the bubble should be amended.

We have also investigated the importance of the specific mixture of gases inside the bubble. First we observed that an air bubble at high gas contents and high acoustic pressures can produce a fast, low intensity type of recycling, where the bubble does not completely burn off the diatomic gasses inside. The colder spectrum suggests lower temperatures inside the bubble. We observe period doubling for the same size bubbles at three different intensity levels, but all at the same acoustic pressure all within a few seconds. A bubble close to being a type 2 recycling, will perform ordinary recycling, but become close to the maximum stable size and while doing so exhibit period doubling, when partially filled with nitrogen and oxygen. The bubble also exhibits period doubling while being an almost pure Argon bubble, just before the recycling event, and finally we observe period doubling in the type 2 situation when the bubble is largest. If we apply too much averaging in the type 2 situation, it will appear to be stable. In fact, this raises questions to an otherwise very interesting paper by Krefting et al [64]. They claim to have stable sonoluminescence in air-saturated water. They show the timing of the flashes most of the time is fairly stable. However, they determine the timing of the collapse by averaging the collapse time of 50 collapses. In air-saturated water we can have type 2 recycling on a time-scale shorter than this. We have experimental evidence of type 2 recycling on a time-scale of ~10 flashes in
almost air-saturated water. Because of the timescale of the recycling is shorter than their averaging time, they cannot see the (extremely) unstable nature of the bubbles. In their honor they do show timing curves which show some instability (Fig. 4 in their paper), and they do comment on it in the text. Their failure to correctly interpret the process must be blamed on their experimental setup lacking flash by flash resolution⁸.

Inspired by the light-emission from the type 2 bubble consisting of air, we wanted to investigate if we could observe type 2 emission from bubbles without any noble gases. That we could, but to our surprise we saw that we could have stable light-emission as well. We observed stable light emission from both oxygen bubbles and nitrogen bubbles. The light from oxygen bubbles fits a colder blackbody spectrum than the nitrogen bubbles. The dissociation temperature for oxygen is lower than for nitrogen, and our measured blackbody temperatures may be connected to actual temperatures inside the bubble. The dissociation of the diatomic gases cost a lot of energy and thus puts a limit to the temperatures reached inside the bubble.

For bubbles with a high Argon content, we observed period doubling with a 3-way symmetry in the horizontal plane, suggesting the bubbles are distorted by an n=3 spherical harmonic. To explain this observation, we must have a smaller light-emitting region inside a bubble, or volume radiation. We can rule out that the light comes from a blackbody filling (almost) the entire bubble.

From the experiments presented in this thesis and our published and submitted papers, I find enough evidence to convince myself and, hopefully in time, many others that there really is a significant temperature and pressure gradient inside a sonoluminescing bubble. The measurements I have done all agree that the light-emitting region is smaller than the bubble itself.

**Future research**

To further back up the conclusion of this thesis, more experiments can be made. Firstly, more measurements on interference patterns should be done. The last minute discovery I made⁹ (when looking at old data and writing this thesis), that it appears that large interference peaks occur when the size of the light-detector is smaller, is potentially very important in determining the size of the light emitting region. This measurement should not be too complex to set up, and I hope Mogens Levinsen will do it after I leave this institute. The measurement should be set up with detectors of varying angular size positioned close to each other. This gives

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⁸ Even though we have a referee report claiming that their method was superior to ours.

⁹ See Figure 24 on page 44.
measurements of both the angular size of the pattern, and the maximum peak intensity as a function of detector size. It would also be interesting to examine if the type of noble gas is important for the interference effects. One could easily imagine that Xenon bubbles have a larger light-emitting region than Argon or Helium bubbles. This would be evident if the maximum peak intensity changes with the noble gas species.

**Final words**

With the work I have done over the last few years I hope to have given a valuable contribution to the scientific community.

All questions regarding this thesis are welcomed. My e-mail address is: jeppe@seidelindam.dk
Bibliography


[7] Jeppe Seidelin Dam and Mogens T. Levinsen (paper on period doubling with n=3 symmetry based on data similar to what is presented in this thesis, not finished by deadline of this thesis.) (2006?).


[35] Private communication during my stay in his group at UCLA.


[46] As reported in various news reports, e.g. http://www.physorg.com/news70035549.html


[49] Data is compressed with the open source zlib codec, which is fast and quite effective, while being unencumbered by patents and copyright. Data files are compressed to about 60% of the uncompressed file size. More information can be found at http://www.zlib.net/


The Quasi Monte Carlo method I use is based on Halton sequences (only recommended for dimensions lower than ~8, which is fine in my case where the dimensionality is 4 at most (3 space and 1 wavelength dimension)). An introduction can be found at: http://www.puc-rio.br/marco.ind/quasi_mc.html

[63] The refractive index of dust-like particles is 1.5 according to http://geo.arc.nasa.gov/sgg/tarfox/whatnew/SpecialSection/JGRspecsec2_ppr1.html or http://kortlink.dk/2rx6 (the last one is a shortcut to the first link, only easier to type).

Size of the Light-Emitting Region in a Sonoluminescing Bubble

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The size of the light-emitting region is a key parameter toward understanding the light-emitting processes in a sonoluminescing bubble. Here we present measurements of interference effects from particles with a diameter of approximately 2 μm situated 6–10 μm from a sonoluminescing bubble. From the angular size of the pattern and from an estimated distance to the particles we conclude that the light-emitting region of a sonoluminescing bubble is smaller than commonly believed (see, e.g., S. Hilgenfeldt, S. Grossman, and D. Lohse [Nature (London) 398, 402 (1999)]. We argue that an upper limit of the size of the light-emitting region is approximately 200 nm.

Knowledge of the size of the light-emitting region is of key interest in the understanding of light-emitting processes in a sonoluminescing bubble. However, different theories predict not only different temperatures and degree of opacity, but also much different sizes of the light-emitting region. Some models (see, e.g., the key paper by Hilgenfeldt et al. [1]) view conditions inside the bubble as uniform, whereas others suggest considerable differences in pressures, and thus temperatures, throughout the bubble [2–4] leading to a smaller and hotter light-emitting region.

Here we present some exciting new experimental results that offer a surprising way of measuring this elusive quantity.

Trentalange and Pandey [5] proposed using Hanbury Brown–Twiss (HBT) single photon correlation to measure the size of the light-emitting region. In Ref. [6] it is found that doing this requires an immense amount of data (correlating of the order of $10^{10}$ pulses from several channels) especially since, as will be seen, individual time tracks have to be recorded. To the authors’ knowledge no results from such measurements have yet been published. In fact, our experiment was originally designed for making the HBT measurement, but so far we have not reached any conclusions. However, in the process of analyzing our data, we found [7–9] (see Figs. 1 and 2 in Ref. [7]) that the autocorrelations are higher than the cross correlations. This experimental fact raised questions, to which we now believe we not only have the answers, but also that from these we can draw conclusions regarding the size of the light-emitting region.

The experimental setup is described in detail in Ref. [8]. We use three optical fibers (1 mm) 6 cm from the center of a spherical cell (diameter 6 cm corresponding to a resonance of $\sim 25.1$ kHz). The optical fibers are mounted near each other as shown in Fig. 1. The angles between fibers as seen from the bubble are calculated using the distances between the centers of the fiber openings and the distance to the center of the flask. The fibers connect to photomultiplier tubes (PMT), which are connected to separate amplifying systems. A computer registers the maximum amplitude of the flashes. As the fibers are relatively thin, covering only a solid angle of 0.4 msrad, and thus catch only a few photons per flash, we usually make a running average of the amplitude of 20 flashes to clearly discern any interesting features.

Visual inspection of the averaged time tracks revealed the presence of some prominent features. In one fiber (channel), e.g., the light intensity suddenly drops to about 50%, rises to over 200%, and then drops down to 50% again. With some overlap, this is seen in another channel. This happens in a time span of 400 flashes (16 ms). The highest relative intensity fluctuations we have observed are down to 30% and up to 500% while the duration in time varies from 25 to 1000 flashes. Often secondary peaks are also seen. The structures are never being seen simultaneously in fibers placed 15° or more apart (including fibers 180° apart). This rules out the effect being directly coupled to the bubble dynamics. The phenomenon is observed at all drive pressures and argon contents, for which stable sonoluminescence with reasonable intensity can be obtained.

These features are responsible for the initial enhancement of the autocorrelations seen in Fig. 1 of Ref. [7].

FIG. 1. Configuration of the experiment. In the enlargement the fibers are seen end on from the bubble, “Hor” being the horizontal plane. The angular distance, as seen from the bubble, between fibers 0 and 2 and 1 and 2 is 2.75°, while the distance between 0 and 1 is 4.76°. The solid angle viewed by fiber is 0.4 msrad.
However, since they only are seen nearly simultaneously with fibers placed close together, they will not affect cross correlations for fibers placed far apart as when studying the spatial dependence of period doubling. As regards the Hanbury Brown–Twiss effect though, the features introduce a disastrous bias.

Quite a few possibilities come to mind when trying to deduce the origin of the features. The sheer size, the secondary peaks, and the fact that they are not seen when a larger solid angle (50 msrad) is employed all suggest that the features can be explained by an interference pattern with a central peak slowly traversing the fibers. The most likely way for such a pattern to be produced is a dust particle being strobé by the bubble while passing the line of sight to the fiber. Such particles are known to be attracted to the bubble after some time (see Fig. 2 in Weninger et al. [10] and also Ref. [11]). Partly because interference thus seems the most plausible cause, but also because of the extremely interesting conclusions that may be drawn regarding the size and nature of the emitting region inside the bubble, we concentrate on this explanation. Later we discuss other possible scenarios and why these can be refuted.

In order to check the hypothesis of interference caused by a nearby small particle we cleaned the system carefully and refilled with water passed through two consecutive 0.2 μm millipore filters. Repeating the measurement twice with water cleaned in this fashion showed the features to be extremely rare, if at all present (a few per hour). We then added to the water a drop containing poly(methyl methacrylate) (PMMA) particles of size 6 ± 1.8 μm having a refractive index of 1.49 resulting in 5 ppm solid mass (manufacturer Polysciences Inc.). The size and refractive index are chosen to match those of common dust [12].

Immediately the patterns reappeared, leaving little doubt about the validity of our interpretation. An example is shown in Fig. 2(a). However, now we not only see features like those already described above, but also less prominent, larger angle features [as estimated from their overlap of all three fibers; see Fig. 2(b)], consistent with interference patterns originating from larger particles or even simple shadowing by large particles. With the large spread in particle size, this is to be expected.

From Figs. 1 and 2(a) it can be deduced that the size of the interference pattern measured from maximum to minimum intensity for this particular case is about 5°, i.e., the distance between the most distant detectors. (Notice that structures less than of order 1° would be smeared across the opening of the fiber.) A noteworthy difference is that the patterns with a strong middle peak are always of size 5° for the PMMA particle while always of size 3° for the dust particles. This is a consequence of the stringent requirements set by the condition of strong constructive interference. We should add that we have tried polystyrene particles of sizes 0.40, 1.05, and 2.84 μm of narrow distributions. These have a refractive index of 1.58 and are not seen.

If the interference patterns are a result of diffraction by a nearby particle presumably having a higher index of refraction than water, simultaneous measurements of the distance between particle and bubble can be used to calculate an upper limit on the size of the light-emitting region. To this end, notice that if the angular size of the light-emitting region, as seen from the particle, is larger than 5°, the interference effect would be quenched. Light emitted from the center of the bubble will result in an interference peak directly behind the particle. Light emitted from a point some distance away from the center will also result in a peak; however, this peak would be displaced, being directly behind the particle as seen from the point of light emission. Thus, to produce the observed peaks, the light-emitting region, as seen from the particle, has to be significantly smaller than the angular size of the observed interference pattern. The validity of this postulate has been checked by superposition of suitably chosen interference patterns. As the interference pattern is of a size near 5°, the light-emitting region must be smaller than 5°, observed from the particle. If we assume that the particle is placed 10 μm from the bubble, in accordance with Weninger et al. [10], an upper limit of the size of the light-emitting region can be estimated.

$$s \ll \sin(5°) \times 10 \, \mu\text{m} = 0.9 \, \mu\text{m}.$$  

Thus the radius $s$ of the light-emitting region would have an upper limit of a few hundred nanometers, i.e., significantly less than the minimum radius of the bubble. In effect the upper limit of the light-emitting region is determined by only two factors, namely, the angular size of the resulting interference pattern and the distance between the light-emitting region and the particle at the time of light emission. Calculations on the following model of interference effects from a particle confirm these results indicating a distance to this of approximately 6 μm giving $s \ll 0.5 \, \mu\text{m}$.

![Fig. 2. Interference patterns caused by PMMA particles. Notice that the patterns in (a) are not exactly simultaneous in the three channels while in (b) there is a nearly complete overlap. Running average over 30 flashes (~1.2 ms).](image-url)
The model is constructed as follows. The light source is modeled as an isotropic point source emitter in water at a given distance from the particle. The amplitude observed at large distances is now calculated according to an integral of Kirchhoff’s equation [13] over a spherical shell centered on the source and containing the particle (see Fig. 3). The wave amplitude is in the model assumed to be constant over the entire shell. The phase of the wave is also constant upon the shell—except for the small region occupied by the particle, where the lower velocity of light in the particle shifts the phase. However, reflection, etc., inside the particle is neglected. The change $\theta$ in relative phase can be estimated as

$$\theta = \frac{2\pi d (1.49 - 1.33)}{\lambda}. \quad (2)$$

Here $d$ is the distance traveled through the particle and $\lambda$ is the wavelength. The refractive index of water is 1.33 compared to that of the particle of 1.49. To obtain a significant interference effect from such a particle, the phase change must be at least of order $\pi$. With the spectral response of our detection system in mind (maximum near 300 nm, see Fig. 2 in Ref. [8]), this translates to particles with a diameter of order 1 $\mu$m, a common size for dust particles according to [12]. Calculations on the model, as well as on the model discussed in the next paragraph, suggest that the distance between bubble and particle is 5–10 $\mu$m which is in good agreement with Fig. 2A in Ref. [10]. An example of a pattern calculated using the model is shown in Fig. 3 revealing a close resemblance to the observed patterns.

The minimum possible distance between particle and the center of the bubble at the point of light emission can be estimated using the following argument. The amount of water closer to the bubble wall than the center of the particle remains constant over one full acoustic cycle, as the particle moves along with the water. A lower limit is given by the center of the particle not being able to get closer than one particle radius at the point of maximum bubble radius. This minimal amount of water between bubble and particle corresponds to a distance of around 10 $\mu$m at the point of light emission, depending on the radius of the particle and the bubble parameters.

In some cases we observe a peculiar phenomenon (here for a dust particle). An example is shown in Fig. 4. These patterns are seen to occur repeatedly every 2150 flashes more or less clearly for more than 150 times. Sometimes the interference patterns occur in a manner suggestive of the strobed position of a particle passing by with different directions for each occurrence. The repetition of the strobed positions of the particle is not inconsistent with the simulations of streamlines by Verraes et al. [14] as shown in their Fig. A1 with the particle being dragged along by the velocity field. Similar short series of periodic behavior, although never as clear as in the case described above, have periodicity ranging from 900–20 000 flashes. A recent measurement [15] with vesicles caught in the flow near a bubble oscillating on a solid surface shows these moving in the flow field and reappearing with a constant frequency similar to our observations in Fig. 4.

Of course other explanations can be imagined. Turning the argument of Eq. (1) upside down, if the light-emitting region covers the whole interior of the bubble more or less uniformly then the distance to a particle should be $\gg 20 \mu$m. However, a distant particle would have to be large thus acting as a classical spherical lens. With a refractive index of 1.5 the particle should, however, cover an angle of $15^\circ$ seen from the bubble in order to reach the central peak height observed, but this is now narrow and surrounded by a large dark region while no secondary peaks are possible. Regarding the regular recurrence of the pattern, it seems more likely that a particle close to the bubble is caught in some kind of a vertex and thus reappears every so often, than would a distant particle. The observed patterns could also result from other phenomena; e.g., if an ingoing jet is formed, the resulting shape of the bubble could be the focusing mechanism. A drift in the position of the jet then explains the change of direction of the features. For other reasons, however, jets are not likely to occur in sonoluminescence far from boundaries (see, e.g., Ref. [16]). Furthermore, it is unlikely that the occurrence of a jet should not have consequences for the emission in other directions.

These alternative explanations have severe problems with reproducing the shape of the patterns even if they are able to produce the magnitude and angular size of this. From this and the reasons stated above, we conclude that the patterns do originate in close-by particles.

To summarize our present Letter, in a previous Letter [7] we presented a correlation analysis. We show here that the elevated autocorrelations (with respect to the cross correlations) are due to the presence of nearby dust particles giving rise to interference. The particles have no effect on the cross correlation from the larger angles, as the size of the interference pattern is small.

Further measurements are needed to pin down the exact size of the light-emitting region. This could be

![Figure 3](image_url)

**FIG. 3.** The model used for the calculation of the interference pattern created by a particle close to the bubble. The pattern calculated from the model using particle diameter 2 $\mu$m, distance 6 $\mu$m, and refractive index 1.49.
done with small angle detectors fitted with short-pass (500 nm) filters viewing from one direction. At an angle of 90° a camera mounted on a long distance microscope is placed using a longer wavelength, pulsed light source to illuminate the bubble and potential particle. Thus one could simultaneously measure the size of the interference pattern and the particle to bubble distance, i.e., the two essential parameters when calculating the upper limit of the size of the light-emitting region. To further improve the experiment, spherical particles with a narrow size distribution should be used. Intimate knowledge regarding the particles would in all circumstances make for a better model—and quite possibly allow one to estimate the actual size, shape, and opacity of the light-emitting region. Furthermore, valuable information regarding the flow around the bubble may be extracted from the regular recurrence of the particles and the apparent shifting directions of passage. These observations could also give a clue to the distance between bubble and particle.

Our interpretation of the features as interference patterns coupled with our model calculations suggest that the radius of the light-emitting region is much less than 0.5 μm. This is in agreement with model calculations by Moss et al. [2,17] and Burnett et al. [18], who find that the light emission is from an optically thick core (~0.1 μm) with an optically thin halo surrounding it.

The validity of this picture is supported by recent measurements of anisotropic light emission and period doubling [7–9]. An important probable consequence is that the temperature in the center is much higher than the approximately 15,000 K given by the uniform model [1].

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Second Mode of Recycling together with Period Doubling Links Single-Bubble and Multibubble Sonoluminescence

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We report the existence of a second type of recycling mode that occurs for air-seeded bubbles. Observation of period doubling in both the stable, the first type, and the second type of recycling mode, together with simultaneous measurement of the relative phase of light emission compared to the drive, shows that the instability boundaries of period doubling and bubble extinction are mainly determined by the bubble size irregardless of the gas composition. The second type mode seems to represent a link between single-bubble and multibubble sonoluminescence.

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The collapse of the bubble in single-bubble sonoluminescence (SBSL) [1] is an extremely nonlinear phenomenon. Driven close to the boundary for extinction, stable anisotropic emission can be observed [2], and a further increase in ultrasound power will lead to destruction of the bubble due to uncontrollable growth of shape instabilities. While the dynamics and chemical processes in SBSL are fairly well understood, the connection to multibubble sonoluminescence (MBSL) is still unclear. Here we present observations on a new mode of recycling SBSL (type 2) that seems to represent a link between air-seeded SBSL and MBSL.

Both argon-seeded bubbles [3] and air-seeded bubbles [4–6] may survive in a recycling mode (type 1 Refs. [4,5]) at sound pressures close to the extinction level when partial gas pressure in the liquid is high. The second type of recycling is observed for partial gas levels at even higher levels than the type 1, but at higher gas levels the band widens up with a hysteretic overlap to the type 1 mode. Since the emission in all cases is spatially anisotropic, this phenomenon is presumably coupled to shape distortion.

The type 2 mode is a true single-bubble mode in the sense that with a resolution of 2.5 ns we never see more than one flash per sound cycle. If the light emitting bubble disappears, a new bubble has to be introduced by external means. This then enters in the previous mode of operation if outside the region of hysteresis. Thus the type 2 mode is completely reproducible. In the following we present studies of the anisotropic period doubling, spectra, spatial stability, and relative phase of the type 2 mode.

The type 1 collapse is believed to be connected to pinch-off (and possible recombination) of microbubbles, which in the case of air-seeded bubbles can lead to a renewed intake of air. According to the dissociation theory [9,10], the surplus of nitrogen and oxygen is burned off while the level of argon builds up to its former level through rectified diffusion. In turn this leads to a slow increase in the light emission until the cycle repeats. The gas in the surviving part of the bubble thus mainly consists of argon. In what follows we shall present arguments that the new recycling mode is related to mostly air filled bubbles that reach the instability threshold before the burn off becomes efficient.

The vessel and the filling procedure is described in detail in a previous publication [8] with experiments performed at drive frequency 22.114 kHz and temperature 9 °C. However, the detection and data collection systems have undergone several changes. The detection system consists of eight photomultiplier tubes (PMT Hamamatsu R3478 & R2076, rise time 1.3 ns) to provide spatial information. These are nearly evenly distributed in the horizontal plane of the bubble viewing it at a solid angle of approximately 0.050 sr. The positioning of narrow band optical filters in front of the PMTs allows for the recording of an eight point frequency spectrum. The signals are fed to preamplifiers (Hamamatsu C7319) and then to two simultaneously digitizing analog-to-digital (A/D) data acquisition cards (ADLink 9810, 12 bits, 20 MHz, 4 channel) run in parallel. Continuous time series (for convenience partitioned into 300 0000 peak values I(\(t_n\)) of the flashes for each channel) can thus be obtained. A second HP 33120A function generator provides a triangular signal phase locked to the ultrasound drive. Suitably phase shifted, this unambiguously produces a voltage proportional to the timing of the pulse. This voltage can be recorded instead of one of the PMT signals by the A/D card, with the external timing signal for this produced by an ninth PMT coupled through a fast discriminator. Finally stroboscopic recordings are obtained using a digital camera together with a homemade long range microscope.

The timing of the flash relative to the phase of the sound field (denoted relative phase) has been shown in the stable case to be related monotonically to the size of the bubble.
[11,12]. Since the time scale of the recycling is much longer than the cycle of the sound, it is fair to assume the bubble to be in a semistable equilibrium state, where a similar relation between size and relative phase holds. Therefore we have investigated the relationship between period doubling and relative phase (indicative of size) in the recycling modes and also the relationship to the surprising existence of period doubled states in the weak emission regime right after the sudden drop in light emission. By period doubling we mean a periodic change between $2^m$ different intensity levels such that $I(\vec{r}, t_n + 2^m) = I(\vec{r}, t_n)$ where $I(\vec{r}, t_n)$ is the intensity at flash time $t_n$ in the spatial direction $\vec{r}$ and $m \geq 1$ a positive integer. For some of the observations we will for clarity in the displays make a running average. This has to be done separately for $n$ even or odd.

When a recycling state is reached either by slowly increasing the forcing or by jolting the bubble with a sudden peak in sound amplitude, the recycling may take place for a very long time before the bubble either breaks or settles down again in a quiet state. Part of a recording of such a state is displayed in Fig. 1 where the intensity from one PMT (upper trace) shows the development in light emission in the type 1 recycling state for two near-extinction events. Degassing is done to 25% atmospheric pressure. However, the bubble suddenly settles into a second type of recycling state (type 2), the nature of which we shall discuss shortly. Just before the sudden drops in intensity a period doubling of the intensity is observed. The lower trace shows the corresponding development of the relative phase of the light pulses with time.

For the steady state sonoluminescing bubble we also find a completely stable period doubling. The intensity level here is just about the same as that of the recycling mode. However, the sound level needed is always somewhat lower than that needed to instigate the type 2 mode. Since the relative phase depends not only on the size of the bubble but also on sound amplitude, a direct comparison between the relative phases of the light pulses is not possible. However, information about the relationship is gained in the following manner. The ultrasound amplitude is set at the threshold value for stable period doubling. The level is then abruptly changed to that of the recycling mode. The time constant of the change of the sound level is given by the quality factor $Q$ of the system which is of the order of $\sim 100$ corresponding to $\sim 5$ ms. Since this time is much shorter than the diffusion times involved, the phase corresponding to the collapse of a bubble of the right size for period doubling under stable conditions, can be measured at the pressure of the recycling mode by extrapolation of this exponential approach in phase. The conclusion from this kind of measurement is that the period doublings in the two cases take place at nearly exactly the same relative phases and that the states therefore are analogous in nature.

Surprisingly a small region of period doubling is seen shortly after the sudden drops in intensity at a much smaller light intensity. Both period doublings are observed to be anisotropic with the same $180^\circ$ symmetry as observed for the steady state period doubling. The vertical lines in the lower part of Fig. 1 denotes the timing of the onset of the period doublings, while the horizontal line shows the relative phases associated with the two period doubled states.

Remarkably we again find a very close agreement between the relative phase of the timing of the period doubling in the two cases even though the pulse height is much reduced in the second case (from 350 to $\sim 90$ mV). This we interpret as showing that the phenomenon takes place at a certain bubble size and as a strong indication of the validity of the above assumption of semiequilibrium.

Conversely we can interpret the above observations as indicating that the relative phase gives a true measure of the size of the bubble also throughout a recycling mode (note though that the relative phase also depends on acoustic pressure). This aspect is of great interest in cases like the present one, where a direct measurement of the size, e.g., by Mie scattering is prevented by the preference of the measurement of other parameters.

Let us now turn to the new type 2 recycling case. In Fig. 2 we display an enlarged picture of the type 2 mode seen in the last part of Fig. 1. Altogether the recycling took place for 85 cycles before the bubble finally succumbed, although much longer tracks can be obtained. With only slight degassing, the type 2 mode can stay on for hours.

![Figure 1](image.png)

**FIG. 1.** Upper trace displays part of a recording of the light intensity with the bubble in the recycling mode. Running average over 30 peak values. Note the various period-doubling regimes. Simultaneous measurements from the other PMTs show the period doubling always to be geometric of nature. Lower trace shows the corresponding development of the relative phase. Vertical lines denote the timing of the onset of period doublings, while the horizontal line shows the associated relative phase. Degassing to 25%.
toward the ultraviolet. From the position of the peak a fit to the spectrum of a type 2 event. Compared to the stably emitting bubble spectrum, the spectrum falls off drastically for the spectrum of a type 2 event. The bubble is mainly composed by argon before and right after the pinch-off. As indicated by the spectra the bubble being much colder in the type 2 mode compared with SBSL and the intermittent recycling mode. Since the lower light level of the type 1 recycling is believed to be caused by the combined effect of pinch-off and diffusion. The bubble is much colder after the collapse. The experiments described above therefore lead us to the following interpretation.

The upper trace shows the variations in the light intensity seen by one PMT. The maximum light intensity varies quite a bit, but in all the cycles period doubling is observable. The variations are partly statistical in nature, but also a sign of the radiation from the bubble being anisotropic as can be ascertained from viewing the other channels. Now, however, the light intensity never reaches the level from before the drop, but stays at a much reduced level of around 45 mV.

The lower trace shows the simultaneous recording of the relative phase. Here we see an extraordinary consensus as regards the bubble size at the period doubling and the catastrophic events, the horizontal level drawn being that of the period doublings of the type 1 recycling mode and the stable mode corrected for the pressure dependence. As this indicates the same size for all the three cases the difference in light emission is most likely caused by a change in gas composition leading to a change in maximum temperature reached in the collapse.

To check this, we have attempted to measure the spectrum of the recycling modes. Since the lower light level of the type 2 mode compared with SBSL and the intermittent nature makes obtaining a spectrum with a spectrometer extremely difficult, we have resorted to making a crude eight point spectrum using narrow band filters (FWHM 10 nm) in front of the PMTs. As a reference we use the spectrum of a stable SBSL bubble. This has, in a previous measurement, been found to be well approximated by blackbody radiation at 14 000 K for ~25% degassing. In Fig. 3 we show the result of such a relative measurement for the spectrum of a type 2 event. Compared to the stably emitting bubble spectrum, the spectrum falls off drastically toward the ultraviolet. From the position of the peak a fit to blackbody radiation would give $T \sim 8000$ K but the spectrum falls off too fast toward both the ultraviolet and the infrared. Whether that feature is related to the 310 nm OH emission line is not known at present. A similar spectrum is obtained in the beginning of the cycle for a type 1 event although it looks slightly hotter, while at the end of the cycle the spectrum is close to that of the stable bubble. These spectra are also shown in Fig. 3. Note that the spectrum for type 2 looks rather similar to that of air-seeded MBSL. The spectrum represents an average over many different events but this is obviously also the case for MBSL. We have employed a homemade long distance microscope together with stroboscopic lighting to image the bubble with a CCD camera. Within the 10 µm resolution obtained there only seems to be a single bubble moving vertically a few tens of micrometers in the beginning of the type 2 cycle.

At a slight degassing level (~ 90%), the range of the sound pressure is sufficient to see the dependence on this as displayed in Fig. 4. Here the stable state spectrum is not known but the figure clearly shows the same qualitative behavior, with the bubble being much colder in the type 2 mode. Furthermore, we see the content of infrared growing as the sound pressure is raised. The sound pressure is not known at this degassing level. However, the drive amplitude is nearly constant throughout the entire range from ~25% to atmospheric pressure, where we observe the type 2 mode. This indicates that the sound pressure too is constant.

As noted above, the type 1 recycling is believed to be caused by the combined effect of pinch-off and diffusion. The bubble is mainly composed by argon before and right after the pinch-off. As indicated by the spectra the bubble is much colder after the collapse. The experiments described above therefore lead us to the following interpretation.

![FIG. 2. Upper trace displays an expanded part of type 2 recycling (later half of the recording shown in Fig. 1). The trace starts with the drop from the end of a cycle of the type 1 recycling mode. Notice the period doubling. Lower trace shows the corresponding development of the relative phase of the light pulses with time with the horizontal line indicating the relative phase at the level of period doubling in the type 1 recycling mode.](image1)

![FIG. 3. Lower curve (× and +): Relative spectra of a bubble in type 2 mode (degassing to ~25%) normalized by the radiation from a stably emitting bubble. A switch of the 404 and 360 nm filters has been performed. Upper curves: Same for type 1, at, respectively, the low (+) and the high emission end (×).](image2)
tation. After the collapse the bubble grows by diffusion toward the size called for by the applied sound pressure but is too cold for efficient rectified diffusion thus having a high content of diatomic gases. This picture is supported by considering the existence of period doubling and the difference in diffusion times for argon and the diatomic gases caused by the strong difference in concentration. Measurement of the relative phase of the flash indicate that the onset of period doubling and shape instability depends mostly on size. This second mode of recycling may give valuable insight in the connection between multibubble and single-bubble sonoluminescence with this mode being a kind of SBSL equivalent of the MBSL state. It is worth noting here the 1 order of magnitude change in MBSL radiation going from argon to air-seeded bubbles found by Sehgal, Sutherland, and Verrall [14]. The authors acknowledge financial support from the Danish National Science Foundation.

FIG. 4. Relative spectrum of a bubble in type 2 mode normalized by the radiation from a stably emitting bubble with drive amplitude as parameter. From below 137.6, 139.5, 140.8, 142.0, and 143.2 mV.

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A commonly accepted view is that stable Single Bubble Sonoluminescence (SBSL) can only be achieved in the presence of a noble gas or hydrogen. In air-seeded bubbles, the content of diatomic gases is burned off to leave the small amount of argon needed to sustain stable operation. Here we report that long term stable SBSL can be sustained with only nitrogen, oxygen, or nitrogen/oxygen mixtures being present. Compared to that of a stable argon bubble, the emission is much weaker and the spectrum looks much colder. Oscillating states as well as recycling states are also observed. An intriguing saturation effect seems connected with the presence of water vapor in the bubble.

Recently we reported a second type of recycling mode in Single Bubble Sonoluminescence (SBSL) [1]. This occurs for air-seeded bubbles with dissolved gas content higher than approximately 20% of the saturation level, when driven past their shape instability limit. In contrast to the usual recycling mode denoted type 1, which reaches the same maximum intensity level as that of the stable bubble, the intensity in the new type 2 mode is down by approximately a factor of 20. Furthermore, spectra of the emission from a single bubble in the type 2 mode look like the bubble is much colder than an argon dominated bubble, thus providing a link to multi bubble sonoluminescence (MBSL).

The dissociation hypothesis [2] was invented to explain the stability of SBSL air-seeded bubbles at dissolution levels, where the bubble growth elsewhere would not be limited. This hypothesis, since verified in many experiments, suggests that the bubbles burn off their content of diatomic gases leaving behind the small amount of argon needed to sustain stable operation. The chemical reaction products, readily dissolvable, diffuse out of the bubble during the expansion and following contraction. Our interpretation of the type 2 mode follows this picture. The bubble is driven so hard that after the splitting off of a microbubble as is the usual explanation for the type 1 event, it sucks in air too fast to burn off the diatomic molecules. Thus it grows to the instability limit while still being predominantly an air bubble. The role of the water vapor could not be established.

In order to check this hypothesis we prepared a cell with gas mixtures involving no noble gasses (nor hydrogen). Gasses used are either pure nitrogen, oxygen, or a 4:1 nitrogen-oxygen mixture to simulate air without argon. The noble gas content in all cases are below 1ppm.

As we had expected, SBSL in the type 2 recycling state is possible and at the same level of emission. The spectrum also looks very similar to that obtained from air-seeded bubbles. Thus our hypothesis regarding the type 2 recycling seems verified.

However, to our surprise, stable SBSL was also possible. This has to the best of our knowledge not been reported before. The stable radiation was obtained for extended periods of up to 45 minutes for nitrogen bubbles. The fluctuations and long term drift of the relative phase of the flashes were 100 ns or less with short term fluctuations within 40 ns and no preferred direction of drift. At higher levels of gas pressure an oscillating state is encountered at the low end of the emission range. Such an oscillating state has previously been reported for nitrogen bubbles [3]. These authors also reported on irregular peaking emission. This phenomenon we find to be associated with small amounts of argon being allowed to diffuse into the cell.

The spectrum of the stable phase very much coincides with that of the type 2 case for air seeded bubbles with total output being similar in the case of nitrogen and the nitrogen/oxygen mixture but an order of magnitude lower for pure oxygen which also seems to be colder. Thus this experiment is further verification of the dissociation theory and fits the picture of the strong and hotter emission from an air-seeded bubble being connected with argon accumulation. To obtain stable emission from non-noble gasses, however, must call for a delicate balance between the diffusion into the bubble and the burning of diatomic gasses. Presumably this leaves the bubble much more susceptible to environmental factors like microstreaming, nearby particles, and chemical balance. All factors, that are expected to affect the long term stability. In view of this, while the levels of fluctuations and long term drift are higher than would be expected for e.g. air or argon seeded bubbles at high levels of emission which can be stable at ns levels, the changes are still surprisingly small.

The cell used is described in more detail in ref. [4], but is essentially a 5 or 6 cm high and 6 cm diameter cylindrical quartz cell with metal caps at both ends so it can be sealed using a pressure relief bag. Piezoelectric transducers are mounted on one or both caps for the drive, which for most experiments reported (6 cm cell), is at a frequency of 22140 Hz. A notable difference is the use of a heater to avoid contamination with hydrogen.

The procedure finally adopted to prepare the water vapor
FIG. 1: Timeseries showing a progression for stepwise increase of sound pressure through stable states into type 2 behavior. Gas content 880 mbar nitrogen/oxygen 4:1 mixture as prepared at room temperature. Measurement performed at 9°C. Upper trace emission at 486 nm ± 5nm. Lower trace the relative phase. Average over 1000 flashes.

As described in ref. [1], a crude spectrum is obtained by placing 7 photomultiplier tubes (PMT’s) around the cell with narrow band (10 nm) optical filters in front. The signals are amplified and collected by a computer with an extra PMT giving the timing signal for the sampling. This method was chosen primarily because it allows us to get spectra of weak and shortlived states but also since it allows us to get flash by flash control of the averaging time of the spectra. Thus we have precise knowledge of the time of collection so the radiative state of the bubble can be established. The timing of the flash is also recorded with a resolution of 50 ns with averaging providing a better resolution for slower fluctuations. For extremely weak signals (oxygen bubbles), however, a different procedure, to be described later, was used.

The stable states were in the case of the nitrogen/oxygen mixture found in the whole interval investigated from dissolved gas levels of 180 to 900 mbar. For nitrogen the range of degassing where a stable state was observed is 100 to 320 mbar, with the intensity level falling below our limit of detection at the low end. However, type 2 emission was observed up to 900 mbar. At low levels of dissolved gas, the interval of stability is quite narrow with the range of stability seemingly being wider for pure nitrogen than for the nitrogen/oxygen mixture. Near ambient pressure the intervals open up and we are able to measure the change in spectrum versus amplitude of the drive. Apart from type 2 (only recycling state present), we also see the oscillating state observed by Hiller et al [3]. These states look very much like those recently observed for air-seeded bubble at very low drives by Thomas et al [5]. This suggests that these low intensity states from air-seeded bubbles have the same origin of being mainly nitrogen/oxygen bubbles.

With gas levels of more than approximately 200 mbar, we encounter the type 2 recycling. This has essentially the same time scale of recycling as the air-seeded bubbles, which strongly supports our explanation of type 2 recycling having its origin in incomplete burn-off. Also it indicates the size $R_0$ of the bubbles at breaking point to be of the same scale of approximately 6 µm. This is corroborated by observation of an anisotropic 10% period doubling in the measurements of the 280 mbar nitrogen bubble just before and at the level of type 2 recycling.

In Fig. 1 we display a 100 s long timeseries obtained for a single bubble in water prepared at a dissolved gas level of 880 mbar (nitrogen/oxygen (4:1) mixture) at room temperature and cooled to 9°C. The amplitude of the drive is stepped up rapidly at 4 distinct short time intervals with stable regions of emission in between, before finally type 2 recycling sets in. Note the overlap in regimes, as the bubble several times relapses into the quiet state, although the sound pressure is kept constant in this last part of the sequence. Since the emission is weak and narrow band optical filters are employed, the average number of photons recorded per flash is of the order of 1. Our claim of stability is therefore based on the absence of oscillations or peaking and the stability of the relative phase.

FIG. 2: Spectra of constant emission (×) dissolved gas level 220 nitrogen, (+) 220 mbar oxygen normalized with emission from stable air bubble. The curves are fitted black body radiation normalized within a constant with black body radiation at 14000 K. Upper curve $T = 8000$ K, lower curve 6580 K. Drive frequency 23200 Hz

is as follows. The water is subjected to alternating degassing at water vapor pressure and flushing with the final gas to be used for several hours in 15 minutes intervals. Under this whole process the water is violently stirred by a magnetic stirrer. After a final 15 min. flushing with the final gas composition and concentration, the water is transferred in a closed system to the cell and cooled to the operating temperature (9°C) for several hours before commencing measurements.
In Fig. 2 we show spectra taken for stably emitting nitrogen bubbles (dissolved gas level of 220 mbar) and oxygen bubbles (220 mbar). Especially the oxygen bubble is so weak that it can hardly be discerned by the unaided eye even after long time adjustment to complete darkness. These spectra are therefore obtained by freerunning the data acquisition cards at 5 MHz alternatingly for a prolonged period. Using the information from the phase-locked sawtooth signal fed to both cards one can then obtain averaged flash intensities from the remaining 6 channels with a highly improved signal/noise ratio.

The spectra have been normalized by the spectrum of an air bubble driven at a sufficiently low level to ensure that the spectrometer is not overloaded. The latter spectrum has in a separate experiment been shown to fit well to 14000 K blackbody radiation in the regime of 300 nm - 700 nm but dropping below this in the extreme ultraviolet range (VUV) (see e.g. Fig. 3). The spectra can be well fitted to blackbody radiation with temperatures 8000 K and 6580 for nitrogen respectively oxygen with the slight overshoot in the VUV range caused by the corresponding drop in the normalizing spectrum. By comparison a stable weakly radiating air-seeded bubble still displays a spectrum like that of the reference bubble apart from a scaling factor. The temperatures obtained are remarkable close to the expected dissociation temperatures for the gasses involved.

The change in the spectrum when increasing the amplitude in the type 2 case displayed in Fig. 1 is shown in Fig. 4. The spectrum starts out looking very much like those obtained for the stable case but gradually change to look like a blackbody spectrum. For comparison we have included the spectrum of a type 2 state measured for an air-seeded bubble at the same level of dissolution. As seen by comparison to Fig. 2 there is an overlap with the constant emission state. For comparison we have included the spectrum of a type 2 state measured for an air-seeded bubble at 8000 K normalized with black body radiation at 14000 K for comparison.

Since we are not able to follow the bubble movements simultaneously with obtaining the spectrum, we resort to using the relative phase for calculating the bubble size. The model is based on the Rayleigh-Plesset equations taking into account water vapor but disregarding the chemical processes. A problem is that for obvious reasons we are not able to use the dissociation theory to calculate the actual composition of the gas inside the bubble as we clearly deal with incomplete burn-off.

The calibration can thus only be based on two fixpoints, the occurrence of period doubling, and the onset of shape instability leading to type 2 recycling. This fixes the bubble size to approximately 6-7 μm at the onset of type 2 behavior. The absolute pressure $P_a$ is more difficult to assess. However, a knowledge of the cell gained from measurements on air seeded bubbles leads us to adopt the value $P_a \approx 1.5$ bar for the corresponding pressure. Assuming a constant average concentration of nitrogen in the bubble, a model calculation leads to $c \approx 0.001$ compared to the degassing level of $c \approx 0.3$. From the diffusion equation we find that the bubble must burn approximately $10^5$ molecules of nitrogen per flash which is the same order of magnitude as found for an air-seeded bubble.

The spectral temperature is determined by fits to the blackbody spectrum which for the nitrogen spectrum fits quite well. For the oscillating state at drive levels below that of the stable state and a transient state into the
stable state, the apparent temperature also oscillates or peaks. For the transient state (results shown in Fig. 4), for most of the time the light emission is quite low compared to the stable emission. In the peaks, however, the emission goes up at all wavelengths but most in the ultraviolet and actually overshoots the stable emission. At the same time the bubble is decreasing in size, the inference being that the bubble is trying to burn off nitrogen and turn into an argon dominated bubble. Due to the normalization with the known spectrum of an argon bubble, the prefactor is proportional to a product of emitting surface area and duration of flash. Exact knowledge of these factors would be necessary for a complete analysis. Unfortunately, both parameters would be very hard to measure due to the short timescale involved. It is clear though, that the lower the drive and the smaller the bubble, the higher the apparent temperature. This trend, also found for air-seeded bubbles, could be connected with the presence of water vapor.$$^3$$

Although it is difficult to get quantitative information from this analysis, some interesting information arise from assuming the bubble radius at the time of emission to be proportional to \( R_0 \), with a proportionality factor that is relatively independent of drive amplitude, and plotting the prefactor as function of the radius \( R_0 \) squared. In Fig. 4 we present this kind of plot for an oscillating state and a peaking state below the stable state, the transient state, and a peaking state above the stable state, together with an analysis for a type 1 state (air-seeded) at a similar level of degassing for comparison. As seen, in the case of pure nitrogen, the prefactors at small ambient radii are proportional to \( R_0^2 \), but at higher values of the ambient radius saturation sets in. Naively one could therefore interpret the slopes to be proportional to the flash duration. A tentative explanation for the saturation could be that the actual volume of emission is smaller than the actual bubble size. As the apparent temperatures do not show much variation, this interpretation is in fact relatively robust independent of whether volume or surface emission is assumed. In fact assuming volume emission would aggravate the problem of reconciliating the changes in emission with the changes in bubble size. We wish to stress that while the absolute values of \( R_0 \) and \( P_a \) are naturally questionable, the functional dependences displayed in Figs. 4 & 5 are robust.

The result for the recycling state of an air-seeded bubble is quite different. The recycling is of type 1 but having the hump that eventually results in type 2 recycling at higher levels of applied sound pressure. For small bubble radius, where the bubble according to our analysis of recycling states is mostly an air bubble we find the same linear behavior as for the nitrogen bubble ending with a trend towards saturation before the shrinking associated with the burn-off takes over. Finally we observe a new linear increase associated with the continued argon intake. Stable states are all located to the right of the transient curves.

To conclude we have shown the possibility of obtaining stable SBSL using non-noble gasses with total removal of noble gasses as a critical condition for stability. The experiments confirm the interpretation of the newly found type 2 recycling mode as originating in incomplete burn-off of diatomic gasses. The presence of stable emission is only permitted if the different gasses are prohibited from accumulating in the bubble. In the case of nitrogen, the only chemical processes possible involve water vapor. This raises the intriguing question of whether the stability could be caused by hydrogen actually accumu-

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**FIG. 4:** Timeseries obtained for a nitrogen bubble prepared with 280 mbar gas content showing a peaking state. The sound pressure converted from the drive voltage as described in the text is approximately 1.42 bar. Also shown are the fits for the ambient radius \( R_0 \), apparent temperature \( T \), and prefactor \( a \).

**FIG. 5:** The prefactors as function of \( R_0^2 \). \( P_a \) 1.36 (+), 1.39 (x), 1.42 (♀), 1.45 bar (●). Air-seeded bubble in type 1 mode (○).
lating in the bubble. Such an accumulation is, however, not possible for the stable oxygen seeded bubble where the only possible reaction products are $\text{H}_2\text{O}_2$ and $\text{O}_3$. It is worth noting that the black body fits give $\approx 8000$ K for nitrogen, and $\approx 6600$ K for oxygen which could be related to the known dissociation temperatures for these diatomic molecules. Finally, gas depletion in a boundary layer around the bubble might very well be an important factor. Furthermore, both the apparent higher temperatures for smaller bubbles and the saturation points to the amount of water vapor being an important factor. Obviously an extension of the dissociation hypothesis also for air-seeded bubbles at low drive and/or low content argon is needed.

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[6] Note that this normalization should not be interpreted as the emission from the air-seeded bubble necessarily being blackbody radiation.
**Period doubling and Single Bubble Sonoluminescence**

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**Abstract.** Single Bubble Sonoluminescence (SBSL) is an extremely nonlinear effect. It is therefore natural to look for period doubling in this system. We do find period doubling, which even can be observed directly in the emission amplitude flash by flash. However, surprisingly it is connected with a shape instability. We discuss the different spatial symmetries involved and touch on the controversy related to previous claims that period doubling could only be observed in the timing of the emission relative to the phase of the ultrasound field.

**Keywords:** Sonoluminescence, hydrodynamics, non-linearity

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**INTRODUCTION**

The most important issues at the moment in the field of Single Bubble Sonoluminescence (SBSL) are related to the question of the maximum temperature attainable in the center of the bubble. Related to this is the question of the temperature profile inside the bubble. Since the only information about this is gained from the emitted light (at least until neutrons or x-rays are detected beyond any reasonable doubt [1]), investigation of the spectrum and spatial distribution is rather crucial. That the distribution could be anisotropic under certain circumstances was shown by Weninger et al. [2]. More surprising was the observation of period doubled anisotropic emission by Dam et al. [3], since the limit for stability of the SBSL bubbles was thought to be related to breaking the spherical symmetry. However, there has been some controversy related to what order of spherical harmonic distortion is relevant in this connection (see e.g. Augsdörfer et al [4]).

Related to the above mentioned issues is the question of the opacity of the bubble i.e. is the bubble really a black body radiator as most experiments performed at drive frequencies in the kHz range would seem to suggest? For instance, in Fig. 1 the spectra of air-seeded bubbles driven at 22 kHz are displayed as function of drive amplitude. The spectra are fitted to blackbody radiation with temperatures starting at 17000 K for low drive amplitudes and gradually decreasing to 14000 K at the highest drive amplitudes.

In what follows we shall describe some new results on period doubling that throw some additional light on these issues.
PERIOD DOUBLING

Single Bubble Sonoluminescence (SBSL) is perhaps the most nonlinear effect ever observed. One of the earliest findings was the extreme precision of the collapse leading to the light emission, a picosecond clock. The very stability of this clock was seemingly violated by the first experiment that touched upon the subject of period doubling [6] in 1994. Here period doubling along the Feigenbaum route to chaos was observed to take place in the timing of the SBSL clock. However, none was seen in the intensity of the light emission itself. For a long time this experiment has been standing alone, not being reproduced by others, and without theoretical explanation. Indeed, theoretical work seemed to prove that the regime of sonoluminescence had not the slightest overlap with the regimes of period doubling found theoretically [7]. This work, however, assumed that the spherical symmetry was unbroken. This was the standing of the field until 2002 when Dam et al [3] discovered period doubling which could be observed directly in and only in the emission amplitude and which was clearly connected to a shape instability.

The effect was found in a correlation analysis but was soon after shown to be visible flash by flash [8] as fine tuning could induce the bubble to periodically change between two shape distorted states having a fixed orientation in space. Tumbling in space of the symmetry axes obviously would destroy the possibility of seeing the effect with a single detector, thus explaining why the observation was so difficult to make. Any trivial averaging would likewise prohibit observation.

The effect was discovered in stably emitting air-seeded bubbles having a certain minimum size close to or just beyond the shape instability borderline [9]. Since then it has been shown that the period doubling mostly depends on size, not on the gas involved and not on the state as nitrogen seeded bubbles [10] and bubbles undergoing relaxation oscillations [11] also show the effect.

The existence of period doubled sonoluminescing bubbles show that nonlinearities to some extent can stabilize deformed bubbles also in the parameter regime for sonoluminescence. The greater interest, however, lies in whether information can be gained about
FIGURE 2. PMT signals from two channels at cross-angles. The upper trace has been displaced up and to the left for easier viewing. The period doubling is seen to be out of phase showing it to be a spatially anisotropic effect.

the state inside the bubble at the time of emission. Nearly all measurements of SBSL, except for extremely small bubbles driven at MHZ drive frequencies [12], suggest that the spectrum is that of blackbody radiation. That the radiation should come from the whole bubble is more or less ruled out by careful calibration of the total amount of power emitted. However, this is a very difficult measurement to do with reasonable absolute calibration accuracy. The observation of period doubling on the other hand gives some information that can be used to exclude this possibility due to symmetry arguments.

**PERIOD DOUBLING WITH TWO-FOLD SYMMETRY**

The observation of period doubling with two-fold symmetry is all done with bubbles of fairly high gas content (200 mbar or more) in the boundary regime of spatial distortion where the bubble is close to distinction. The bubble in this situation is fairly big, around 6 µ in rest radius \(R_0\).

In Fig. 2 we show a typical measurement of spatially stable period doubling with output from two PMT’s positioned crosswise. The out of phase period doubling is easily discernible. The size of the period doubling is shown in Fig. 3 and seen to have a two-fold \(180^\circ\) symmetry.

For an \(n=2\) spherically deformed bubble (an ellipsoid) this could be explained by light being emitted from the surface of the bubble since only the cross-section plays a role for the intensity observed in a given direction. In this case the period doubling should be caused by the bubble shifting periodically between two ellipsoids sharing a common symmetry axis turned away from the normal to the plane of the detectors. (In the case of alignment the period doubling would be indistinguishable from that caused by period doubled spherical collapse.) However, other experiments on interference patterns in the
bubble emission generated by nearby transparent particles [13], show clearly that the emission is from a smaller region central to the bubble. The obvious deduction that follows from this is, that the period doubling is caused by diffraction in the distorted bubble surface. Model calculations indicate that to avoid too strong a distortion that would destroy the bubble, the refractive index at the inside bubble wall has to be no more than $\approx 1.1$, proving that the temperature and pressure inside the bubble must be inhomogeneous. Furthermore, the simulations show that a transparent light emitting region under these conditions can not explain the size of the period doubling. The off-set of the sine is connected to the tilt of the common symmetry axis.

The two-fold symmetry is also found in relaxation oscillating states of air seeded bubbles (see Figs. 4 & 5). From measurements of the timing of the flashes (relative phase [14]) compared to the phase of the ultrasound drive signal, one can deduce that the radius $R_0$ is the same for all the events observed and that the difference in intensity therefore must be caused by differences in gas composition. This is collaborated by measurements of the spectral color temperature which is around 7000 K for the low intensity states and 14000 K for the high intensity state. In the high intensity (type 1) relaxation state the final bubble content is argon, while in the later low intensity relaxation state the bubble content is mostly nitrogen and oxygen at all times. The salient point in the present context is that even the cool states seem to emit blackbody radiation.

**PERIOD DOUBLING WITH THREE-FOLD SYMMETRY**

New measurements on air seeded bubbles with additional argon content has shown that the two-fold symmetry is not the only state with broken spherical symmetry.

The addition of argon has the effect that the bubble can attain a larger size before reaching the shape instability border line. As seen in Fig. 6 there can now exist two high
FIGURE 4. Upper trace displays part of a recording of the light intensity with the bubble in the recycling mode. Running average over 30 peak values. Note the various period-doubling regimes. Simultaneous measurements from the other PMTs show the period doubling always to be anisotropic. Lower trace shows the corresponding development of the relative phase. Vertical lines denote the timing of the onset of period doubling, while the horizontal line shows the associated relative phase. Degassing to 25%.

intensity regimes of period doubling (this happens repeatedly in the course of a long series of relaxation oscillations).

The regime reached first has the two-fold rotational symmetry as described above. However, the latter state cannot be fitted with a \( \sin(2\theta) \) but needs a \( \sin(3\theta) \) except for an offset as demonstrated in Fig. 7. (The \( \chi^2 \) test gives a result between 5 and 20 times as high when a fit is attempted to a \( \sin(2\theta) \), ruling out this symmetry.)

Thus the state has a three-fold (120°) spatial symmetry. This points to an underlying \( n=3 \) spherical harmonic spatial distortion. The radius of the spatially deformed bubble can then be expressed in spherical coordinates at time \( t \) as

\[
r(t, \theta, \phi) = R(t) + a_n(t)Y_n(\theta, \phi)
\]  

(1)

If the bubble surface is giving out blackbody radiation in this case, the radiation would have six-fold (60°) spatial symmetry. The simplest possible period doubling should involve a switching back and forth between such two \( n=3 \) mirror states. Mirror states of \( n=3 \) spherical distortion, however, has identical cross-sections seen from any two opposing directions. If there is no other symmetry breaking mechanism no period doubling would result. Indeed, it would take a truly substantial change in refractive index of the water in the boundary layer next to the bubble to break the symmetry enough to have any detectable effect. The same symmetry argument shows that a spherical
FIGURE 5. Upper trace displays an expanded part of type 2 recycling (later half of the recording shown in Fig. 4). The trace starts with the drop from the end of a cycle of the type 1 recycling mode. Notice the period doubling. Lower trace shows the corresponding development of the relative phase of the light pulses (with bubble radius being a monotonously growing function of this). The horizontal line indicates the relative phase at the level of period doubling in the type 1 recycling mode.

FIGURE 6. The first large region of period doubling starting at \( \approx 4.8 \) s can be well fitted to \( \sin(2\theta) \) with an offset. At \( \approx 6.2 \) s the symmetry axes start to tumble and after about one second, the symmetry is three-fold (see Fig. 7).

bubble surrounding an \( n=3 \) distorted core will not be able to produce the observed threefold symmetry. The only explanation left that does not require extreme assumptions is that of a small central emission core in an \( n=3 \) distorted bubble. As before the refractive index close to the bubble wall should not be too high consistent with an inhomogeneous temperature distribution. This indeed has the three-fold symmetry for the period doubling although one would not expect a fit necessarily to be to a perfect sine.
PERIOD DOUBLING IN TIMING

In some of our cells we have accidentally excited period doubling in the cell response. This period doubling can be heard directly by ear as the cell emits the audible half harmonic. Coincidentally we have a few times observed period doubling in the timing of the flashes. We have tried to make simultaneous recordings of the signal from a microphone attached to the cell and the timing and intensity of the emitted light. But although we have recorded period doubling in the signal from the microphone, we have not succeeded in recording any simultaneous period doubling in the timing. As the time constant of the sound field in the fluid corresponds to $\approx 100$ ashes, it is perhaps unrealistic to expect the effect to originate in this fashion. However, that such generation of subharmonics could be behind the old observations, can not be ruled out yet.

DISCUSSION

The observations of period doubling certainly seem to favor the picture of the light emission coming from a hot opaque core. However, we still have no inkling of the nature of the feedback mechanism that allows the bubble to enter the period doubling states. Furthermore, we are still in need of self consistent simulations on the wave equation inside the bubble that take into account the position dependent refractive index and the diffraction in the bubble surface in order to turn the experimental information into precise knowledge about the extent of the core and the temperature distribution inside the bubble. And the truly exciting riddle is still: how does such a small body succeed in emitting what seems to be blackbody radiation at temperatures that range from 6000 to 25000 K depending on the gasses and liquids involved.
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