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Single-bubble sonoluminescence

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Abstract

Single-bubble sonoluminescence is a phenomenon in which an acoustically trapped and periodically driven bubble collapse so strongly that the energy focusing at collapse leads to light emission. In the seminar we talk about experimental and theoretical efforts for undertanding this phenomenon and we concentrate on light emission proceses.

1 Introduction

Sonoluminescence is a process where sound transforms into light. This happens when bubbles in a liquid are excited by sound so strongly that they collapse. When bubbles implode they burst short emission of light. The phenomenon was discovered in 1934 by H.Frenzel and H.Schultes[1] as a result of work on sonar. They exposed a photographic plate to acoustic waves generated in a water bath. With that they hoped to speed up the development process. Instead, they noticed tiny dots on the plate after developing. The luminescence they observed did not result from the sound field directly but through a process called cavitation. When the pressure amplitude P_a is larger than ambient pressure $P_0 = 1$bar, the pressure in the flask becomes negative, putting the liquid under tension. Tensile portion of the pressure variation generates voids filled with gas and vapor within the liquid (bubbles). The collapse of these voids during the compression portion of the acoustic wave is extremely violent. These cavitation clouds collapse with enormous force, powerful enough to do serious damage to the surfaces of solid bodies in their vicinity. It was too difficult to analyze the effect in early experiments because of the complex environment of a large number of short-lived bubbles. This phenomenon is now referred to as multiple-bubble sonoluminescence (MBSL). Here many bubbles grow and collapse throughout the regions of most intense acoustic stress.

Sonoluminescence has been poorly understood because it is associated with a large number of cavitation bubbles. Spatial scale of individual event is on the order of a micrometer, and the temporal scale is on the order of a few nanoseconds. Because of the random nature of MBSL it was difficult to learn much about the physics of the individual cavitation events and also the resulting electromagnetic emission.

2 Discovery of single-bubble sonoluminescence

Single-buble sonoluminescence(SBSL) was discovered in 1989 by Felipe Gaitan[2], then a graduate student working with Larry Crum. Crum had seen first hints of light emission from a single bubble in 1985[2]. Gaitan's objective for his thesis was to systematically search the conditions under which a single, stable cavitation buble would produce sonoluminescence each acoustic cycle. He was carrying out a set of experiments on the oscillation and collapse of bubbles, using a flask of liquid lined with transducers tuned to set up an acoustic standing wave at the resonant frequency at the jar.

In his research for single-buble sonoluminescence, Gaitan found a regime

with moderate forcing pressure $P_a/P_0 \approx 1.2 - 1.4$ and with the water degassed to around 20% of its saturated concentration of air. He observed that "as the pressure was increased, the degassing action of the sound field was reducing the number of bubbles, causing the cavitation streamers to become very thin until only a single bubble remained. The remaining bubble was approximately 20μm in radius and [...] was remarkably stable in position and shape, remained constant in size and seemed to be pulsating in a purely radial mode. With the room lights dimmed, a greenish luminous spot the size of a pinpoint could be seen with the unaided eye, near the bubble's position in the liquid"[2]. Figure 1 shows a sketch of a typical setup for generating sonoluminescence(left), and an actual experiment(right). The blue dot in the center of the flask is the bubble emitting light. Reducing the dissolved gas

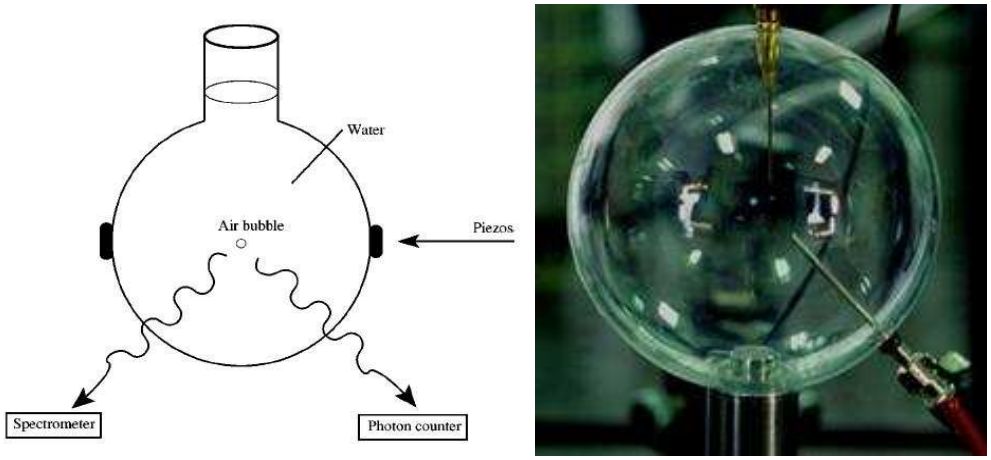

Figure 1: Sketch of a typical setup for generating sonoluminescence(left)[2], and an actual experiment(right), where the blue dot in the center of the flask is the bubble emitting light[4].

concentration makes it possible to produce a single, stable cavitation bubble that undergoes large radius excursions each cycle. Gaitan was able to find the necessary conditions for sonoluminescence. Once those conditions are achieved the system is amazingly robust. If there are no significant changes in the acoustic or liquid parameters, SBLS can be maintained for unlimited periods of time.

3 Researching SBSL

All previous work with light-emitting bubbles involved many unstable bubbles being simultaneously created and destroyed. Using a laser, a photode-

tector and the Mie-scattering algorithms it is possible to invert scattered intensity and obtain a radius-versus-time curve. Gaitan and co-workers demonstrated that their setup in fact generated a single bubble, undergoing its oscillations at a fixed, stable position at a pressure antinode of the ultrasound field in the flask. The oscillation frequency ν is that of the sinusoidal driving sound usually $20 - 40$kHz. The dynamics ob the bubble radius is strongly nonlinear. Once during each oscillation priod, the bubble, whose ambient radius R_0 is tipically around 5μm, collapses very rapidly from its maximum radius $R_max \approx 50\mu$m to a minimum radius $R_min \approx 0.5\mu$m, changing its volume by a factor of 10^6[3].The bubble expansion caused by the negative pressure is followed by a violent collapse, during which light is emitted. The process repeats itself with extraordinary precision. This is shown on figure 2.

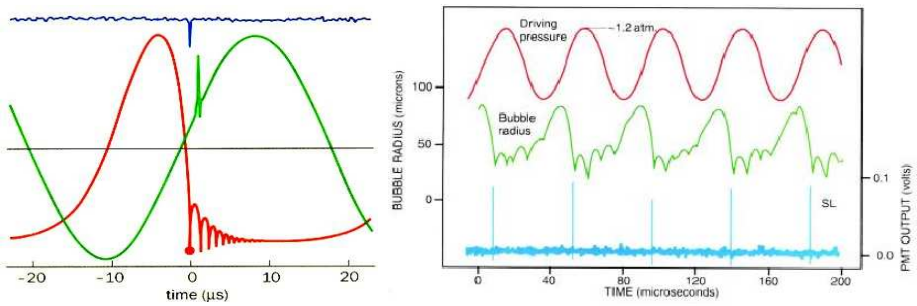

Figure 2: On lhe left side we have a single cycle, where green is the sound field, red is the bubble radius and blue is light intensity[3]. The implosion also generates an outgoing pulse of sound(spike on the sound wave). Right side shows process repeating itself[5].

A group headed by S. Putterman used improved light-scattering technique to obtain radius-time curves for SBSL to a high level of precision[1]. The results are shown in figure 3. These curves in figure 3, illustrate the transiton from a nonsonoluminescent bubble to a sonoluminescent one. As the acoustic-pressure amplitude is increased, there is a transition at which the bubble's equilibrium radius, its maximum radius and its rebound from implosive collapse are all suddenly reduced. At this pressure sonoluminescence emissions begin to occur. Computation of these radius-time curves using standard models of nonlinear bubble dynamics predicts the rebound reduction at the reduced bubble size; however, the sudden decrease in equilibrium radius is still not clearly understood[1].

R. Hiller[2] measured the spectrum of SBSL and demonstrated that it in-

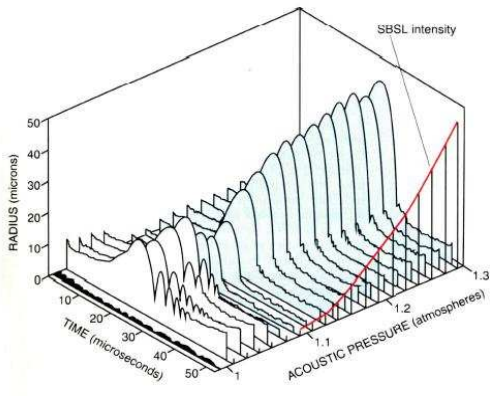

Figure 3: These measurements illustrate the chance in the beheavior of the bubble when sonoluminescence conditions are achieved. The unshaded regions are for nonSL bubble[1].

creases toward the ultraviolet(figure 4). The apparent peak in spectra is due to the strong absorbtion of wavelengths below $\sim$ 200nm by the water in the flask. A SBSL spectrum shows a smooth continuum, without spectral lines. Spectral lines are overwhelmed by continuous emission processes at high temperatures. By fitting the observed spectra to a blackbody emitter(figure 4) indicates temperature of approximately 16000K.

S. Putterman and his colleagues also tried to measure the pulse duration of the sonoluminescence flash. They found that as they selected photomultiplier tubes with increasingly faster response times, they continued to measure only the impulse response of the tubes. When they compared the impulse response of the SBSL flash with that of a 34-picosecond laser, they determined that SBSL flash extinguished faster than that of the laser, probably due to some residual ringing in the laser that is absent in SBSL[1]. It was concluded that the width of the light pulse was less than 50ps.This upper bound of the pulse width was much smaller the time during which the bubble remained in its most compressed state.

B. P. Barber demonstrated that the light intensity and amplitude of the oscillations of the bubble also depend on the concentration of the gas dissolved in the liquid, the temperature of the liquid or small amounts of surface active impurities. When the air dissolved in the liquid was replaced with pure nitrogen, SBSL disappeared. With a gas composed of (80%) nitrogen and (20%) oxygen, there was still no sonoluminescence. Only when the inert gas argon was added did SBSL light emission return. In figure 5 we can see the dependance of SBSL intensity of nobel gas mixture with nitrogen, and the

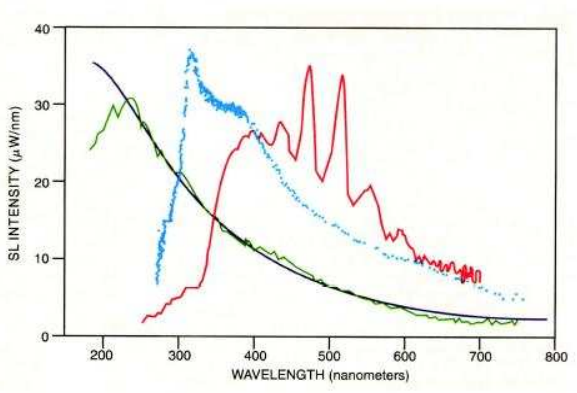

Figure 4: Sonoluminescence spectra. The green curve is for SBSL and the smooth is the tail of a blacbody for a temperature 16200K. The red and blue curves are for MBSL at different parameters, and have been shifted for comparison[1].

spectrum for various gas mixtures dissolved into water at the same conditions. Duration of the flash also depends on the gas dissolved in the bubbles. Xenon bubbles are the brightest SBSL emitters and yield the longest pulses, with widths up to 350ps and more. Only bubbles in highly degassed water can very faint flashes of about $40 - 60$ps duration be found[2].

4 Light emission

4.1 Overview

This micrometer-sized bubble in a water-filled flask can undergo oscillations of incredible violence and still maintain the stability of its spherical shape, showing precise repetitions of this highly nonlinear dynamics for millions and billions of driving cycles. In order to release a photon of visible wavelength, an atom, ion, or molecule must be excited a few eV above its ground state. A sound wave of 1-atm amplitude, by contrast, carries an energy density of typically 10^{11}eV per particle. The required tremendous energy concentration of almost 12 orders of magnitude[2] is precipitated by the rapid collapse of the sonoluminescent bubble, where the layers of water surrounding the bubble act as a radial piston compressing its interior. Since the discovery of sonoluminescence, researchers have come up with lots of creative ideas concerning light-emission processes. Almost all of them can be classified

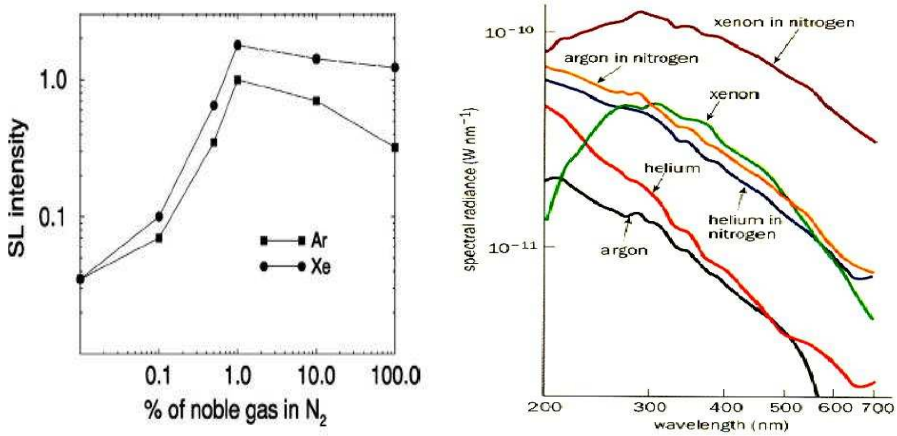

Figure 5: Left side shows dependence of SBSL intensity as a function of percentage of nobel gas mixed with nitrogen[2]. On the right side is spectrum for various gas mixtures at the same conditions[3].

under one of two headings: thermal or electrical processes.

The first attempts at explaining the mechanism behind the light emission favored electric discharges[2]. Lecshin and Rzhevkin[2] initially brought up the subject of sharge separation in cavitation bubbles. Harvey[2] thought of the bubble as the spherical capacitor with charges at the center and the wall. Upon collapse, the capacitance decreases and voltage increases until electric breakdown takes place. Since a symmetric charge distribution cannot radiate light, discharge theories in general have to assume that emitting bubble undergo an asymmetric collapse, and would predict increasing intensity of light emission as the asymmetry increases. This contradicts recent systematic studies of SBSL. Experiments under microgravity performed by Matula[2] show that an increase in symmetry leads to slightly more light rather than less. A number of theories placed the location of light emission in the liquid, rather than inside the bubble. All nonthermal models have to explain why their mechanism of emission would not be swamped by thermal radiation.

In thermal theories the energy for light emission is supplied by thermal energy resulting from an adiabatic bubble collapse. Noltingk and Neppiras[2] were the first to use Rayleigh-Plesset bubble dynamics to deduce bubble internal temperatures as high as 10000K at collapse of spherically symmetric bubble. Depending on the actual temperatures achieved during collapse, different excitations become dominant in the compressed gas. Phrase thermal emission can refer to a large variety of different processes. As temperatures

increase from several hundred to many thousand Kelvin, those processes can be molecular recombination, collision-induced emission, molecular emission, atomic recombination, radiative attachment of ions, neutral and ion bremsstrahlung, or emission from confined electrons in voids. The uncertainty about the precise temperatures of SBSL bubbles would allow most of these mechanisms. Serious argument against thermal processes was the SBSL pulse width; it is much shorter than the time for which the bubble is maximally collapsed.

The need for ultrashort pulses fueled the popularity of shock-wave models, in which a focusing shock causes the light-producing region to be much smaller than the bubble size. Gompf[2] demonstrate that a simple thermal model can result in a light pulses with the essential characteristics. Conversely, ultrashort pulses were no longer necessary. Calculations showed that shock waves for noble gases would be far less intense than for molecular gasses or might be altogether absent[2]; which is obviously not the case. All the evidence indicates that, while shock waves might be present in collapsing bubbles under certain circumstances, they do not substantially contribute to SBSL light emission.

4.2 Thermal volume emitter

Gompf[2] determined the widths of the light pulse in different parts of the optical spectrum.The measurements are shown in figure 6. Between the ultraviolet $(300-400\text{nm})$ pulse and the red $(590-650\text{nm})$ pulse there was hardly any detectable difference. This contradicted thermal models favoring black-

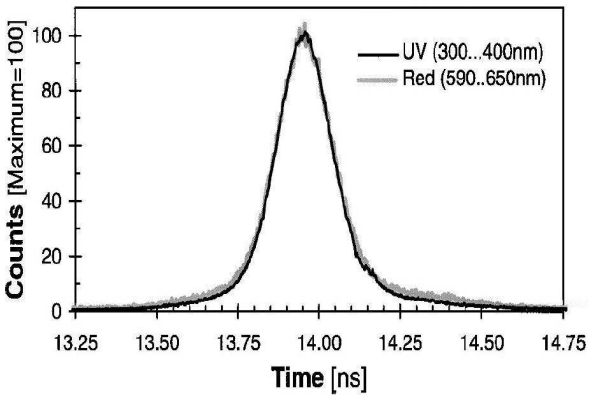

Figure 6: First measurement of SBSL pulse width in different wavelenghts[2].

body emission. These predict that the bubble is capable of emission at longer

optical wavelengths for a longer time than at short wavelengths. Meaning the red pulse should be about twice as long as the ultraviolet pulse[2]. A blackbody of a given temperature emits a spectral light intensity at wavelength λ of

$$I_{\lambda}^{Pl}[T] = \frac{2hc^2}{\lambda^5[\exp(\mathrm{hc}/\lambda \mathrm{k_B T}) - 1]} \tag{1}$$

the Planck intensity, with Planck constant h and Boltzman constant k_B and the speed of light c. Experiments measure the spectral radiance, for which we must integrate over the projected bubble surface and all solid angles:

$$P_{\lambda}^{Pl}[t] = 4\pi^2 R(t)^2 I_{\lambda}^{Pl}[T(t)]. \tag{2}$$

This formula reflects the fact that blackbody is a surface emitter. Radiation comes from its surface alone, as photons originating in deep layers are absorbed. So we can integrate over the corresponding wavelengths ranges and compare the prediction with light pulse measurement ($mogo$ceslik$ozaprim$erjavo FIG41$inFIG$13). In this model the intensity of the pulse is about two orders-of-magnitude larger than the experimental values, the duration of the pulses is at least a factor of 2 longer, and the length of the pulse dramatically varies with wavelength, which is in direct contradiction with the experiments. Experiments show that it is necessary to seek a modification of the blackbody model.

Local thermodynamic equilibrium must hold even over the short time scales of the bubble dynamics at collapse. At the immense particle densities ($n \sim 10^28\mathrm{m}^{-3}$) and high temperatures ($T \sim 10^4\mathrm{K}$) collisions between particles are very frequent (collision times are well below a picosecond)[2]. Blackbody picture assumes that the bubble is black. This requires that the mean free path of photons ($\kappa_\lambda{}^{-1}$) be much smaller than the size of the object. Here κ_λ is the absorption coefficient for photons of wavelength λ, and when divided by the gas density, κ_λ is often referred to as the opacity. A blackbody of extent radius (R) has $\kappa_\lambda R \gg 1$. The product $\tau_\lambda \equiv 2\kappa_\lambda R$ is optical thickness of the object at λ. Researchers speculated and discussed that opacity may play a role for SBSL and that the bubble might be transparent to its own photons. Calculations show that the emission volume of the optically thin shell is much larger than that of the optically thick core. The idea of a transparent bubble is the only proposition put forth to date that explains the wavelength independence of sonoluminescence radiation, and it is now widely accepted as a key ingredient to a consistent view of SBSL light emission.

For a volume emitter, the light intensity from a volume element of the bubble is dependent on the location inside the body (the depth s along the

ray of vision to the element). Using the laws of absorption and emission in a medium[2],

$$I_\lambda(s,T) = I_\lambda^{Pl}[T](1 - \exp{-\kappa_\lambda[\mathrm{T(t)]s}}), \qquad 0 > \mathrm{s} > 2\mathrm{R} \tag{3}$$

is the intensity at wavelength λ from a depth s, provided the temperature of the body is spatially uniform. Note that the source function of the emission is still I_λ^{Pl}, because local thermodynamic equilibrium is obeyed, and that for infinite absorption coefficient κ_λ, the Planck emissivity is recovered. To obtain the spectral radiance, one must carefully integrate the upper equation. Assuming isotropy of SBSL radiation, the spectral radiance is then

$$P_\lambda(t)d\lambda = 4\pi^2 R^2 I_\lambda^{Pl}[T(t)] \left(1 + \frac{\exp(-2\kappa_\lambda \mathrm{R})}{\kappa_\lambda R} + \frac{\exp(-2\kappa_\lambda \mathrm{R}) - 1}{2\kappa_\lambda^2 R^2}\right) d\lambda. \tag{4}$$

Note that the Planck radiance is now multiplied by a factor whose magnitude varies between 0 and 1, depending on the optical thickness $\tau_\lambda = 2\kappa_\lambda R$. As $\tau_\lambda \to \infty$, the Planck spectrum is recovered. In the transparent limit $\tau_\lambda \ll 1$ the radiance reduces to

$$P_\lambda^{thin}(t)d\lambda = \frac{2}{3}\tau_\lambda P_\lambda^{Pl}[T(t)]d\lambda \tag{5}$$

a spectral power that is much smaller than that for blackbody emission.

The dimensionless optical thickness satisfies $\tau_\lambda < 1$ throughout the collapse for all wavelengths, and $\tau_\lambda \gg 1$ for the dominant ultraviolet part of emission. ($mogocesklicnaslikoFIG44$)

Comparing with the blackbody calculation, it is seen, first, that the total intensity is reduced dramatically (to a few 10^5 photons per pulse). This is because the small optical thickness, leads to a drastic reduction of radiance compared to an opaque bubble. Second, the light pulses are now considerably shorter, and finally, the dependence of pulse width on wavelength has almost disappeared. All of these properties of the light pulse are now in much better agreement with those of experimentally observed SBSL pulses.

Identifying the major contributions to the absorption coefficient automatically identifies the main light emission mechanisms of SBSL. This follows from Kirchhoff's law, which states that every absorption process must balance a corresponding emission process. Inverting the absorption events identifies SBSL light emission as a combination of electron-ion bremsstrahlung, electron-neutral bremsstrahlung, and radiative recombination. None of the three processes seems to be dominant over the whole range of the SBSL parameter space.

5 A SBSL oscillation cycle

In figure 7 one cycle is shown in detail and each step of the cycle is marked. The cycle begins, by definition, when the driving pressure begins to dip into

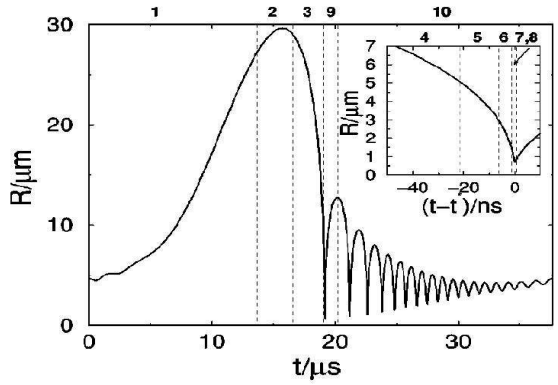

Figure 7: Classical bubble dynamics calculation for driving pressure amplitude $P_a = 1.2$atm, frequency$\nu = 26.5$kHz and ambient bubble radius $R_0 = 4.5\mu$m[2].

the negative half of its cycle, and the bubble is therefore allowed to expand. The surrounding liquid far from the bubble is degassed to some level and maintained at room temperature. Consequently, the bubble contains about 10^10 argon atoms and about $2\text{x}10^8$ water molecules at the outset.

1. Expansion. The bubble expansion is comparatively slow and the growth is sustained for almost half a cycle ($\sim$ 15ms). In this phase, the bubble is in both thermal and mass transfer equilibrium with the liquid. Because of the falling pressure inside the bubble, it gains large numbers of water-vapor molecules (evaporating from the wall) and also some gas molecules from the liquid.

2. Turnaround at maximum radius. The driving pressure begins to increase again, and the expansion comes to a halt. At maximum radius ($R_max = 7R_0$), the bubble contains little more than the initial 10^{10} argon atoms, but has collected up to 10^{11} water molecules.

3. Rayleigh collapse. As the external pressure increases, the inertial collapse of the liquid layers around the bubble begins. Even with the increased number of molecules, the internal pressure is still very low, and the collapse proceeds almost exactly like the classical collapse of an

empty cavity treated by Lord Rayleigh (1917). The radius decreases quickly (over about 4ms) from R_max to a value comparable to R_0. During this collapse, water vapor recondenses at the wall and the argon atoms again become the dominant species inside the bubble.

4. Decoupling of water vapor. About 50ns before the minimum radius is reached, the time scale of the bubble collapse ($\sim R/|\dot{R}|$) becomes smaller than the time scale for the diffusion of water vapor. The water vapor still left inside the bubble is now trapped until the reexpansion. Calculations show that about 20% water vapor should be mixed with the argon. Up to this moment, the polytropic exponent of the gas mixture has not increased significantly above 1, and the temperature has only risen to about 500K.

5. Thermal decoupling. Only $\approx$ 30ns later, the accelerating bubble wall becomes fast enough that heat can no longer escape the bubble. Until reexpansion, the bubble is now thermally isolated from the liquid as well, and the polytropic exponent γ rises quickly to its adiabatic value. The latter is determined by the mixture of 80% Ar and 20% H_2O to $\gamma \approx 1.6$. From now on, the temperature increases rapidly.

6. Onset of dissociation reactions. Once the temperature exceeds roughly 4000K, water-vapor molecules start dissociating into OH and H radicals. While the dissociation products have lower adiabatic exponents than H_2O, this endothermic reaction also consumes much energy and curbs the temperature rise. At this stage, faint molecular band light emission is a possibility.

7. Onset of light emission. Despite the temperaturelimiting influence of water vapor, about 10000K are finally reached in the bubble about 100ps before maximum compression. At this temperature, a small fraction of the Ar as well as of the O and H atoms now present undergo ionization and release free electrons. The electrons interact with the ions and neutral atoms, and emission of electromagnetic radiation (thermal bremsstrahlung and radiative recombination) begins, which spans the spectrum of visible and ultraviolet wavelengths. Sonoluminescence is observed.

8. Maximum compression. At this point, the gas density reaches (almost) solid-state values. The deceleration of the bubble wall down to zero speed has begun to enhance random shape perturbations (Rayleigh-Taylor instability) and leads to massive energy loss through acoustic

wave emission. The temperature and light emission peak, helped by the high densities that prevent further endothermic dissociation reactions.

9. Reexpansion. The bubble loses about 90% of its energy in the collapse, mostly due to acoustic emission. The reexpansion is much slower than the collapse. The Rayleigh-Taylor instability grows and may overwhelm a strongly driven bubble during this stage. Only a small increase in radius and decrease in temperature are sufficient to dramatically reduce the photon absorption coefficient and quench the light emission uniformly for all wavelengths, about 100-200ps after it has begun. Subsequently, reaction chemistry stops and thermal as well as diffusive equilibria are reestablished.

10. Afterbounces. The bubble rebounds to a much smaller size than the maximum radius before the main collapse, and for no parameter combination realized so far is there enough energy left to induce sonoluminescence during the afterbounces. The afterbounces provide a parametric excitation that can accumulate and render the bubble shape unstable. The radial motion is, however, damped rapidly until the driving pressure dips into its negative cycle once again, and the oscillation starts anew. Over the whole cycle, shape perturbations may have been enhanced (then the bubble is parametrically unstable), or a net gain or loss of gas may have resulted (diffusive instability). In the correct parameter range, the bubble is stable with respect to both processes and continues to oscillate and emit light in exactly the same fashion.

The above 10 steps hold for SBSL bubbles in water with only argon (or any other noble gas) dissolved. If in addition molecular gases such as nitrogen and oxygen are dissolved, not only water dissociates at step 6, but also these gases (at around 7000K for N_2 and O_2). The reaction products subsequently dissolve in water.

6 Consclusion

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