Single-bubble sonoluminescence

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As a result of some spectacular experiments, it has recently become possible to drive a single gas bubble of a few microns to such an extreme nonlinear motion that light is emitted which is visible to the naked eye in a darkened room. This phenomenon, associated with stable acoustic cavitation, is termed here as *single-bubble sonoluminescence* (SBBL). Even though the emitted light appears continuous to the naked eye, it actually consists of a sequence of individual flashes which are highly synchronous with the driving acoustic field. Each flash, containing about one million photons, is established to last for about 50 ps.

The spectrum of the emitted light is known to be broadband, with higher energy content in the ultraviolet. A black-body fit to the spectrum suggests temperatures nearing 10^5 K under certain conditions. However, whether SBSL is associated with simple black-body radiation obeying the Planck distribution has not yet been clearly established. Irrespective of that, the phenomenon of SBSL does involve spontaneous energy amplification by roughly 11 orders of magnitude since very low level acoustic energy in the liquid is converted to highly energetic photons. It is now proven beyond doubt that light emission occurs during the violent collapse phase of an acoustically driven gas bubble, this being the mechanism for energy amplification. The intensity of SBSL, in terms of the number of photons emitted per flash, is known to depend on several parameters, viz. drive pressure amplitude and frequency, temperature of the liquid sample and the amount and type of dissolved gas in the liquid. The upper bound on the intensity is yet to be established. The primary aim of this review article is to provide experimental evidence to substantiate many of the above statements without much discussion, since some crucial aspects of the phenomenon are still not understood.

SONOLUMINESCENCE (SL) is a term used to define the phenomenon of light emission from cavitation fields. Cavitation itself is a physical process wherein voids or bubbles are formed in a liquid under the action of reduced pressure. Even though SL has been observed from both hydrodynamically and acoustically induced cavitation, it is the latter which has been studied the most. Sonoluminescence originates from weak spots, or 'nuclei', in the liquid, which is subjected to an intense acoustic excitation. Cavities or bubbles grow from these spots during the expansion phase of the acoustic pressure field and tend to collapse during the compression phase. The motion of the bubble interface being a nonlinear one, the volume compression ratios are normally substantially higher than the volume expansion ratios. Thus, if the motion is adiabatic then the gas (and vapour) inside the bubble can get heated to very high temperatures during the bubble collapse phase, resulting in light emission. Here is, thus, a mechanism by which low-level acoustic energy is focused onto a few gas molecules, resulting in SL. The question whether SL is of black-body origin or chemiluminescence from formation and recombination of excited molecules or radicals within the bubble is not completely settled. There are other possible mechanisms as well, like 'bremsstrahlung', if the gas gets ionized. In any case, it should be noted that SL in general is quite 'dim' and the interest in its study was related to the fact that it is an indirect indicator of the extreme conditions reached within a collapsing cavitation bubble. This has implications in understanding, among other things, (a) sonochemistry, which has become a field by itself (see, for example, Lorimer and Mason[1]) and (b) potential damage to human tissue during ultrasound diagnostics.

In general, a liquid sample which is subjected to ultrasonic excitation will contain a large number of nuclei distributed randomly in the bulk. Thus, there are many potential locations from where SL can originate. This type of multibubble SL is basically uncontrolled and has been the subject of extensive studies in the past. Dependence of SL intensity on the nature of the dissolved gas, the type of liquid, and the acoustic pressure amplitude and frequency have all been investigated. There have been attempts also to correlate the phase of SL with the phase of acoustic pressure field. In addition, SL spectra have been measured primarily to understand the mechanism responsible for the observed light emission. An excellent and exhaustive summary of these findings is contained in a relatively recent review article by Walton and Reynolds[2]. Study of a physical phenomenon which is uncontrolled has its own limitations; for example, it is not possible to observe in detail the behaviour of bubble interface motion during or prior to light emission. The foundations for the study of SL from controlled cavitation were laid in a paper by Saksena and Nyborg[3]. Instead of relying on natural nuclei, they seeded artificial nuclei in the form of a stream of gas bubbles of different sizes. In particular, with glycerine–water solution as the liquid medium they found a significant difference in the threshold pressure amplitudes between SL with and SL without seeding of artificial nuclei. Thus, at certain pressure amplitudes SL could be detected only in the presence of seeded gas bubbles. From these controlled experiments they demonstrated that SL can originate not only from 'transient cavities', which grow and collapse (so violently that they disintegrate) within one acoustic cycle, but also from stably oscillating (nonlinearly) gas bubbles. At the end of their article, Saksena and Nyborg[3] indicate that there is no reason why their observations cannot be repeated with a single bubble. In fact, Gaitan and Crum[4] and Gaitan[5] were able to achieve this by observing SL from an oscillating single gas bubble trapped at the node of a standing-wave acoustic pressure field. This type of SL, which can appropriately be termed as single-bubble sonoluminescence (SBSL), is bright enough to be visible to the naked eye in a darkened room. This fantastic discovery was followed by some outstanding and revealing experimental studies of the phenomenon by a group of scientists at UCLA under the supervision of Seth Putterman. It is appropriate at this point to quote Maddox[6], who, while reporting on the phenomenon of SBSL, states that 'one puzzle with sonoluminescence is that it seems to be a means of making energy run uphill. A coherent beam of sound waves is made to travel through a liquid whereupon, when the conditions are right, flashes of light can be observed: phonons seem to have been converted into more energetic photons, some of them with ultraviolet energy'. In this article, we consider certain aspects of this exciting discovery, starting with a description of the methods for producing single-bubble sonoluminescence (SBSL).

Producing SBSL

The methods and conditions required to produce SBSL are fully described in Gaitan[5] and Barber[7]. Basically, an acoustic levitation cell has to be constructed from a glass vessel filled with a liquid and driven with one or more piezoelectric transducers. A schematic diagram of the apparatus and instrumentation used by Gaitan *et al.*[8] to produce SBSL and study some of its characteristics is shown in Figure 1. They used cylindrical and square containers driven by two hollow cylindrical piezoelectric transducers attached at the top and bottom. When the system is driven at one of its natural resonant frequencies, large-amplitude acoustic pressures are generated near the centre of the glass container. Barber[7] and Barber and Putterman[9] used spherical boiling flasks for their experiments since acoustic efficiencies are significantly higher. We[10], at the Indian Institute of Science (IISc) have used a system very similar to the one used by Barber and Putterman. A schematic diagram of this set-up is shown in Figure 2. A 500 cc boiling flask with its neck cutoff is driven by a hollow cylindrical piezoelectric transducer (ID = 19 mm; OD = 25.4 mm and $L = 50.8$ mm), which is epoxied to the flask at the bottom, with a brass spacer matching the curvatures. The transducer is excited at the desired frequency through a power amplifier driven by a signal generator. Spherical levitation cell has very sharp resonance (high Q); hence, the ability to control frequency setting from the signal generator has to be extremely good. Typically, resolution of a few hertz in tens of thousands of hertz (10 kHz) is required. The maximum pressure amplitudes are obtained at the centre of the flask, as expected, since acoustic waves generated by the surface movement get focused at this point. Hence, the centre being a node (velocity = 0) is a convenient location for stabilizing a bubble.

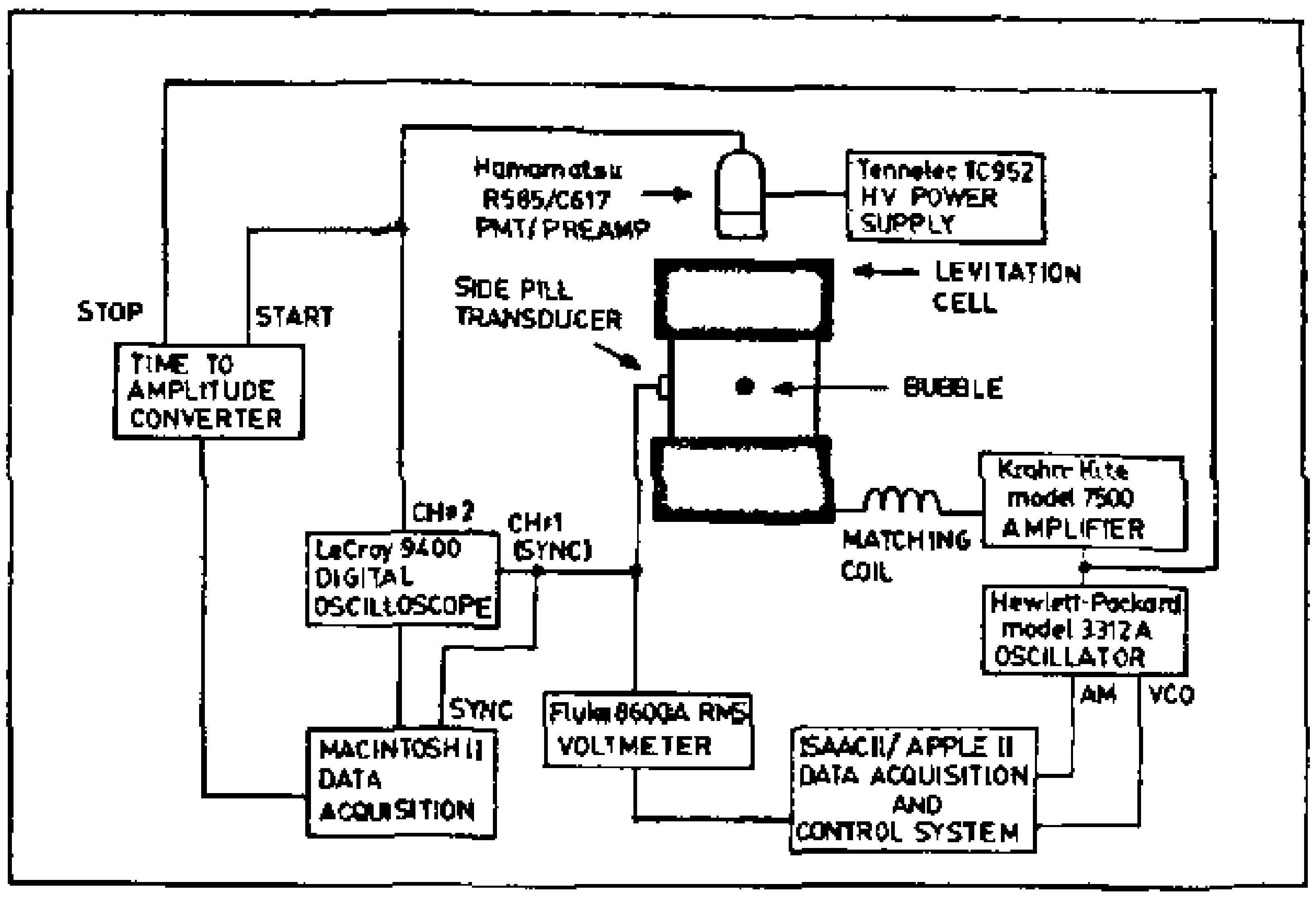

Figure 1. Schematic diagram of the experimental apparatus used by Gaitan *et al.*[8] to establish stable SBSL and measure its phase relative to acoustic pressure.

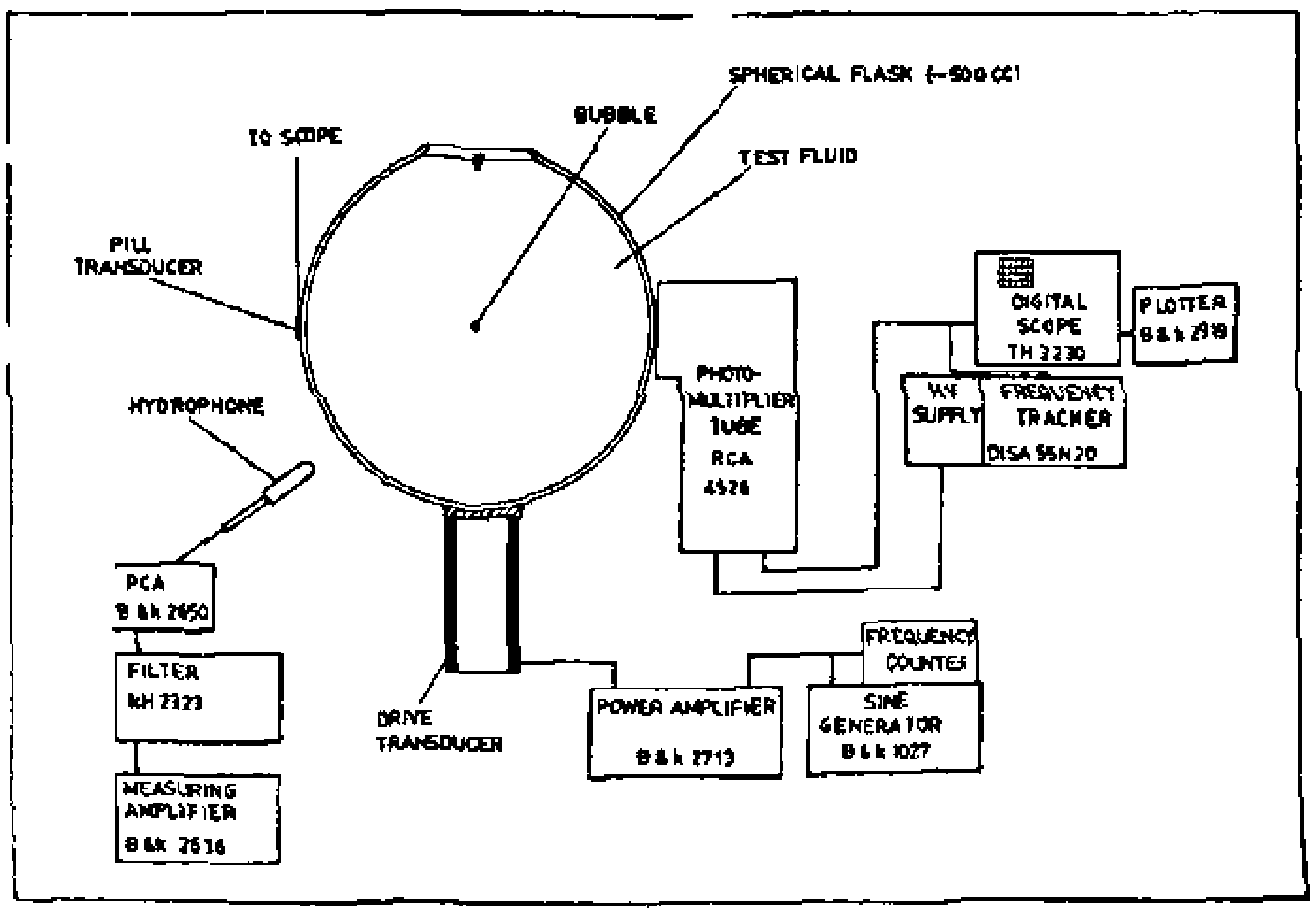

Figure 2. Schematic diagram of the experimental apparatus used at IISc for establishing stable SBSL and measuring its intensity.

Let us now describe briefly the method for producing SBSL in a spherical levitation cell, though the methodology should be applicable to other geometries as well. The first step is to look for the exact resonant frequency; this can be done by monitoring the pressure field in the flask with the help of a pill transducer attached to the wall of the flask or with a hydrophone (or microphone) located few centimeters away from the flask (see Figure 2). At resonance, there will be a sharp increase in the output from the sensing transducers. Once the operating frequency has been ascertained, at low operating power, an artificial nucleus in the form of a gas bubble is introduced into the liquid medium. This can be done with the help of a hyperdermic needle or just by tapping a glass rod at the free surface. The gas bubble so introduced will travel to the centre of the flask under the influence of the pressure gradient. Whether this can be achieved is a good test on the correctness of the operating frequency. The bubble can be levitated at the centre with sufficient power input to the drive transducer. Subsequently, the power levels can be increased gradually; the sequence of observed events is summarized in Figure 3. Thus, there is a range of pressure amplitudes for which stable single-bubble sonoluminescence (SBSL) is possible. What is truly fascinating here is that after the bubble fragments, it settles down to such an extreme nonlinear motion that light flashes are emitted continuously. This light is visible to the naked eye as a tiny blue spot in a darkened room. A photograph of SBSL obtained with the apparatus at IISc is shown on the cover. The exposure time is 8 min and the flask outline is made visible by double exposure with background lighting.

The liquid mediums used to produce SBSL so far have been filtered distilled water and glycerine-in-water mixtures of different compositions. Whatever be the liquid medium, it is essential to degas them partially to achieve stable SBSL. Furthermore, it is not possible to establish stable SBSL without the introduction of the gas bubble initially since the pressure amplitudes required to initiate cavities from natural nuclei are generally much higher than the upper threshold indicated in Figure 3. Under these conditions, transient cavities result which, typically, have lifetimes of one acoustic period. It is entirely possible and most likely that light emission originates from such transient cavities, but the characteristics of this light may be quite different from those of the light emitted from stable SBSL.

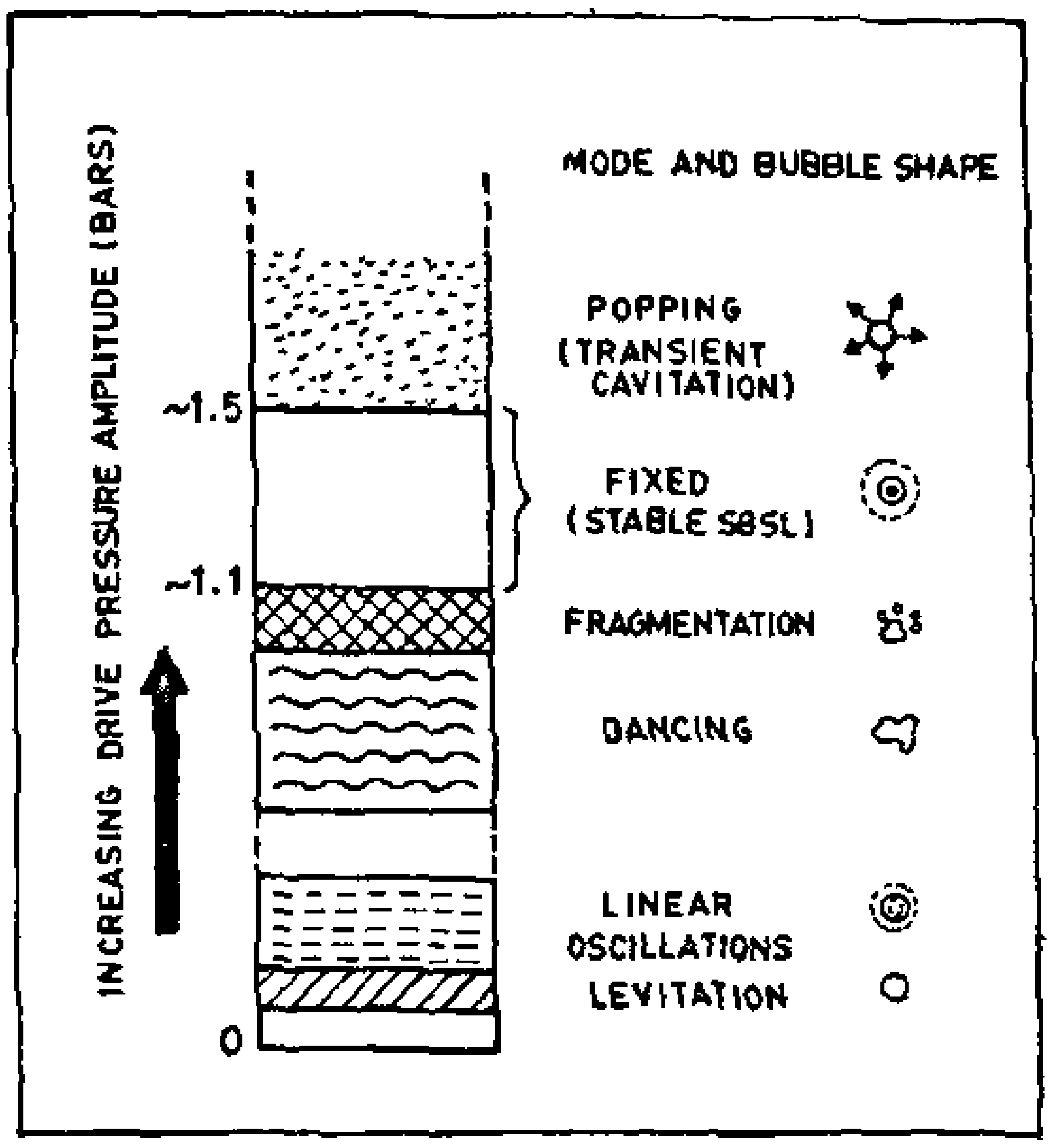

Figure 3. Diagram indicating threshold pressures for different modes of bubble motion. Stable SBSL has an upper and lower threshold pressure. In highly degassed liquids it is not possible to go through the modes at lower pressure amplitudes.

Characterization of light from SBSL

Even though Gaitan[3] and Gaitan and Crum[4] were the first ones to report the establishment of stable SBSL, their primary interest was studying the radius time history of a sonoluminescing bubble and compare it with several theoretical models. They did show that SBSL appears in a train of light flashes whose phase is highly synchronous with the driving sound field. In addition, they found that the appearance of a light flash in SBSL coincides with the collapse phase of the bubble[8]. These findings are indeed very significant, but a more startling revelation about the characteristics of the light from SBSL came from the studies of Barber and Putterman[9], and Hiller *et al.*[11]. First of all, they confirmed the synchronous property of SBSL flashes and, further, addressed questions like 'What is the duration', intensity and spectral characteristics of the light flashes?. In spite of the use of the best available equipment, they came to the conclusion that the fastest available photomultiplier (PM) tubes are not able to resolve the SBSL flash duration. Attempts to use a streak camera by Barber[7] failed since the SBSL intensity was insufficient. Using an indirect method involving a comparison of the PM output from SBSL with that of a 35 ps laser, Barber *et al.*[22] made a conservative estimate of 50 ps for flash duration. The integrated charge output from the PM tube can be used to arrive at the intensity of flashes. Initially, Barber and Putterman[9] estimated that each flash is comprised of more than 10^5 photons (over 4π radians) and that the intensity is remarkably uniform from flash to flash. The variation in intensity (represented by distribution of pulse heights) could be explained on the basis of the statistics of a few photons reaching the

PM tube. In view of some of the above characteristics of the present type of SBSL, Barber and Putterman termed it as synchronous picosecond sonoluminescence.

The spectral content of the light from SBSL as measured by Hiller *et al.*[11] is shown in Figure 4. It is clear that the spectrum is broadband, with high ultraviolet (UV) energy content; in fact, the energy increases as UV cut-off of water is reached. This provides a direct evidence of SBSL being a phenomenon wherein low-level acoustic energy in the medium (estimated to be 10^{-10} eV/molecule) is concentrated to such an extent that photons with energy level > 6 eV are emitted. This represents energy concentration by roughly 11 orders of magnitude. A black-body fit to the spectra as indicated in Figure 4 shows that the temperatures reached within the bubble could be as high as 25,000 K. In view of these extraordinary findings, it would be of interest to know what parameters influence the establishment of stable SBSL and its intensity.

Parameters influencing SBSL

As indicated earlier, as of now, stable SBSL has been established in water and glycerine–water mixtures. In the latter medium it is found that SBSL blinks on–off once every second or so[9, 13]. In degassed water it is extremely stable. From some investigations conducted by the author, it has been found that surface tension is an important parameter in establishing stable SBSL. For example, it was not possible to establish SBSL in ethylene glycol or in water when a small quantity of *n*-butyl alcohol (which reduces the value of surface tension) was added to form a 2% aqueous solution.

Intensity of SBSL (in terms of the number of photons per flash, N_p) increases by a factor of about 6 as the acoustic pressure amplitude is increased from the lower to the upper threshold value (see Figure 3), the increase being to a lesser extent as the operating frequency is decreased[9]. Cooling of the liquid sample has a very significant effect on the intensity[11, 13]. Using a quartz flask, Hiller *et al.*[11] found that in water N_p increases by a factor of 10 from 4×10^5 to 4×10^6, as it is cooled from about 20° C to 5° C; this corresponds to SBSL peak power levels of about 30 mW.

Other factors which seem to influence the intensity rather strongly are the type and amount of dissolved gas in the liquid[10, 13]. In water the number of photons per flash, N_p, increases as the dissolved gas concentration, α, measured in parts per million moles (ppm), is decreased (Figure 5). However, there is hardly any influence of α on N_p in glycerine–water mixture; the fact that SBSL blinks on-off in this medium may have some relevance. The effect of various dissolved gases on the intensity is presented in Figure 6. With oxygen as the dissolved gas the intensities are maximum, and with nitrogen they are the minimum. Here we do not attempt to explain or discuss further the above findings since the exact mechanism for light emission has not yet been established.

Theoretical aspects of SBSL

In principle, the problem of SBSL can be treated as

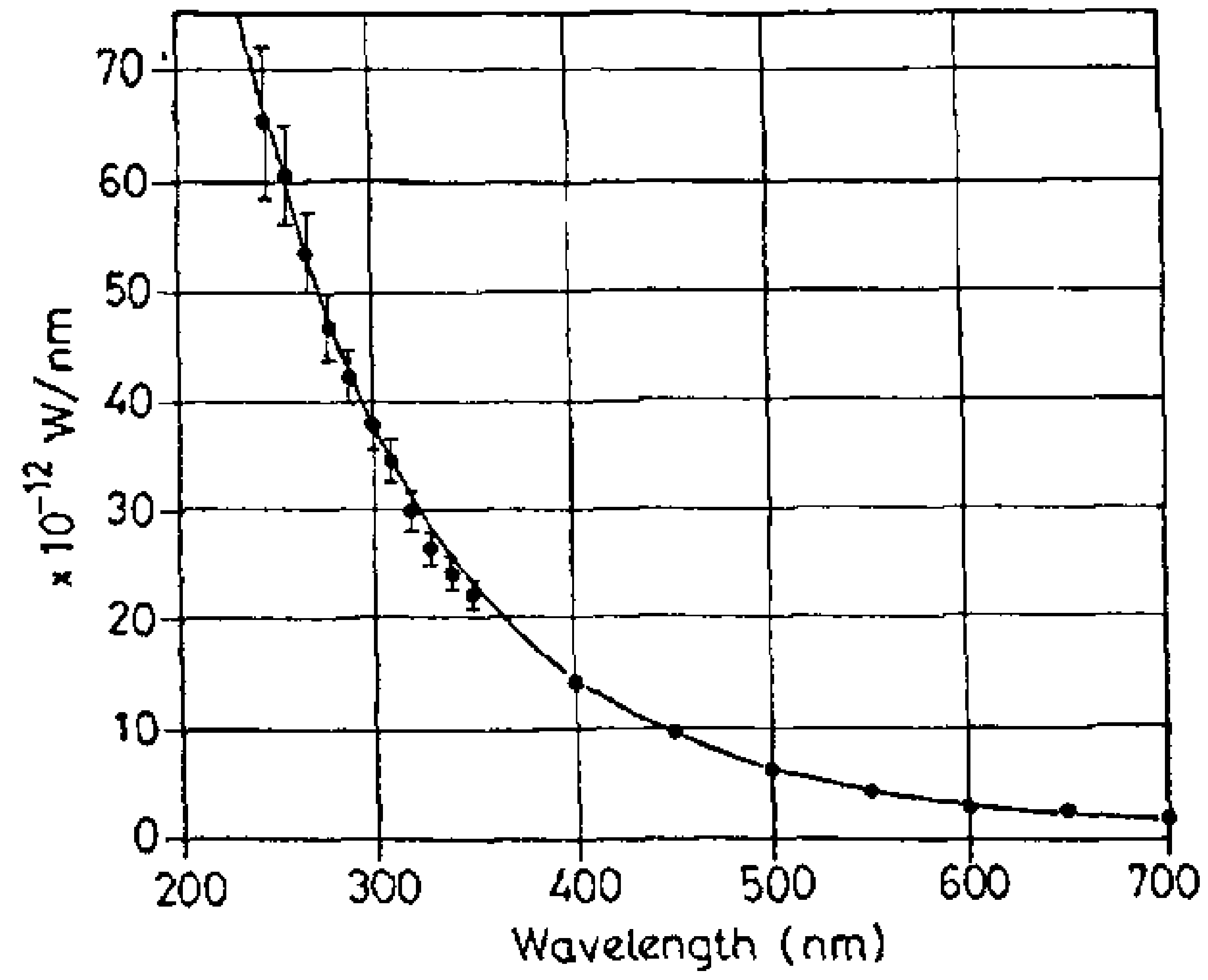

Figure 4. Spectrum of single-bubble sonoluminescence. The solid line is a 25,000 K black-body spectrum (after Hiller *et al.*[11])

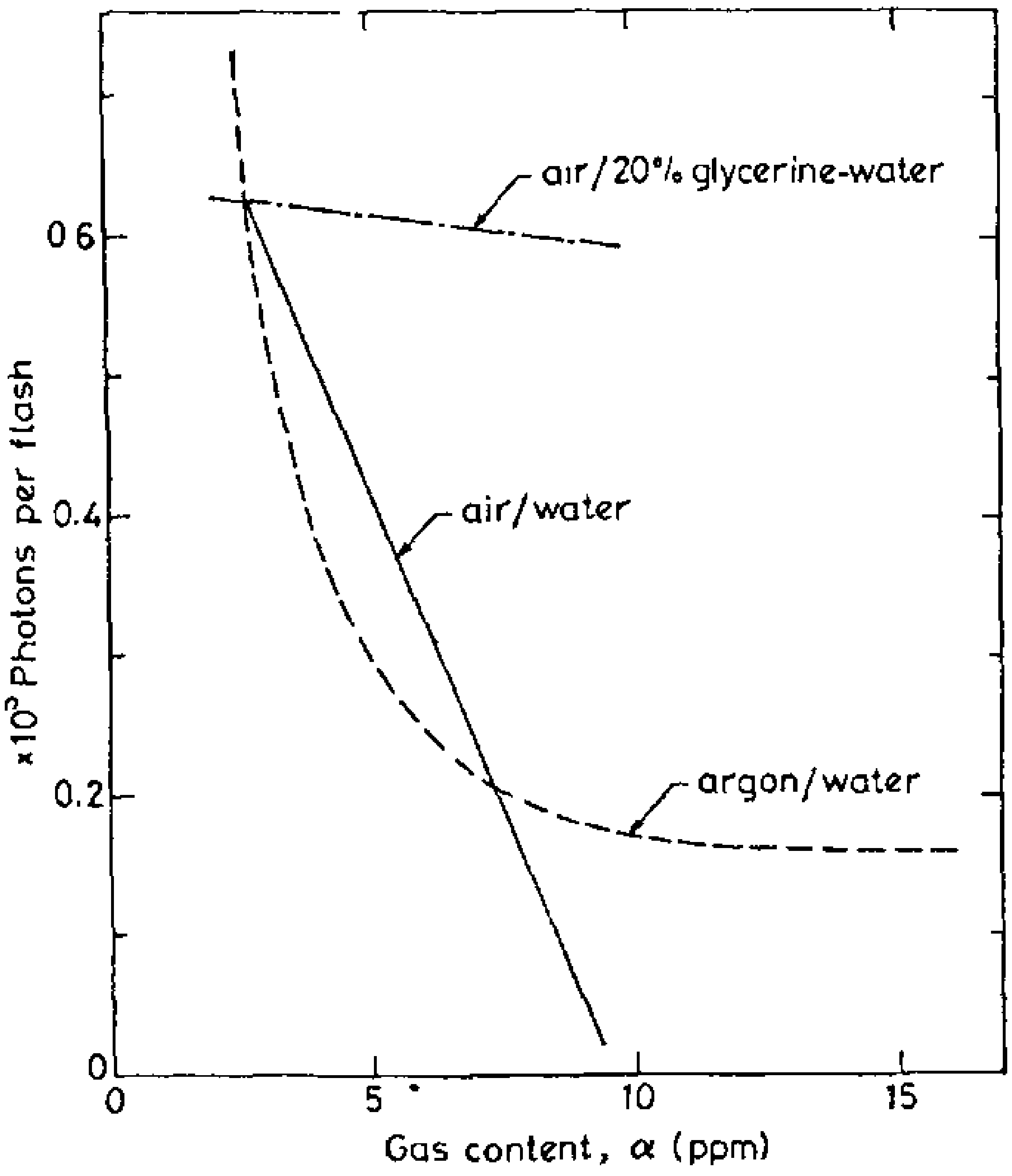

Figure 5. Effect of dissolved air concentration on maximum SBSL intensity at 20° C (after Arakeri[10]).

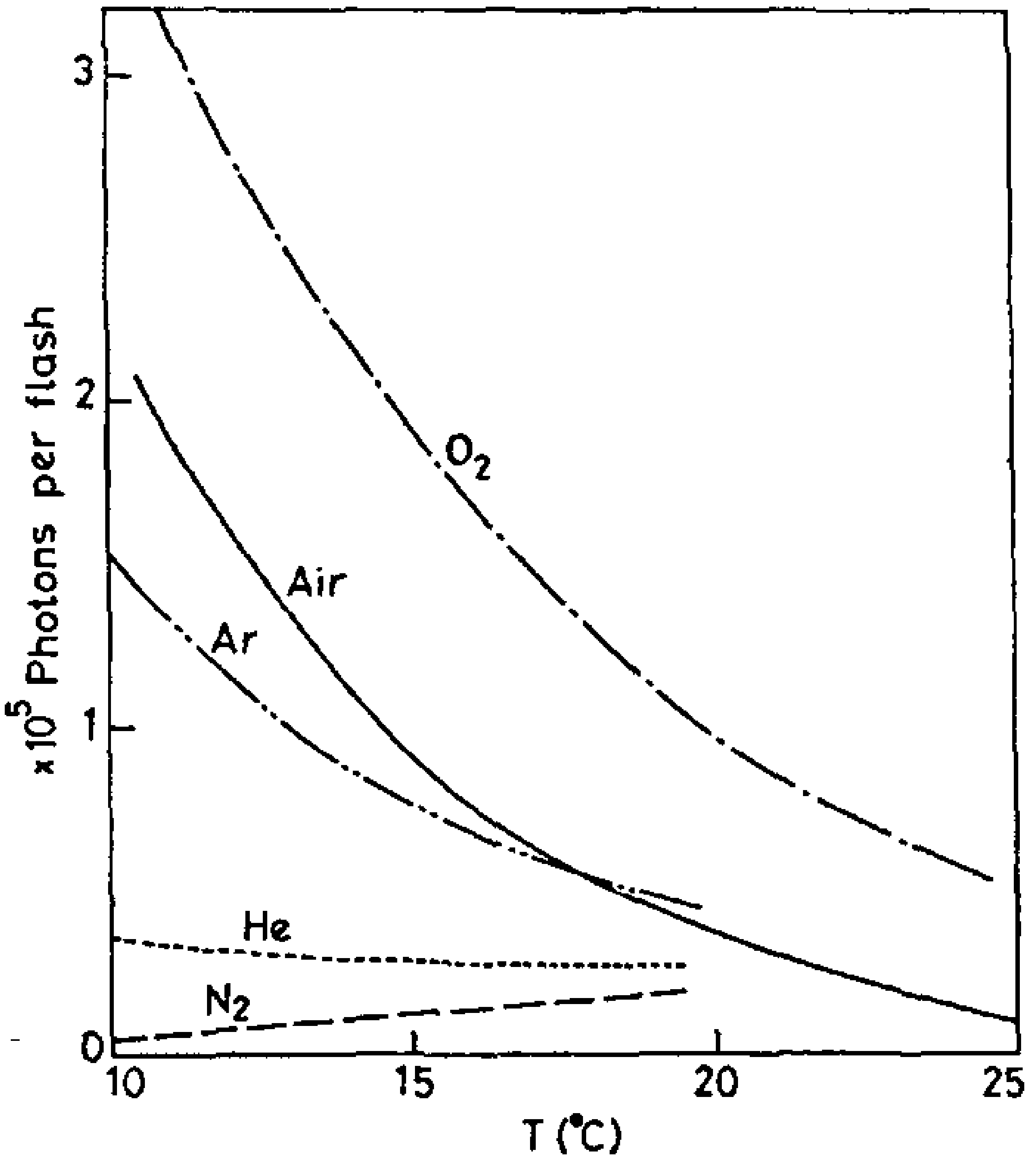

Figure 6. Maximum SBSL intensity in water as a function of temperature for various dissolved gases with $\alpha = 5$–6 ppm (after Arakeri[13]). Additional investigations have shown no measurable emission from H_2.

being equivalent to determining the state of a gas which is expanded and compressed by nonlinear motion of a spherical piston. In the present case, the bubble interface is the piston and its motion is coupled to the state of the gas inside and the forcing function, viz. the acoustic pressure field in the liquid. Therefore, the bubble dynamics problem involves determining the radius time history of a bubble subjected to a time-varying pressure field surrounding it. Solution to the exact formulation[14] would be quite a formidable task and may not always be required. A number of approximations to the solution have been reported[8, 15, 16]. Recently, Barber and Putterman[17] have indicated that most of the physics of bubble motion is recovered by solving a bubble dynamics equation which is accurate to first-order acoustic approximation in the liquid[18]. Further, the thermodynamic path of the gas inside the bubble is assumed to follow the van der Waal's adiabats. Typical solution to the bubble dynamics equation is shown in Figure 7. The bubble grows to a maximum size due to pressure reduction and then collapses violently. It rebounds to go through cyclic oscillations before repeating the motion in the next acoustic cycle. The runaway nature of the collapse was predicted over a few decades ago by Rayleigh[19], who derived and solved the incompressible version of the bubble dynamics equation. In view of Rayleigh's original contribution and later modifications by Plesset, the bubble dynamics equation is now commonly referred to as the Rayleigh–Plesset (RP) equation. The magnitudes of some significant parameters obtained from the solution indicated in Figure 7 are summarized in Table 1.

Theoretical solutions to the RP equation have been verified experimentally[8, 17]. Using the excellent timing properties of SBSL, Barber and Putterman[17] were able to resolve the phenomenon at micron level length scale and nanosecond time scale. They established clearly that the flash of light is emitted few nanoseconds before the minimum radius is reached and at that time bubble wall velocities are close to the gas speed of sound. In view of this, they suggested that a shock wave may be forming inside the bubble during the last stages of collapse, which upon focusing and rebounding at the centre may result in extremely high temperatures. Recently, Wu and Roberts[20] have derived theoretically the above-postulated mechanism for the origin of the very

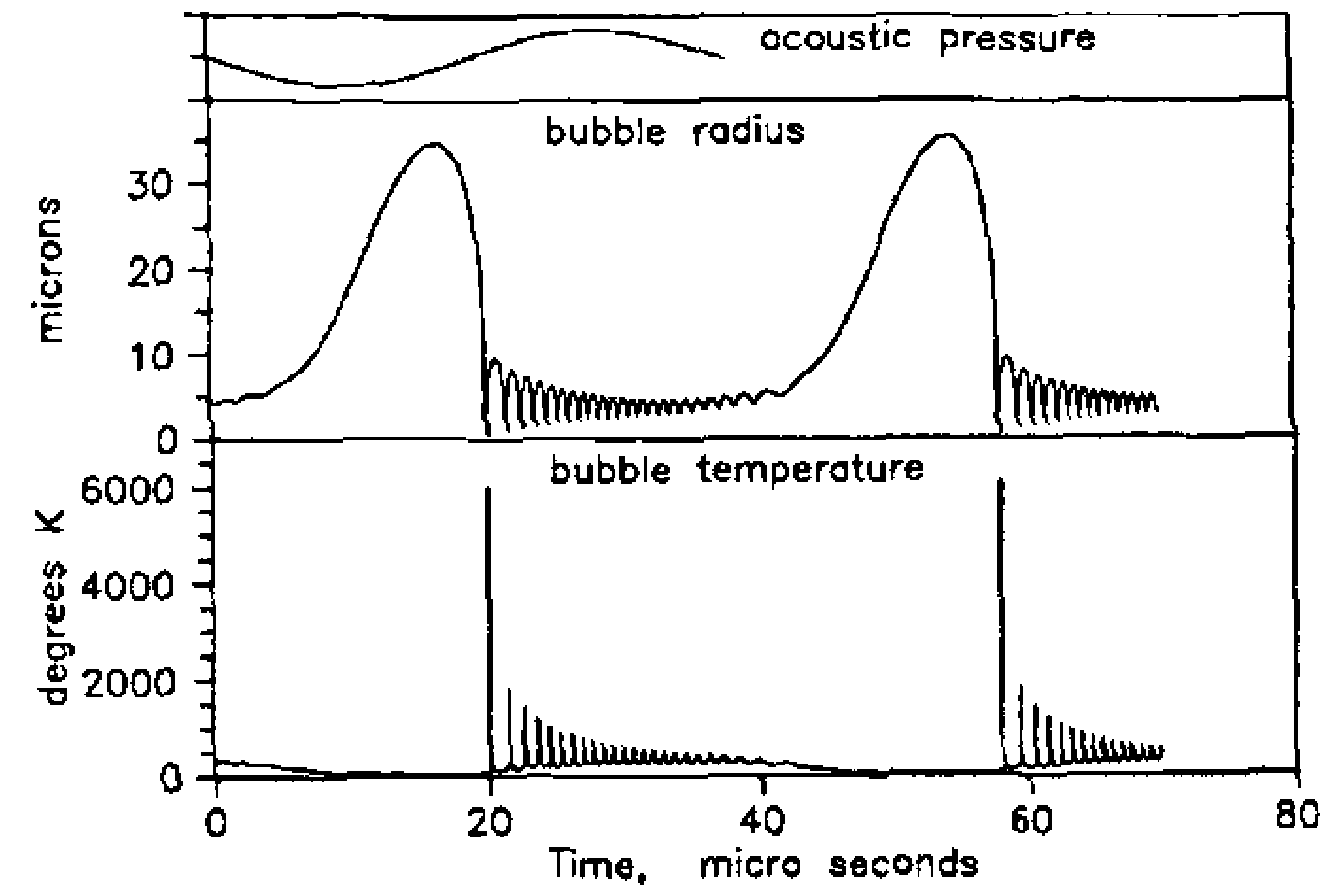

Figure 7. Typical solution to the bubble dynamics equation for the conditions of stable SL. Initial bubble radius is 4.5 µm. The acoustic pressure amplitude and frequency are 1.325 bar and 26.5 kHz, respectively. The latter corresponds to an acoustic period of 37.74 µs.

Table 1. Magnitude of bubble interior parameters obtained from the solution to the bubble dynamics equation with the use of van der Waal's equation of state

	Gas phase properties				
State of bubble motion	R (µm)	T (K)	P (bar)	ρ (kg/m^3)	$\dot{R}_{max}$ (m/s)
Initial	4.5	300	1.032	1.2	-
Point of maximum radius	35.1	25.5	1.8×10^{-4}	2.52×10^{-3}	-
Point of minimum radius	0.583	6065	3.6×10^{4}	552	-
During first collapse	-	-	-	-	1365

high temperatures and the accompanying SL. Therefore, it is clear from the above discussion that the high temperatures inside the bubble can originate from simple adiabatic compression (Figure 7) or by formation of a spherical shock and its focusing and rebounding from the centre. To explain SBSL, one must then examine the possible mechanisms for light emission. In the past, several have been suggested[2], but the ones relevant in the present context are the following: (a) chemiluminescence, (b) bremsstrahlung and (c) black-body radiation.

Chemiluminescence in general could involve the formation of electronically excited molecules and/or radicals with radiative transition to a lower state or radiative recombination. For example, Saksena and Nyborg[3] considered the radiative recombination of OH and H radicals formed from H_2O at high temperatures. In addition, Sehgal *et al.*[21] suggested that SL spectra from a multibubble cavitation field could consist of bands corresponding to transitions of excited OH and H_2O molecules. Only when the gas is active could band systems like Schuman–Runge for O_2 be important. There is a fundamental difficulty in ascribing chemiluminescence to SBSL because the radiative life times for fully allowed electronic transitions are[22] of the order of 10^{-8}–10^{-7} s, whereas the flash duration of SBSL has been estimated to be 50 ps. Bremsstrahlung is associated with radiation from a free electron in an ionized gas, which gets slowed down as it approaches the field of an ion or a neutral atom. This emission is continuum and has been used[20] very recently to predict quantitatively the light intensity during SBSL. Blackbody radiation follows the standard Planck distribution.

Therefore, theoretical considerations of SBSL would require a proper modelling of (i) the bubble dynamics process, (ii) the dynamics of the gas within the bubble and (iii) the mechanism of light emission. Among these, at present the first one can be handled with some confidence, but there are a lot of uncertainties about the latter two.

Concluding remarks

Single-bubble sonoluminescence (SBSL) is a controlled physical process with apparently continuous light emission consisting of highly synchronous flashes of light having an estimated duration of 50 ps and measured peak power of > 30 mW. The excellent timing properties of the flashes have made SBSL a valuable tool in the study of the behaviour of a sonoluminescing gas bubble. In addition, the short duration of the flashes could be useful in determining the rise time characteristics of photomultiplier-tube-based electronic systems. Therefore, there is a potential in developing light-emitting bubble as a picosecond light source with practical utility. There is a need to identify the mechanism of light emission since it will have a direct bearing on the estimated temperatures reached within a light-emitting bubble. Black-body fit to the emission spectra indicate temperature estimates approaching 10^5 K under some conditions. Whether higher temperatures and powers can be achieved may crucially depend on the possibility of stabilizing a bubble with larger initial size and at higher acoustic pressure amplitudes than possible now.

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