Sonoluminescence and bubble fusion*

Vijay H. Arakeri
Department of Mechanical Engineering, Indian Institute of Science, Bangalore 560 012, India

Sonoluminescence (SL), the phenomenon of light emission from nonlinear motion of a gas bubble, involves an extreme degree of energy focusing. The conditions within the bubble during the last stages of the nearly catastrophic implosion are thought to parallel the efforts aimed at developing inertial confinement fusion. A limited review on the topic of SL and its possible connection to bubble nuclear fusion is presented here. The emphasis is on looking for a link between the various forms of SL observed and the severity of bubble collapse or implosion. A simple energy analysis is also presented to enable the search for an appropriate parameter space and an experimental technique for achieving energy densities required for triggering fusion reactions within the bubble.

THE motion of a sonoluminescing bubble involves the growth of a nucleus to a maximum size during the low-pressure phase of an imposed ultrasonic sound field followed by a rather violent collapse once the imposed pressure recovers to the ambient value or above. It is now well-established that light emission coincides with the last stages of bubble collapse and is associated with high temperatures and pressures generated within the bubble during this time. This is commonly termed as the hot spot model for sonoluminescence (SL). The above noted transient motion of a bubble repeats itself every cycle, and SL thus consists of extremely short duration flashes of light which are synchronous with the drive frequency of the sound field. SL is a fascinating phenomenon since it involves an extreme degree of energy focusing; basically, during this process, low-intensity sound energy in the liquid medium is converted to light energy involving energetic photons. The estimated level of energy focusing is twelve orders of magnitude[1]. It is natural, then, to ask whether bubble nuclear fusion is possible through this mechanism (see ref. 2). In this article, some aspects of SL in the context of the above question will be examined, and a perspective on bubble fusion also will be given. It should be emphasized that the present perspective is based on simple energy considerations and does not involve any detailed computations of the type, for example, contained in Moss *et al*.[3]. However, the present analysis does serve the purpose of identifying important gross parameters of the problem and may prove to be helpful in designing future experiments.

*Dedicated to Prof. S. Ramaseshan on his 80th birthday.
e-mail: vijay@mecheng.iisc.ernet.in

Various forms of SL

In view of recent developments, SL can broadly be classified into two types, namely multi-bubble sonoluminescence (MBSL) and single-bubble sonoluminescence (SBSL). MBSL, which involves light emission from a bubble field, has been known and investigated since 1934, and several relatively recent review articles on the topic are available[4,5]. On the other hand, SBSL, which involves light emission from a single, levitated bubble in a standing-wave sound field was discovered only about a decade ago[6,7]. This remarkable finding has paved the way for renewed interest in the general subject of SL and related topics like bubble dynamics. Since SBSL is such a controlled phenomenon, the understanding generated about its physical aspects is both deep and extensive. As indicated in recent reviews[8–10], many aspects of SBSL are now well-understood. One of the models[11], which is free of adjustable parameters, is capable of explaining many of the experimental observations; however, some key aspects remain unresolved. One is extreme sensitivity of the phenomenon to variations in experimental parameters; for example, the number of photons emitted per flash is observed[12] to increase by a factor of 100 with a decrease in the ambient temperature from about 30 to about 5°C. There are questions as to the exact mechanism of light emission and energy focusing, in particular, as to whether it is due to simple adiabatic compression[11] or involves the formation of shocks[13–15]. It is beyond the scope of the present article to go into any details about these matters (the interested reader can consult the review articles cited earlier); however, at this point it is worthwhile to mention the motivation (other than bubble fusion) for studying MBSL and SBSL.

The MBSL spectra are considered to be useful signatures of the extreme conditions, in terms of temperatures and pressures, reached within the nonlinearly oscillating bubble fields. Such fields, commonly known as acoustic cavitation, are known to be responsible for many of the chemical, physical and biological effects due to high-intensity ultrasound[16,17]; one recent application is synthesis of nano-particles[18]. On the other hand, SBSL being such a controlled phenomenon, can be considered to be a micro-laboratory for studying such diverse topics as high energy physics and chemistry, nonlinear dynamics, nonequilibrium thermodynamics and transport processes, etc. One immediate application has been associated with SBSL flashes being of extreme short duration of the order of hundred picoseconds[19,20]. Thus, light from a SBSL flash has been used to determine the rise time characteristics of photomultiplier-based instrumentation[21,22]. Similarly, the shock wave emitted in the liquid medium during the last stages of bubble collapse associated with SBSL[23] can find application in determining the response characteristics of hydrophones.

Now we discuss some results from our own studies of SL[22,24] which have enabled us to infer the existence of different types of bubble motions. Long-exposure photographs of two different forms of MBSL and one form of SBSL are presented in Figure 1. A medium was found from which a nearly pure line emission (in this case golden yellow sodium D line) from MBSL was possible. The spectrum of this emission shown in Figure 2 confirms what is observed in Figure 1 *a* – that the dominant emission is the sodium resonance radiation near 589 nm. The doublet was not resolved due to the unusually large broadening, which from Figure 3 is estimated to be about 4 nm. From the same figure, it is apparent that there is asymmetry towards the red in the profile. Both these features are indicators of high-density environment at the instant of light emission[25]. One of our key findings was that the optical pulse widths of MBSL flashes in the form of sodium resonance radiation were of the order of tens of nanoseconds[22]. This was at first quite surprising, since these were considerably longer than the previously reported pulse widths for both MBSL[21] and SBSL[19,20]. An explanation (which we believe is convincing) for this observation was provided in Giri and Arakeri[22]. Further support was found from modelling sodium emission using bubble dynamics formulation developed by Kamath *et al.*[26], and showing that the synthetically generated optical pulse shape agreed well with the measured one[24]. Thus, we demonstrated the existence of synchronous nanosecond sonoluminescence. This is in contrast to SBSL, which in one study[1] has been characterized as synchronous picosecond sonoluminescence. We will return to indicate the implication of this finding in the next section.

To continue the discussion on various forms of SL, it is worthwhile to point out that the spectra of MBSL as depicted in Figure 1 *b* and SBSL are both broadband. The MBSL spectrum extends from about 350 nm to in excess of 700 nm and possesses a broad peak near about 450 nm[22]. The SBSL spectrum covers a wider range but does not show a discernible peak; its intensity continues to increase even at wavelengths of 200 nm where absorption in water (friendliest of fluids for establishing SBSL) becomes significant[9]. A blackbody fit to a spectrum[27] indicates bubble temperatures of the order of 25,000 K. All the forms of SL shown in Figure 1 are visible to the naked eye in a darkened room. In the case of SBSL, where bubble sizes at the instant of light emission are estimated to be of the order of one micron[9], the radiation is visible at a distance of almost one million times the source size. Therefore, it is not surprising that one description for SBSL has been ‘star in a jar’.

Implications on the existence of different types of bubble motion

In the previous section, we have presented some evidence for the existence of different forms of SL characterized

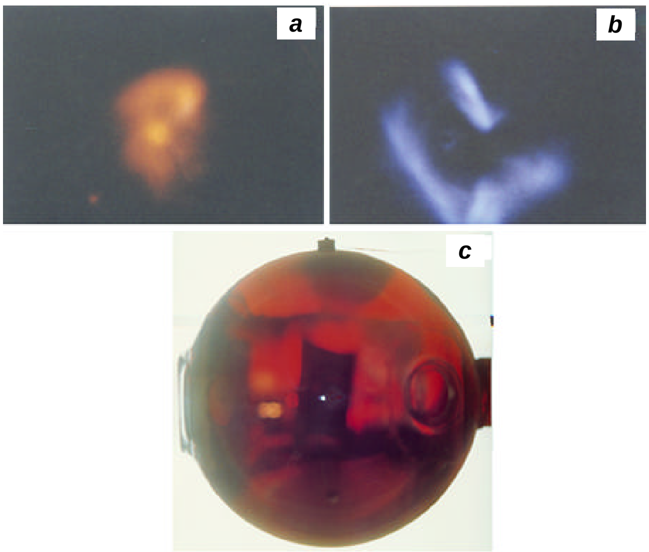

Figure 1. ***a***, MBSL from an argon-saturated 1 N sodium chloride–ehylene glycol solution. The emission, golden yellow in appearance, is narrowband sodium D line resonance radiation. ***b***, MBSL from air-saturated ethylene glycol sample. The emission, bluish in appearance, is broadband and the source of emission is unknown (for further details see ref. 22). ***c***, SBSL (bright spot at the centre) from slightly degassed water contained in a spherical flask[41].

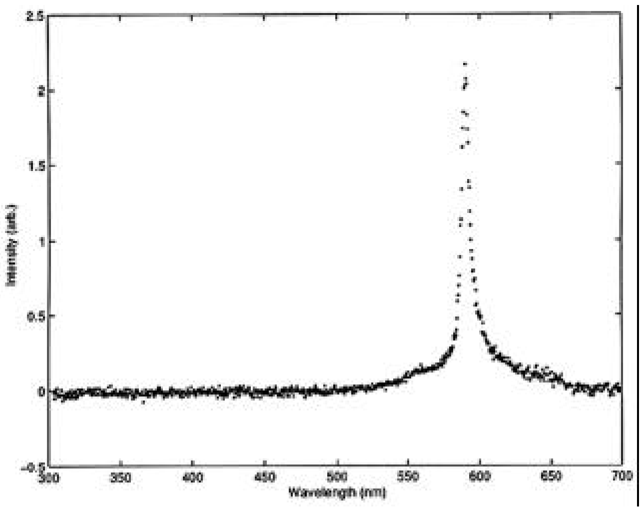

Figure 2. Low-resolution (3 nm FWHM) spectrum of MBSL in the form of sodium resonance radiation (see Figure 1 *a*). After Giri and Arakeri[22].

by varying spectral distributions and timescales. The motion of a bubble under the influence of high-intensity ultrasound is known to depend sensitively on some parameters like the drive amplitude and frequency[28]. Therefore, it may be reasonable to ascribe different forms of SL to varying severity of bubble implosion. Here, we examine this possible connection further. First, a summary of some of the physical characteristics of different forms of SL is provided in Table 1. The inferred type of bubble motion with soft collapse and hard collapse, as indicated in Table 1, was suggested by Giri and Arakeri[22] to explain the vastly different timing characteristics of MBSL flashes. The SBSL flash widths are even shorter than those associated with MBSL flashes, and this may indicate that during SBSL the bubble collapse is even more severe and hence termed here as super collapse. In Table 1, we hypothesize on the existence of bubble motion with hyper collapse, and this may lead to truly extreme temperatures within the bubble. We follow-up on this in the next section.

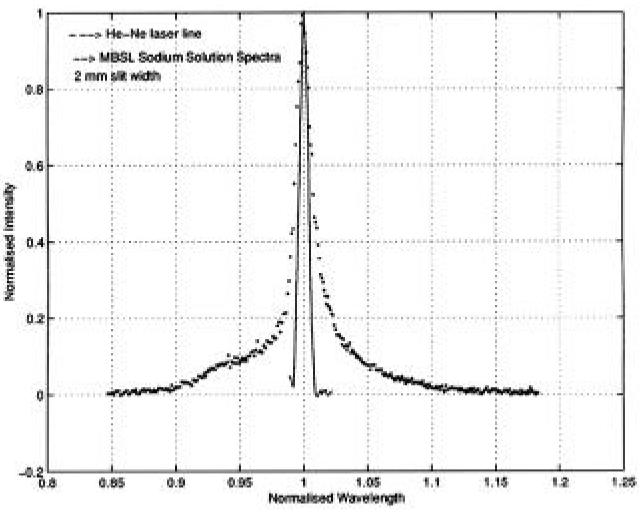

Figure 3. Comparison of the normalized spectrum line profiles of MBSL in the form of sodium resonance radiation (·····) and that of a helium-neon laser (——). These were recorded with a scanning monochromator by shining attenuated laser light on a sonoluminescing bubble field.

Perspective on bubble fusion

Prospects for initiation of thermonuclear fusion reactions within a sonoluminescing bubble were suggested when theoretical simulations of the SBSL phenomenon by Wu and Roberts[13] showed the existence of maximum bubble temperatures of the order of 10^8 K! These extreme temperatures were limited to a small region of the bubble interior and were made possible by the launch of a shock wave within the already compressed gas. The shock focuses as it approaches the bubble centre and doubles its strength when reflected from the origin. In an another study, Moss *et al.*[3] showed that hydrodynamic simulations of a collapsing bubble containing D_2 and D_2O vapour provide the possibility for a small number of thermonuclear D–D fusion reactions in the bubble. A more recent and complete shock-wave model used to compute the optical emissions from a single sonoluminescing bubble is that due to Moss *et al.*[30]. Even though, the agreement with general experimental observations is good, it has not been possible to verify experimentally the involvement of shocks in any SL process. The potential role of shocks in the SL process will remain a controversial topic, in particular, since good agreement with the same dataset as used by Moss *et al.*[30] has also been possible using a model that assumes energy focusing by simple adiabatic compression[11].

In a stunning development in 2002, Taleyarkhan *et al.*[31] reported detection of nuclear products from a sonoluminescing bubble field. However, repetition of their work by Shapira and Saltmarsh[32] has cast some doubts on the original interpretations by Taleyarkhan *et al.*[31]. We do not want to dwell further on this issue, except to state

Table 1. Summary of physical characteristics of some forms of MBSL and one form of SBSL. Also indicated are inferred types of bubble motion. Numerical values shown are typical

Type of SL	Medium	Spectrum character	Optical pulse width of SL flash	Number of photons per flash	Power per flash	Bubble temperature (K)	Acoust. ampl. (drive level)	Exp. ratio R_{max}/R_o	Inferred type of bubble motion
MBSL	Argon-saturated NaCl–ethylene glycol solution	Broadened asymmetric sodium D line emission	50 ns	10^6	7 µw	3×10^3 Comp. est.[24]	1 bar (low)	2.2 Est.[24]	*Soft collapse*
MBSL	Air-saturated ethylene glycol	Broadband extending from 350 to ~ 700 nm with a peak at 450 nm	1 ns	5×10^5	1 mw	5×10^3 Est. from Cr spectra[30]	3 bar (high)	5 Probable	*Hard collapse*
SBSL	Degassed water	Broadband extending from 200 to > 700 nm; No peak	100 ps	5×10^5	8 mw	2.5×10^4 Est. from blackbody fit to a spectrum[27]	1.4 bar (medium)	10 From experiments[9]	*Super collapse*
SBL (single bubble luminescence)	Degassed low vapour-pressure liquid	Broadband	< 1 ns	?	?	$> 10^6$ Desired	15 bar (high)	100 (see text)	*Hyper collapse*

that the approach taken by Taleyarkhan *et al*.[31] was interesting. They did not take the route of SBSL, where a seeded nucleus is levitated as a bubble at the pressure node of a standing-wave acoustic field and then the acoustic pressure is gradually increased until the bubble starts to glow and become brighter. However, this process cannot be continued indefinitely; at critical pressure amplitude, P_{Ac}, the bubble is destroyed. From several experimental studies the magnitude of P_{Ac} is found to be about 1.35 bar, with the liquid ambient pressure being 1 bar. Therefore, a limit to maximum energy focusing that can be achieved through this approach exists. Even though, as indicated earlier, the limit (~ 10^{12}) is quite impressive, it does not seem to be sufficient for initiation of fusion reactions within the bubble. In terms of bubble dynamics parameters, the above limit can be expressed in an alternate form as a limit on the maximum expansion (characterized by R_{max}/R_o, where R_{max} denotes the maximum radius attained by the bubble and R_o is its radius at the start of the growth phase) that can be achieved through the SBSL route. This follows since R_{max}/R_o is known to depend strongly on P_A, with other parameters held constant[28]. The maximum expansion is also known to be a critical parameter in determining the severity of bubble collapse or implosion. With R_{max}/R_o near 10, the maximum inward velocity is found to be supersonic with respect to the gas-phase speed of sound[9]; with R_{max}/R_o about 2.5, the inward velocity is predicted to be only about 35 m/s, being a small fraction of the speed of sound[24]. The expansion ratios of 10 and 2.5 are typical of those associated with SBSL and MBSL in the form of sodium resonance radiation. Hence, our terming of bubble motions in Table 1 with respect to SBSL as super collapse and those with respect to MBSL as soft collapse may be appropriate.

In order to achieve an expansion ratio higher than 10, Taleyarkhan *et al*.[31] took partially degassed deuterated acetone (C_3D_6O) in an acoustic levitation cell and subjected the sample to acoustic pressure amplitudes of the order of 15 bars; the nuclei were seeded by dumping high-energy neutrons (14 MeV) to initiate bubble activity or acoustic cavitation. (Recall that in SBSL experiments, a nucleus is first seeded and then acoustic pressure is gradually increased until a self-limiting value of about 1.35 bars is reached). By following the above procedure, Taleyarkhan *et al*.[31] claimed to have achieved an expansion ratio of the order of 10^5; however, no direct proof was provided. In essence, it can be stated that on the basis of both experimental evidence and theoretical considerations, the extent of energy focusing is primarily determined by the magnitude of R_{max}/R_o. It turns out that this parameter comes out naturally, if one considers the energetics of a cavitation bubble. As indicated earlier, the transient motion of a sonoluminescing or cavitation bubble involves a growth phase from an initial radius R_o to a maximum radius R_{max} under the influence of applied low pressure (in most cases it is actually negative or tensile). During this process it acquires potential energy, which to a good approximation can be expressed as,

$$\mathrm{PE} = P_c\left(\tfrac{4}{3}pR_{max}^3\right). \qquad (1)$$

In the above, P_c is the ambient pressure at the beginning of collapse and hence commonly termed as collapse pressure. It is the above potential energy, when deposited on few atoms or molecules of gas present in the bubble, which results in SL. Therefore, it is important to consider the energy density, that is the potential energy per atom or molecule of gas involved. Assuming certain equilibrium conditions for the bubble contents when its radius is R_o, the number of atoms or molecules, N_o, of gas in the bubble can be estimated. As expected, it turns out that the value of N_o is proportional to the bubble volume given by $(4/3)\,pR_o^3$. Using this information, the energy density $E_d = \mathrm{PE}/N_o$ in units of eV per atom or molecule (note: 1 J = 6.242 × 10^{18} eV) works out to be:

$$E_d = 0.025(P_c/P_o)\,(R_{max}^3/R_0^3). \qquad (2)$$

The constant value 0.025 in eq. (2) is approximate and includes the values of Avagadro number and universal gas constant. The collapse pressure P_c can be taken equal to the ambient liquid pressure P_o, unless special efforts are made to spike the acoustic wave with a pressure pulse at the right instant, and efforts in this direction are underway[33]. Taking $P_c = P_o$,

$$E_d = 0.025(R_{max}^3/R_0^3) \qquad (3)$$

is then only a function of the expansion ratio. For example, with an expansion ratio of 10 (typical of SBSL phenomenon[9]), the energy density is 25 eV per atom or molecule. The expressions given above for E_d do not include the possible effects due to vapourization; the tacit assumption is that any vapour formed during the growth phase will condense out during the collapse phase. However, due to the nonlinear nature of the bubble motion, this need not be the case. Recent computations[34] show that some vapour does escape condensation and hence the bubble potential energy now gets distributed over both gas atoms or molecules and the remaining vapour molecules. By assuming that a fraction of the vapour molecules present in the bubble at its maximum radius escapes condensation, it is straightforward to show that the modified expression for E_d becomes:

$$E_d = 0.025(R_{max}^3/R_0^3)\left[\frac{1}{1+k\left(\frac{P_v}{P_o}\right)\left(\frac{R_{max}^3}{R_0^3}\right)}\right]. \qquad (4)$$

Here k is the fraction of vapour escaping condensation and P_v is the vapour pressure of the host liquid.

In Figure 4, a plot of E_d versus R_{max}/R_o is presented for the following three cases: (a) $k = 0$; (b) $k = 0.025$ and

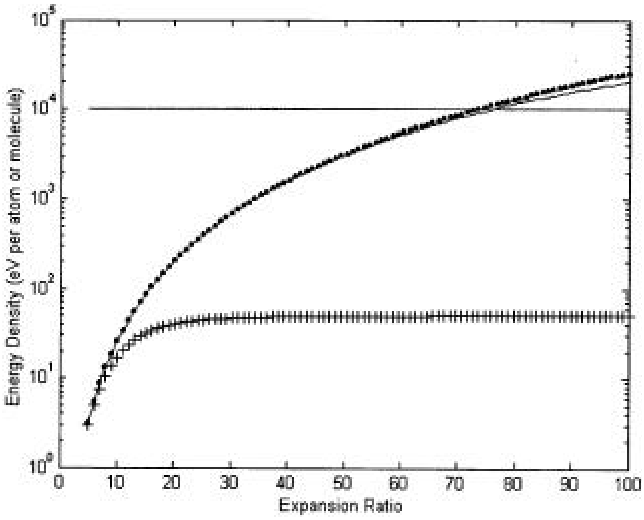

Figure 4. Bubble energy density versus expansion ratio, R_{max}/R_o for various cases. · · · · ·, $k = 0$; ++++, $k = 0.025$ and $P_v / P_o = 0.02$; —— $k = 0.025$ and $P_v / P_o = 10^{-5}$. Also shown is constant value for fusion threshold, 10^4 eV per atom or molecule[37] (note: 1 eV = 11,600 K).

$P_v/P_o = 0.02$ (typical of water near room temperature and $P_o = 1$ bar); and (c) $k = 0.025$ and $P_v/P_o = 10^{-5}$ (typical of fluids like ethylene glycol and $P_o = 1$ bar). The value of $k = 0.025$ is chosen on the basis of results due to Storey and Szeri[34]. Some interesting features are apparent from the results shown in Figure 4. With $k = 0$ there is no limit on the value of E_d and its value increases monotonically with R_{max}/R_o. On the other hand, with $k \neq 0$, there is an asymptotic limit for E_d given by

$$\left(\frac{0.025}{k}\right)\left(\frac{P_o}{P_v}\right) \quad \text{as} \quad R_{max}/R_o \to \infty.$$

The limit for E_d can be taken to strongly depend on the value of P_v, since P_o, is generally 1 bar (in principle, P_o can be increased but there are other difficulties like requirement of increased drive acoustic pressure amplitude; hence, in our discussion here we will assume $P_o = 1$ bar). For example, with a fluid like water the limit is about 50 eV per atom or molecule. It is worthwhile to point out that some limitations on upscaling of SL by working with standard SBSL apparatus but with reduced acoustic drive frequencies has been noted by Toegel *et al.*[35]. This limitation was not predicted by computation[36] and has been ascribed to the effect of water vapour.

Also shown in Figure 4 is the value of $E_d = 10^4$ eV per atom, that is taken to be a representative value for fusion initiation[37]. In the case with $k = 0$, the required energy density seems possible with expansion ratio of about 75; it should be noted that this is an order of magnitude higher than what has been possible with the SBSL apparatus. The plot for the low vapour-pressure fluid in Figure 4 is nearly coincident with the plot for the case of $k = 0$, and similar conclusion as reached above is applicable. It is important to note that the energy density possible with a fluid like water is predicted to be well below 10^4 eV per atom.

Concluding remarks

The bubble potential energy available at its maximum radius gets partitioned only partially to the bubble contents and the remaining, to a large extent[38], goes out in the form of acoustic energy in the liquid. To compute the details of partitioning of energy into various components will again require solution to the complete governing hydrodynamic equations. Similarly, the expressions given here for energy densities are average values and do not include the possibility for sharp gradients within the bubble; such gradients are likely to exist, for example, with formation of shocks. In such cases, the expression for the potential energy remains the same; but most of the fraction of energy going into the bubble will get distributed over a smaller number of atoms or molecules present in the sharp gradient region. The effects due to sharp gradients and neglect of any efficiencies of the overall implosion process could to a certain extent compensate each other. The present analysis based on average values does serve a useful purpose. It appears that working with a fluid like water and driving a seeded bubble with high-intensity ultrasound (as, for example, done in SBSL experiments) may not be the ideal way for achieving extreme energy densities of the order required for initiating fusion reactions within the bubble. Also, the required energy densities are unlikely to be possible with multi-bubble cavitation field, since under these conditions the expansion is limited by bubble interference effects[39] and loss of bubble stability[40]. If the implosion is not spherically symmetric it will not only reduce the energy focusing, but there is also the possibility for injection of some fluid into the bubble leading to effects similar to vapour trapping. Therefore, development of an experimental configuration which can create an isolated bubble in a low vapour-pressure fluid with large expansion ratio (~ 100), but with nearly spherically symmetric implosion, is what will be required to realize conditions close to those needed for bubble fusion.

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