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Physics Letters A 372 (2008) 3496–3500
PHYSICS LETTERS A
www.elsevier.com/locate/pla

Sonoluminescence: Two sources of light

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Received 15 October 2007; accepted 3 December 2007

Available online 11 January 2008

Communicated by V.M. Agranovich

Abstract

The model of sonoluminescence with two sources of light is presented. The first of them is gas inside the bubble heated during collapse. The second source (main) is plasma in the breakdown channel in liquid.

PACS: 78.60.Mq

Keywords: Sonoluminescence; Model; Two sources of light

1. Introduction

Sonoluminescence (SL) is defined as the pulse light emission from collapsing gas bubbles in liquid induced by insonation with an acoustic field [1–3]. The two types of SL are known: a multi-bubble (MBSL) type and a single-bubble (SBSL) one. In case of MBSL the light emitter is a system of bubbles (density of 10^3–10^5 cm^{-3} [2]); in the second case it is one pulsating bubble created by special equipment [1,3]. In most SBSL experiments the acoustic wave frequencies f_a range from 10 to 50 kHz [1,3]; in some works $f_a = 1$ MHz [4]. According to [2], diapason f_a for MBSL is 1 Hz–1.5 MHz.

Dynamics of the SBSL bubble is strongly nonlinear. During each oscillation period there are several stages [1]: comparatively slow bubble expansion as the reaction for sound field rarefaction (~ 15 μs)[1]; growth of bubble up to its maximum radius R_m (~ 5 μs); collapse to its minimum radius R_c (time of collapse from $0.5R_m$ to R_c is about 0.5 μs) and oscillation of bubble around its ambient (undriven) radius R_0. Values of radii are: $R_c \approx 0.5$ μm, $R_0 \approx 5$ μm, $R_m \approx 50$ μm. At the final stage of collapse $R_0 \to R_c$ (5–10 ns) it occurs light flash generating and a shock wave appears in liquid [1,3,5].

The main question of SL is the mechanism of light emission. Since 1930, when sonoluminescence was discovered, different models have been offered. Almost all light-emission mechanisms discussed since then can be classified into one of two groups: thermal or electrical processes [1–3].

For all thermal models the light emitter is the gas inside the bubble heated during the collapse. According to calculations, the temperature of the gas can attain to 5×10^3–10^4 K for SBSL [1,3] and to 3×10^3–6×10^3 K for MBSL [6] when the adiabatic heating takes place. Some researchers consider SBSL situation, when a shock wave is initiated in the gas during the final stage of bubble collapse. Such a shock wave heats the gas to very high temperatures: $T_{\max} \approx 10^5$–10^8 K [1,7,8]. Some theories have explained the emission of photons from the high temperature zone by means of a blackbody model or a thermal volume emitter model [1,3]. Other theories have used the mechanism of thermal bremsstrahlung [1,3,4].

Electrical theories place the location of the emission in the gas inside the bubble or in the liquid. Some of them are based on the idea of the asymmetric collapse of the bubble. Such a collapse initiates water or gas jets. Authors of [9] have speculated on sparklike discharges around water jets invading a bubble. According to [10] the light is emitted due to fractoluminescence when the collision of the gas jets with the bubble wall initiates "cracking" of water. Models [9,10] are developed for SBSL only.

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[1] This example is for $f_a = 26$ kHz [1].

0375-9601/$ – see front matter
doi:10.1016/j.physleta.2007.12.060

Presented model of MBSL in [2] is based on the well-known physical effect of double electric layer (spontaneous polarization) which appears in liquid near to a liquid–gas boundary surface [11]. Deformation of this layer under an asymmetric collapse generates an electric field inside the bubble and an electric breakdown might take place under certain conditions. The channel of breakdown in gas is the light emitter. Model [2] predicts increasing of intensity of light emission as the asymmetry increases.

Thus, there are two groups of SL models: thermal and electrical ones. Every model can explain some experimental facts, but the mechanism of SL is not quite clear nowadays [1–3]. Such experimental results as a very high efficiency of water for SBSL experiments in comparison to other liquids [1,3], a strong sensitivity of SL to the temperature of liquid [1], the great influence of surface-active substances and noble gases on light intensity [1–3], the presence of hard ultraviolet in the spectra of SBSL [1] and the increase of its intensity as frequency of driving sound increases [4], the presence of line emission in SBSL and MBSL (lines of liquid molecules fragments and metals (for MBSL only)) at low forcing pressures and their absence at higher ones [1–3,12–14], absence of any lines in spectra originated by gas atoms and molecules [1–3], weak directional asymmetry of SBSL radiation [15–17], high dependence of shock wave amplitude upon driving pressure [5] and the fact that the size of emitting region is less than minimum bubble radius [4,15] did not obtain a convincing explanation by the developed theories. The subject of debate is also: are SBSL and MBSL same or different physical phenomena [1–3,13]?

2. The model

In this Letter we present a model of SL based on the assumption that two sources of light exist. One of them is, well known for SL, adiabatically heated gas inside the bubble during the collapse. The second one is plasma in the channel of electric breakdown in liquid. Let us consider the possibility of formation of such kind a source for SBSL in water.

For this consideration we apply, as it used in [2], the known physical effects of electric potential jump and a double electric layer (spontaneous polarization) appearance in liquid near to the liquid–gas boundary surface [11]. The origin of these surface phenomena is bounded up with the adsorption of ions and dipole molecules. The arising effect of polarization is the strongest for polar liquids. Water is a polar liquid: the dipole moment of molecule $\mu \approx 1.8$ D ($1\ \mathrm{D} = 3.34 \times 10^{-30}$ C m) [18]. Let us suppose that spontaneous polarization is generated in spherical layer of water during a bubble expansion stage by means of some dipoles regulating. Let us also suppose that uncompensated value of polarization is equal P_0 when maximum bubble radius R_m has reached. Uncompensated polarization takes place if period of sound wave $T_a \leqslant 2$–3τ ($\tau = \varepsilon\varepsilon_0\rho_0$, where ε is dielectric permittivity, ε_0—electric constant, ρ_0—specific resistance). For pure water, which is used generally for SBSL experiments, $\rho_0 \approx 2 \times 10^7$ Ω cm [1] and $\varepsilon \approx 80$ [19]. Hence, $\tau \approx 140$ μs and such a stable effect has to take place for frequencies of forcing sound $f_a = T_a^{-1} \approx 3$–4 kHz and P_0 has to increase as f_a increases. Appearance of polarization P_0 is an equivalent to arising of electric charge with density $\sigma_0 = P_0$ on the layer boundaries. Electric field strength E inside the layer is given by the known expression:

$$E = \frac{1}{4\pi\varepsilon\varepsilon_0}\frac{q}{r^2}, \tag{1}$$

where $q = 4\pi R_m^2\sigma_0 = 4\pi R_m^2 P_0$, r—radius ($r \geqslant R_m$).

Near the layer boundary, when $r \approx R_m$, the field strength is

$$E_m = \frac{P_0}{\varepsilon\varepsilon_0}. \tag{2}$$

If $q = \text{const}$ during the bubble collapse, then for $r = R_c$ one can obtain

$$E = \frac{P_0}{\varepsilon\varepsilon_0}\left(\frac{R_m}{R_c}\right)^2. \tag{3}$$

As far as $R_m/R_c \approx 10^2$ usually for SBSL, we obtain

$$\frac{E_c}{E_m} = \left(\frac{R_m}{R_c}\right)^2 \approx 10^4. \tag{4}$$

Let us determine value P_0 which is sufficient for electric breakdown. The electric strength of technical pure water is $E_{\mathrm{st}} \approx 0.7 \times 10^7$ V/m [20]. Applying expression (3) for $E_c = E_{\mathrm{st}}$ we have: $P_c = \varepsilon\varepsilon_0 E_{\mathrm{st}} = 0.5$ μC/cm^2 and, consequently, $P_0 = 0.5 \times 10^{-4}$ μC/cm^2. The maximum possible value of polarization for water is $P_{\max} \approx 20$ μC/cm^2, when the orientation of all molecular dipoles is the same. So that $P_0/P_{\max} = 2.5 \times 10^{-6}$. This estimation shows the possibility of high strength electric field arising in water. During the final stage of collapse (about 100 ps before maximum compression) the gas undergoes ionization and releases free electrons. Some of there electrons penetrate into the thin spherical layer of water. The strong electric field $E \geqslant E_{\mathrm{st}}$ and free electrons presence are the necessary and sufficient conditions for local electronic breakdowns in water. Plasma in the breakdown channel is the main source of light. Plasma expansion from the initial state forms a shock wave in water. The hot gas inside the bubble is the second light source. It basically forms red region of the SBSL spectrum. The presented polarization model can be employed for MBSL too. In this case the effect ought to be lower because of the collapse asymmetry. Let us discuss some not quite explained experimental results with usage of our model.

3. Experimental results

3.1. "Friendly" water

It is known that water is the most effective liquid for SBSL experiment [1,3]. In Table 1 some parameters of the liquids used for SBSL investigation are presented. There are dielectric permittivity ε, specific resistance ρ_0, time constant $\tau = \varepsilon\varepsilon_0\rho_0$, dipole moment μ, surface tension Σ and dynamic viscosity η. This dataset is taken out from reference books [18–22]. It corresponds to the normal conditions.

Table 1
Parameters of the liquids

N	Liquid	ε	ρ, Ω m	τ, μs	μ, D	$\Sigma \times 10^3$, N m^{-1}	$\eta \times 10^3$, Pa s
1	Water (H_2O)	78.3	4.4×10^6* $(1–4) \times 10^4$** 2×10^5***	140	1.84	72.8	1.05
2	Heavy water (D_2O)	78.3	–	–	1.78–1.87	72.6	–
3	Ethanol (C_2H_6O)	25.2	1.6×10^5	36	1.68	22.0	1.22
4	Pentanol ($C_5H_{12}O$)	14.4	–	–	1.66	–	–
5	Propanol (C_3H_8O)	–	2.9×10^3	–	1.78	26.0	2.2
6	Butanol ($C_4H_{10}O$)	17.7	10^5	16	1.65	–	2.95
7	Adipnitrile ($C_6H_8N_2$)	–	–	–	2.17	–	–
8	Dodecane ($C_{12}H_{26}$)	2.0	–	–	0	25.5	1.49
9	Silicone oil	2.3	–	–	0	–	–

* Distilled in vacuum; ** Distilled; *** Used for experiments [1].

In comparison with other liquids, water has the greatest values of surface tension Σ and τ. The first parameter is important to form a liquid–gas boundary surface, the second one estimates an uncompensated value of polarization. These are very important parameters for SL, but it is necessary to emphasize that for our model the liquid–gas system is the most important because both of these media take part in formation of the boundary surface.

3.2. Surface—active substances

According to [1,3], intensity of SBSL emission strongly depends on some substances added in liquid or gas. Small amount (some few ppm) of ethanol [3] or butanol [1] added to water quench the emission sharply in the system of water–air, while the bubble is still oscillating in the stable fashion. The light emission becomes weaker as compared with the system of ethanol(butanol)–air. On the base of a polarization model, this effect can be explained in view of the fact that ethanol and butanol are surface-active substances. Such substances tend to be accumulated on the bubble surface and decrease the surface tension. It results in decreasing polarization P_0 and, consequently, diminution of the light emission.

3.3. Noble gases

Apparently, the influence of noble gases on the light emission can be explained in the similar manner. It was showed [1] that stable and visible SBSL flashes were not observed in experiments with pure N_2, O_2 and their 80/20 mixture dissolved in water. For the system of water–$N_2(O_2)$ the light intensity is 0.2–0.3 (0.3–0.4) in comparison with the system of water–air. Adding inert gases results in stability and intensification of SL. For example, adding He, Ar and Xe to N_2 results in increase of light intensity when concentration of noble gas does not exceed 1%. Maximum intensity is 0.6 (He), 1.0 (Ar) and 1.2 (Xe) in comparison with the system of water–air [1]. While concentration of noble gas has increased over 1%, the intensity has been decreasing slowly to 0.3 (He, Ar) and 1.1 (Xe) for pure noble gases [1]. Thus, the mixture of air and 1% of noble gas is more effective then the pure noble gas. In our opinion this fact points out that noble gases partake in boundary surface and polarization in liquid.

3.4. Liquid temperature and shock wave

There is one general fact for all SBSL experiments: intensity of the bubble light emission increases as temperature of the liquid falls [1]. For example, the emission power in the system of water–air rises by factor of $\sim 10^2$ when the temperature of water decreases from 40 °C to −6 °C [1]; in the system of water–helium that parameter rises by factor of 16 as temperature falls from 25 °C to 0 °C (Fig. 53 in Ref. [1]). A possible explanation of this effect on basis of a polarization model follows bellow.

Viscosity and surface tension for liquid rise as temperature falls. Water viscosity, for example, is two times as high at 0 °C then at 25 °C [19]. Under such change of temperature the surface tension increases by factor of 1.05 [21]. Such change of the parameters must promote the increase of the polarization P_0 and, consequently, the increase of the emission intensity. Rising of the intensity can be very strong in view of the fact that the heightened driving pressure is used to achieve the maximum effect [1]. The latter statement based on the results of investigations of shock waves which are generated in water at the final stage of the bubble collapse. Pecha and Compf showed [5] that shock wave amplitude p_{sw} depends strongly upon driving pressure p_a: change of p_a from 1.28 to 1.39 bar results in increasing p_{sw} from 38 to 60 kbar.

In our model a shock wave source is the channel of breakdown in liquid. That is why it is reasonable to suppose that in such kind of source the energy output and, consequently, the shock wave amplitude is proportional to the density of electric field energy w. The latter parameter is proportional to the electric field strength in power two. From expression (3) we have

$$w \sim \left(\frac{R_m}{R_c}\right)^4. \tag{5}$$

As the bubble dynamics is nonlinear let us assume that $R_m/R_c \sim p_a^{\alpha}$ and get next expression for shock wave amplitudes ratio.

$$\frac{p_{swm}}{p_{swn}} \sim \left(\frac{p_{am}}{p_{an}}\right)^{4\alpha} \cong k_{mn}, \tag{6}$$

where m and n are experiment numbers.

In Table 2 the results [5] and calculated coefficients k_{nm} are presented. These data show that $p_{swm}/p_{swn} \sim k_{mn}^5$ ($\alpha \approx 1.25$) and, therefore, explain the strong dependence of a shock wave amplitude from driving pressure.

Table 2
Shock wave parameters

Number of experiment	p_a, bar	p_{sw}, kbar	k_{mn}	p_{swm}/p_{swn}	k^4_{mn}	k^5_{mn}	k^6_{mn}
1	1.28	38	1.04 (21)	1.18 (21)	1.17	1.22	1.27
2	1.33	45	1.05 (32)	1.33 (32)	1.22	1.28	1.34
3	1.39	60	1.09 (31)	1.58 (31)	1.41	1.54	1.68

3.5. Sonoluminescence spectra

One of the main characteristic of the light source is its radiation spectrum. Till recently MBSL and SBSL are regarded as different physical phenomena because their spectra are different [1,3,13]. The spectrum of MBSL contains spectral lines which originate from neutral hydroxyl radicals or other molecular species presented in liquid. In contrast to this, SBSL shows a smooth continuum without line emission. Later, when line emission for SBSL was discovered [12,14], the situation has changed cardinally and been now characterized by the following.

1. SBSL spectral density increases as wavelength decreases. At low driving pressures, right at the onset pressure for SL, the spectrum contains spectral lines originated from molecular species of liquid: radicals of OH* (water) [12] or CN* (adiponitrile) [14], for example. Young et al. [12] observed line emission in water at other wavelengths too. The origin of these lines is unclear today. The increase of driving pressure results in disappearing lines. The spectrum is getting smooth.

The spectra are significantly more ultraviolet at sound frequency 1 MHz then at $f_a \approx 10^4$ Hz [4].

2. The spectrum shapes of MBSL are analogous to SBSL ones. They contain lines originated from molecules or molecular species of liquids [1–3]. At high driving pressure the spectral lines disappear [12].

3. MBSL spectrum for system of 0.1 M water solution of NaCl(KCl)–air contains the lines of Na* (K*) and OH*. The SBSL spectrum for the same system is smooth [13]. The presence of metal lines in MBSL spectrum is unanswered question for the known models [3,13].

4. Lines originated from atoms or molecules of gas inside the bubble are absent in MBSL and SBSL spectra [1–3].

SBSL and MBSL spectra and their peculiarities have a simple explanation by means of the presented model. As it was noted earlier, the main emitter in our model is plasma being generated in the local breakdown channels in liquid. It is known that the emission spectrum of the low-temperature plasma (ionic temperature $T \leqslant 10^5$ K) is linear. That plasma is a light source for SBSL and MBSL at low driving pressure. The most probable emission mechanisms is the recombination of electron–ion and molecular radicals and relaxation of exited states of molecules and molecular radicals. According to [12] for example, the recombination of the radicals H and OH is able to create a smooth part of the MBSL spectrum. High-temperature plasma is the source of emission for bright SBSL and MBSL at high level of acoustic pressure. The main emission mechanism here is thermal bremsstrahlung.

3.6. Some other results

According to our model the main light source locates outside the bubble. Thus, the dimensions of a source are not obligatory equal to the bubble radius. This fact is a qualitative explanation of the experimental result [15–17], where it is shown that the size of the light-emitting region is smaller than the minimum bubble radius and the light emitted by a bubble has a weak directional asymmetry.

3.7. Sonofusion

We also note the use of SL high pressure and temperature to produce nuclear fusion [7,8,23–27]. Taleyarkhan et al. reported in 2002 about the observation of d–d thermonuclear reaction occurring in collapsing bubbles in deuterated acetone (C_3D_6O) [23]. The researches used a pulse of neutrons in order to seed the bubbles. This new method permits to produce stable bubbles that could expand to nearly a millimeter in radius. The experiments were conducted with driving pressure $P_a = 15$ bar, sound frequency $f_a = 20$ kHz and liquid temperature $T = 3\,^\circ$C. Measurements of neutron emission and tritium production were performed. These experiments with deuterated acetone showed the evidence of d–d fusion reaction, whereas, null results were obtained with normal acetone.

These experiments were repeated by Shapira and Saltmarsh [24] but they "find no evidence for 2.5 MeV neutron emission correlated with sonoluminescence form collapsing bubbles". However, after analyzing this work R.I. Nigmatulin et al. [25] determined, that the results of Shapira and Saltmarsh were consistent with fusion. Recently Taleyarkhan et al. [7,26], Xu and Butt [8], Forringer et al. [27] confirmed the results presented in paper [23]. In works [7,8] the physical setup was similar to the original experiment [23]. Intensity of neutron emission was (1–4) $\times 10^5$ n/s (5–20 neutrons per each sound cycle) [7]. Authors [26,27] used mixture of deuterated acetone (C_3D_6O), deuterated benzene (C_6D_6) and tetrachloroethylene (C_6Cl_4) as a liquid. Alpha particles from radioactive decay of natural uranium (dissolved in liquid uranyl nitrate) were used to nucleate bubble clusters. In this case intensity of neutrons was 5×10^3–10^4 n/s [26]. The researchers suppose that the conditions needed for the fusion are created by a spherical shock wave formed inside the collapsing bubble [7,26]. The shock wave model of supercompression of vapor bubbles was developed by R.I. Nigmatulin et al. (see for example [25,28,29]. This model describes dynamics of cavitation of bubble cluster during the acoustic cycle. Theoretical simulation [28] has predicted the experimental results with deuterated acetone [7,23]. In our view this kind of conditions have to be realized in the breakdown channels in acetone. Note, that acetone is a polar liquid, its dipole moment is $\mu \approx 2.8$ D [18]. One of the reasons of lesser neutron intensity in the experiment [26] (mixture of organic liquids) in comparison with experiment [7] (pure acetone) is probably bound up with diluting the polar acetone with non-polar benzene and tetrachloroethylene.

The presented model allows to propose other experiments for sonofusion investigation. Such SBSL experiments have to

be conducted with a system of D_2O-gas. Gas has to ensure the brightest SL in this system. The driving pressure and sound frequency have to be as high as possible, the liquid temperature—extremely low. The best liquid for sonofusion might be a mixture (50/50) of D_2O and T_2O. This mixture can be used to realize a thermonuclear reaction $d+t \rightarrow \alpha+n+17.6$ MeV. The d–t reaction has lower Coulomb barrier and higher cross-section in comparison with the d–d reaction. Methods of creation of the bubbles with high collapse ratio (maximum radius R_m divided by minimum radius R_c) are also very important. The shock wave model [28] predicted that inorganic liquids such as heavy water (D_2O) may not be good for cavitation bubble fusion because of its relatively low molecular weight, high sound speed in vapor, low accommodation (i.e. condensation/evaporation) coefficient, and low cavitation strength. Experiment [26] confirmed this theoretical prediction: no evidence of fusion could be observed in the system of D_2O plus vapor D_2O. But, it is known [1], that systems of water (H_2O, D_2O) plus H_2 or D_2 do not produce bright sonoluminescence because hydrogen bubbles are unstable in water. Possibly, behavior of D_2O vapor bubbles in D_2O is the same. On the other hand, water is the most effective liquid for sonoluminescence experiments when other gases (air, noble gases) are used [1,3]. And such system of D_2O plus gas (or mixture D_2O+T_2O plus gas) under abovementioned conditions are perhaps good for sonofusion.

4. Conclusion

Thus, the presented model explains qualitatively the main experimental results. There is, however, still one problem: an experimental confirmation of the presented model. Probably, the simplest way to do this is to investigate the light output dependence upon the sound frequency for SBSL under the same other conditions. According to our model the light output has to increase as frequency increases, at least, for frequencies f_a that do not attain values $f_a \gg \tau^{-1}$ ($\tau = \varepsilon\varepsilon_0\rho_0$).

Acknowledgements

Author is grateful to R.P.Taleyarkhan for valuable discussion. Author thanks S.V. Borissenok, A.A. Lebedeva, A.V. Pluznikov, D.R. Goncharova, and V.G. Simakov for help with manuscript.

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