Peculiarities of localization of several sonoluminescent bubbles in spherical resonators

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Abstract

Experiments on generation of 1, 2, 4, and 6 sonoluminescent bubbles in water with an external ultrasound source in an acoustic sphere resonator with glass walls have been carried out. Theoretical examination has shown that the observed excitation frequencies could be described with a good accuracy taking into account that the velocities and pressures of the contacting media on the external and internal resonator surfaces are equal. The necessity of accounting for oscillations with non–zero self–values of angular momentum operator has been shown when describing the features of localization of several bubbles. To explain a strangely small distance between the bubbles in the case of two–bubble sonoluminescence the following possible explanations have been proposed: a) mechanism of space splitting of a mode with a singular angular momentum and b) mechanism of secondary excitation when one of the bubbles is trapped into the acoustic trap created by high–frequency vibrations arising simultaneously when the other bubble sonoluminescence occurs.
1 Introduction

The phenomenon of sonoluminescence – light emission by gas–vapor bubbles under the influence of acoustic cavitation – was observed in 1930 during examination of chemical reactions initiated by ultrasound. The great interest in this problem was aroused from the early ’90s due to discovery of the effect of single bubble sonoluminescence in the field of a standing sound wave [1]. The most part of the experimental and theoretical works on this subject are dealing with its different aspects (see, for example, the reviews [2–4]). Less studied remain the processes where several sonoluminescent bubbles are involved, particularly, their space localization inside the acoustic resonator is insufficiently studied. To describe standing waves in a glass sphere resonator, it is proposed in [2] to consider the velocity potential value on the glass–air boundary equal to zero and the transition from liquid to glass was neglected. The analysis of our experiments has shown, however, that within this approximation it is impossible to explain the observed resonance frequencies of the small number (one to six) of sonoluminescent bubbles. In addition, in our experiments when several sonoluminescent bubbles were excited, they formed rather stable space structures, which cannot be easily described with spherical symmetric velocity potentials corresponding to zero orbital momentum (described in [2]). This work describes the experiments on generation of several sonoluminescent bubbles in a sphere resonator as well as theoretical analysis of the results obtained.

2 Experiments

Present experiments were aimed at studying special features of SBSL in acoustical resonators made of glass with configurations slightly different from spherical.

2.1 Resonators

Resonator #1. Spherical chemical retort with a long narrow neck. The maximum size of the retort was 64.5 mm, minimum one 63.0 mm; wall thickness 1.0 mm. The neck: length 105.0 mm, external diameter 7.5 mm, internal diameter 4.5 mm.
Resonator #2. Spherical chemical retort with a spherical bottom and wider neck. Diameter of the retort 63.0 mm, thickness of the wall 1.5 mm, diameter of the neck 20 mm, length 30 mm.

Resonator #3. Thin wall retort with ellipsoid–of–revolution form, and a short neck. Maximum and minimum retort cross sections: 62.0 and 59.0 mm. Neck: diameter 11 mm, length 5 mm.

During the experiments Resonators #1 and #2 were supported by their necks with the help of laboratory three–fingered clamp stand. Resonator #3 was placed into a type of a string–bag made of strong sewing suspended on a stand.

2.2 Electroacoustic transducers.

To induce the vibrations three types of hollow cylinder transducers were used.

Transducer #1. Diameters: 30/26 mm, height: 16 mm.

Transducer #2. Diameters: 18.5/16.0 mm, height: 22 mm.

Transducer #3. Diameters: 18.5/16.0 mm, height: 10 mm.

As a sensor of acoustical vibrations a self–made piezoelectric cells were used. Resonator #1 was equipped with one activator located within the equatorial plane, Resonator #2 was equipped with two activators located symmetrical within the equatorial plane. Resonator #3 was equipped with one activator located at the bottom of the retort.

2.3 Electronics

Activators were connected to the transformer output of low frequency 100 W amplifier with 60 kHz band pass and 150 V maximum voltage. The amplifier was fed by low–frequency generator G3–110. The scheme contains also series LC circuit: L – variable inductance of a coil with a ferrite core, C – capacitance of activator; the circuit being tuned to one of the fundamental frequencies of a resonator.
2.4 Seeding of cavitation centers

Seeding of cavitation centers was carried out by dropping a special nail–headed rod onto a surface of water in the base of resonator necks, or by injection of air bubbles into the water by a microsyringe in the case of Resonator #3.

2.5 Results

In the case of Resonator #1 a satellite bubble was observed, i.e. a second luminescent bubble. Brightness of this second bubble was much less than that of the main bubble. Satellite oscillated a little. Both bubbles located along the axis of the neck of the retort above (the main bubble), and below (the satellite) the center of the retort. The frequency was $f = 27048$ kHz, the quality factor of the system $Q = 1000$.

In Resonator #2 there were four luminescent bubbles in the vertexes of an imaginary quadrant. Plane of the quadrant changed a position a little depending on the frequency.

In Resonator #3, a stable luminescence took place concurrently for six bubbles, alternately fading and arising again in different positions. The frequency was $f = 53840$ kHz (the second harmonic), the quality factor $Q = 234$.

3 Excitation of single sonoluminescent bubbles

It is known that standing monochromatic acoustic waves of spherical form are described with solutions

$$
\psi = Ae^{-i\omega t} \frac{\sin kr}{r}, \quad k = \frac{\omega}{c}
$$

of radial wave equation

$$
\frac{1}{c^2} \frac{\partial^2 \psi}{\partial t^2} = \frac{1}{r^2} \frac{\partial}{\partial r} \left( r^2 \frac{\partial \psi}{\partial r} \right)
$$

for the velocity potential $\psi$. This solutions satisfy the finiteness condition in a point $r = 0$, which is true if there is no source at the point of origin [5]. We will assume
for liquid contained in the spherical flask that the acoustic field inside it is described by a potential (1), and inside its wall with a potential
\[ \psi' = A'e^{-i\omega t}\frac{\sin(k'r + \alpha)}{r}, \quad k' = \frac{c}{c'}, \]
where \( c \) and \( c' \) are sound velocity in water and in glass, respectively, and \( \alpha \) is a phase shift caused by the transition of the wave to the material with different mechanical properties. The condition that the pressure on the outer side of the wall is equal to zero (we neglect wave radiation into environment) gives
\[ p|_{r=a_+} = -\rho'\frac{\partial\psi'}{\partial t}|_{r=a_+} = 0, \]
where \( a_+ \equiv a + d \) is an external radius of the flask and \( d \) is the wall thickness. From here we have
\[ k'a_+ + \alpha = n\pi, \quad n = 0, 1, 2, \ldots \]
Further we will assume that \( n = 1 \) (it appeared that this very \( n \) value agrees with the experiment), which corresponds to the solution
\[ \psi' = -A'e^{-i\omega t}\frac{\sin k'(r - a_+)}{r}. \]

We will describe wave reflection on the water–glass boundary accounting for the requirement of medium continuity and the condition of pressure equality at the both sides of the boundary [6]. The last condition gives
\[ \rho A \frac{\sin ka}{a} = \rho'A' \frac{\sin k'd}{a}, \]
or
\[ A' = \frac{\rho}{\rho'} \frac{\sin ka}{\sin k'd} A. \]
The requirement of medium continuity on the surface \( r = a \) is reduced to the equality in the points of velocity of both media \( v = \partial\psi/\partial r \) and \( v' = \partial\psi'/\partial r \). Therefore, we find
\[ A \left( \frac{k \cos ka}{a} - \frac{\sin ka}{a^2} \right) = A' \left( \frac{k' \cos k'd}{a} + \frac{\sin k'd}{a^2} \right). \]
This with the account of (4), gives the following transcendental equation for finding the wave number \( k \):
\[ ka \cotg(ka) - 1 = \frac{\rho}{\rho'} \left[ 1 + \frac{c}{c'} ka \cotg \left( \frac{c}{c'} kd \right) \right], \]
where $c/c' \simeq 0.25 \div 0.3$ and $\rho/\rho' \simeq 0.4$ are the ratios of sound velocities and material densities of water and glass. First solution (5) with a minimum value of wave number $k$ corresponds to $ka \simeq 3.685$, which, for example, for $a = 3.15$ corresponds to the glass thickness $d \simeq 2.6$ mm, which is rather close to a real one. Acoustic oscillation frequency is connected with the wave vector $k$ by the relation $f = ck/2\pi$, which also gives a value close to the observed $f \simeq 27$ KHz.

According to the existing theory which is in a good agreement with the experiment, cavitation bubbles are located, depending on their size, either in nodes, or in points of local maximum of pressure amplitude in standing acoustic wave (anti–nodes). Thus, small–size bubbles (smaller than a certain resonance value) are located in nodes, while big ones – in points of maximum [7]. Experiments with several sonoluminescent bubbles have shown that in this sense they are “big” since they are located in anti–nodes [2].

Equation (5) allows one to identify the experimentally observed bubble trapping at a center of water–filled glass flasks with outer radiuses $a_+$, equal to 3.15 and 3.20 cm, at excitation frequencies 27 and 26.5 KHz, respectively, as their trapping by the acoustic traps corresponding to the solutions (5) with minimum values of wave number $k$ i.e., with a first harmonics of the resonator self–oscillations). At the same time, the bubble observed at the frequency of 53.82 KHz at the center of the water–filled glass flask at $a_+ = 3.025 \pm 0.075$ cm, can be corresponded with the second harmonics. Single sonoluminescent bubbles corresponding to third and higher harmonics have not been studied in these experiments.

4 Excitation of two, four, and six sonoluminescent bubbles

Since at acoustic wave excitation under real conditions the spherical symmetry condition is violated surface excitations in the flask should be described with the following function:

$$r_s(\theta, \varphi, t) = R(t) + \sum_{l,m} a_{lm}(t) Y_{lm}(\theta, \varphi),$$
where \( Y_{lm} \) are spherical functions accounting for the dependence of surface deformations on \( \theta \) and \( \varphi \) angular coordinates. Then the wave equation for the potential should be as well taken in the general form

\[
\frac{1}{c^2} \frac{\partial^2 \psi}{\partial t^2} = \frac{1}{r^2} \frac{\partial}{\partial r} \left( r^2 \frac{\partial \psi}{\partial r} \right) - \hat{l}^2 \psi,
\]

where

\[
\hat{l}^2 = -\frac{1}{\sin \theta} \frac{\partial}{\partial \theta} \sin \theta \frac{\partial}{\partial \theta} + \frac{1}{\sin^2 \theta} \frac{\partial^2}{\partial \varphi^2}
\]

is the “angular momentum”\(^1\), which satisfies the condition

\[
\hat{l}^2 Y_{lm} = l(l+1) Y_{lm}.
\]

For the monochromatic wave with the potential

\[
\psi = e^{-i\omega t} R(r) Y_{lm}(\theta, \varphi)
\]

The radical wave equation takes the form of

\[
\frac{1}{r^2} \frac{\partial}{\partial r} \left( r^2 \frac{\partial R}{\partial r} \right) + \left( k^2 - \frac{l(l+1)}{r^2} \right) R = 0,
\]

from where it can be seen that the radical part of the potential depends on \( k \) and \( l \) parameters and does not depend of \( m \):

\[
R = R_{kl}(r).
\]

The solution of equation (6), satisfying the finality condition in the point \( r = 0 \), has the form

\[
R_{k0}(r) = \frac{\sin kr}{r},
\]

in agreement with the results of the previous section. General solution of equation (6), satisfying the finality condition in the point \( r = 0 \), has the form (see, for example, [8])

\[
R_{kl}(r) = C r^l \left( \frac{d}{dr} \right)^l \frac{\sin kr}{r} = c_{j_e}(kr),
\]

\(^1\)Since angular variables in equations (4.1) and (4.2) describe anisotropy and not rotary motion, the notion “angular momentum” should be understood here as a certain convention suitable for classification of the solutions.
where \( j_e(k) \) is the spherical Bessel function. Particularly,

\[
R_{k1}(r) = C_1 \left[ k \frac{\cos kr}{r} - \frac{\sin kr}{r^2} \right],
\]

\[
R_{k2}(r) = C_2 \left[ \left( \frac{3}{r^3} - \frac{k^2}{r} \right) \sin kr - \frac{3k \cos r}{r^2} \right]
\]

etc. The velocity potential

\[
\psi_{klm} = Ce^{-i\omega t} R_{kl}(r) Y_{lm}(\theta, \phi)
\]

(8)

corresponds to several oscillating out of phase standing spherical waves the symmetry centers of which are very close to each other. For example, we have for two centers

\[
\psi = C \left( \frac{\sin kr_2}{r_2} - \frac{\sin kr_1}{r_1} \right) = C \frac{\partial}{\partial r} \left( \frac{\sin kr}{r} \right) \frac{\partial r}{\partial s} ds = C' \frac{\partial}{\partial r} \left( \frac{\sin kr}{r} \right) \cos \theta,
\]

(9)

where \( r_1 \) and \( r_2 \) are the distances from the centers to the observation point, \( ds \) is the distance between the centers, which is assumed to be infinitely small, \( \partial r/\partial s = \cos \theta \) is a direction cosine of the observation point and the line connecting the centers. This potential form corresponds to \( l = 1 \). Similarly the velocity potentials, corresponding to the higher angular momentums, are interpreted.

The points of pressure extremals location showing bubble position in the resonator, are described with a system of equations

\[
\begin{align*}
\frac{d\psi}{dr} &= 0, \\
\frac{d\psi}{d\theta} &= 0, \\
\frac{d\psi}{d\phi} &= 0.
\end{align*}
\]

(10)

The velocity potential \( \psi \) at a preset excitation frequency \( f_{kl} \) is, generally speaking, a superposition of functions (8) with different values of \( m \) projections of angular momentum, \( m = -l, ..., +l \). In the case of frequency degeneration (within the \( \Delta f \) width determined by the system \( q \)-factor by \( l \) parameter , it is necessary as well to take into account the superpositions of the states with different values of orbital
momentum. For separate $\psi_{k l m}$ modes the system (10) can be simplified and obtains the following form:

$$
\begin{cases}
\frac{dR_{kl}}{dr} = 0, \\
\frac{dY_{lm}}{d\theta} = 0, \\
\frac{dY_{lm}}{d\varphi} = 0.
\end{cases}
$$

(11)

From (11) for the $Y_{1, \pm 1}$ mode corresponding to $l = 1$,

$$
\begin{align*}
\theta_1 &= 0, & \theta_2 &= 0, \\
\varphi_1 &= 0, & \varphi_2 &= 0.
\end{align*}
$$

The obtained angular coordinates correspond to the experimentally observed at the frequency $f \simeq 49.5$ KHz four–bubble configuration located in the flask with the radius $a_+ = 3.2$ at the vertexes of an imaginary square.

If for the approximate estimate of probable distances from the bubbles to the center of the flask according to [2], we take soft conditions at the external boundary

$$
p|_{r=a_+} = -\rho \frac{\partial \psi}{\partial t}|_{r=a_+} = 0,
$$

where $\psi$ is the velocity potential of acoustic oscillations in liquid, then, accounting for the explicit expression for $R_{k1}$, we obtain the equation

$$
tg ka_+ = ka_+,
$$

the solutions of which are

$$
ka_+ \simeq 4.4934, \quad 7.7253, \quad 10.9041 \quad \text{etc.}
$$

Taking into account that $k = 2\pi f/c \simeq 2.075$ cm$^{-1}$, we find that only the first of these solutions corresponds to the experimentally observed excitation frequency.

Probable positions of the bubbles $r_i$, are determined from the condition

$$
\frac{\partial p}{\partial r} = 0,
$$

which leads to the equation

$$
tg kr_i = \frac{2kr_i}{2 - (kr_i)^2}.
$$
It can be seen that the first of the solutions
\[ kr_i \simeq 4.4934, \quad 7.7253 \quad \text{etc.}, \]
of this equation with the account of \( k \) value gives
\[ r_1 \simeq 2.2 \text{ cm}, \]
which is somewhat greater than the experimentally observed value \( r_{\text{exp}} \simeq 1.5 \text{ cm} \). This shows that the speculations of the previous section, accounting not only for the external boundary conditions but for the water–glass interface conditions as well, should be true for the case of various sonoluminescent bubbles (for each of the radial functions \( R_{kl} \)).

It would be natural to take as a potential a combination of states corresponding to \( l = 2 \) for the six–bubble configuration observed at a frequency of \( f \simeq 53.8 \text{ KHz} \) located in the flask at the same plane approximately one centimeter from the glass wall. In this case the solution of system (11) for angular coordinates gives
\[ \theta = \frac{\pi}{4}, \frac{3\pi}{4}; \quad \varphi = 0, \pi \]
for \( Y_{2,\pm1} \):
\[ \theta = 0, \frac{\pi}{2}, \pi; \quad \varphi = 0, \frac{\pi}{2}, \pi, \frac{3\pi}{2} \]
for \( Y_{2,\pm2} \). On the assumption that not all of the bubble traps were filled up (though no special measures had been undertaken for this purpose), among the solutions obtained one could find the sets similar to the experimentally observed configuration. It is especially interesting to analyze a two-bubble configuration located along the vertical line near the center of the flask (with a radius \( a_+ = 3.15 \text{ cm} \)) at \( f \simeq 26.9603 \text{ KHz} \), frequency where one of the bubbles was emitting light more intensely than the other. The distance between them is unexpectedly small (only few millimeters), which is difficult to explain with the use of equation (11) for a radical part of the potential\(^2\). One of the possible explanations of this fact is that

\(^2\)Small distances of bubbles from the center of the flask correspond to large values of the wave number. In this case, however, \( k = 1.13 \text{ cm}^{-1} \).
in this case one of the bubbles was trapped by an acoustic trap created by the high–frequency pulsations of the other arising simultaneously with its sonoluminescence. It is well known that the wave length of these high–frequency oscillations is approximately 20 times smaller than the standing wave length exciting sonoluminescence [2], and in our case is within the millimeter range. If the flask had the shape of a spheroid with a small distance between the focuses and if one of the bubbles was located at one of the focuses, then the emitted by it high–frequency waves should be accumulated at the other focus [11] thus creating an acoustic trap capable of supporting sonoluminescence of another bubble. The other possible explanation of these results is connected with formula (9), which shows that the solutions corresponding to \( l = 1 \), and the configuration of two bubbles, located at a finite though small distance between each other, are close. The transition of the common solution with \( l = 1 \) into the configuration of two bubbles oscillating in anti–phase is possible on the assumption of a certain instability connected with excitation of vertical oscillations due either to the fact that the flask has a neck, or to periodic deformations of the surface of the flask by the attached piezoelectric transducer. At present this problem remains open.

5 Conclusion

This paper describes an experimental setup permitting to study the processes of single– or several–bubble sonoluminescence in a sphere resonator. Though seemingly simple, an experiment of this kind is not a trivial task and it requires a number of conditions (such as fine tuning of the excitation frequency, requirements to the resonator \( Q \)–factor, temperature, and pressure in cavitating liquid, etc.). At the same time it is very important to study these processes if regarded that at present no generally accepted theory exists explaining the nature of the observed light emission [2, 4], and also because recently the chances appeared to attain extremely high temperature and pressures in these processes [2, 9, 10]. Therefore, it is clear that the precise description of the conditions under which stable laboratory reproducibility of these phenomena is possible is of much importance.
The analysis performed in this paper has shown that when calculating resonant excitation frequencies, at which one could expect sonoluminescent bubble arising, boundary conditions should be accounted for not only at the external but at the internal side of the resonator wall as well. In the case of several bubbles excitation, radial oscillation modes with different from zero orbital momentum should be accounted for. Further, to check the correctness of the proposed description one should have the precise measurements of bubble position in the resonator with the account of optical distortions due to light passing through the water–filled glass flask. It seems important as well to study the reason of the anomalous approach of two sonoluminescent bubbles.

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References

[1] Gaitan D.F., Crum L.A., Roy R.A. // J. Acoust. Soc. Am. 1992. V. 91. PP. 3166–3172.

[2] Barber B.P., Hiller R.A., Lofstedt R., Putterman S.J., Weninger K.R. // Phys. Rep. 1997. V.281. PP. 65–143.

[3] Putterman S.J., Weninger K.R. // Annu. Rev. Fluid. Mech. 2000. V.32. PP. 445–476.

[4] Margulis M.A.// UFN. 2000. V.170. Pp. 263–287.

[5] Landau L.D., Lifshits E.M. Hydrodynamics. M., Nauka. 1986. P. 379.

[6] Skudrzyk E. Bases of acoustics. Volume I. M. Edit. IL, 1958. P. 141.

[7] Yosioka K., Kawasima Y., Hirano H. Acustica. 1955. Vol.5. PP. 173 – 178.
[8] Landau L.D., Lifshits E.M. Quantum Dynamics. M., Nauka. 1974. P. 135.

[9] Taleyarkhan R.P., West C.D., Cho J.S., Lahey Jr. R.T., Nigmatulin R.I., Block R.C. Science. 2002. V.295. PP.1868–1873.

[10] Beliaev V.B., Kostenko B.F., Miller M.B., Sermyagin A.V., and Topolnikov A.S. Ultrahigh temperatures and acoustic cavitation. 2003. JINR Communication P3–2003–214. Dubna.

[11] Encyclopedia of elementary mathematics, volume 5. M. Nauka, 1966. P. 575.