OBSERVATION OF SONOLUMINESCENCE AND SUBHARMONICS

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In this paper we describe results of experimental investigation of multibubble sonoluminescence. Experiments were done in an acoustic resonator with water. It was found that sonoluminescence and subharmonics occurrence follows by each other when the amplitude of acoustic pump wave in the resonator increases and decreases. It was also observed a hysteresis effect.

INTRODUCTION

The effect of a synchronous buildup of sonoluminescence (SL) in water and occurrences of ultrasonic signals on subharmonics of the pumping frequency (SG) has been known for a long time [1], but its properties and mechanism have been still poorly studied. We have carried out synchronous measurements of SG and SL characteristics at various levels of ultrasonic pumping.

1. EXPERIMENT

The ultrasonic pumping field was created in a reactor which corresponded to an upright oriented cylindrical vessel which was formed by a piezoelectric ceramic emitter plate. For each measurement the same amount of water was poured into the reactor with the water layer thickness equal to 1 and 1/4 of the pumping sound wave length. The upper water boundary was free and contacted to air. A pumping frequency of 500 kHz has been chosen. Thus a standing wave with a pressure antinode at the emitter surface was set in. The wave had also an antinode of oscillatory velocity at the free surface of water and two other pressure antinodes within the water column, alternating with two oscillatory velocity antinodes. In these experiments the amplitude range of ultrasonic pumping pressure at the emitter surface was within (0.8-1.8) 10^5 Pa.

The SG signals were measured on the coatings of the same piezoelectric plate which was used for the ultrasonic pumping. For SG signal filtration and amplitude measurement a circuit of an passive eliminator and a selecting voltmeter was used. The SG signals were measured in 10 kHz frequency range with the central frequency of 250 kHz.

The SL pulse-repetition rate from a multiplier photocell with its photocathode oriented from above to the water surface was measured with a frequency meter. The multiplier photocell perceived SL signals in the optical part of the spectrum (up to 300 nanometers). In experiments distilled water was used.

The measurements were carried out under the air atmosphere at normal pressure. The water temperature was kept near 43 °C. Some tens of measurement cycles with step-by-step pumping level variation have been carried out. In some cycles the pumping stepped up, starting from the level, when SL and SG still missed, crossed the area of SL and SG occurrence and was increasing up to the maximum level. Then it stepped down in the same manner to the initial minimum level. In other cycles the measurements started at the maximum pumping level, the pumping stepped down until SL and SG signals disappeared, and then it stepped up again by the same steps up to the maximum level.

2. RESULTS AND DISCUSSION

As a result of separate data averaging for both measurement cycles it has been shown, that the SL and SG characteristics for both types of measuring appeared rather similar with the only difference in the area of occurrence and disappearance SG and SL. For example the SL and SG characteristics obtained only by averaging over the cycles when pumping was first stepping up from the minimum value and up to the maximum level and after returned to the initial value with the same steps are shown in Fig.1. In the figure X-axis corresponds the amplitude of ultrasonic pressure on the emitter surface. The Y-axis corresponds the following mean values:

1) SG levels averaged over 10-30 seconds at each step of pumping in each cycle, and

2) mean SL pulse counting rates also averaged over 10-30 seconds at each step of pumping in each cycle.

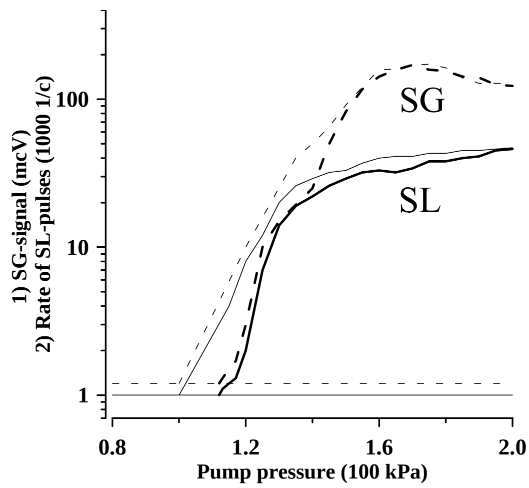

Fig.1 Average characteristics of SL buildup (heavy line) and quenching (thin line) and SG ccurrence and failure in the measuring cycles starting and ending at small pumping level.

The heavy lines show the mean SL and SG characteristics at pumping increase, while the thin lines show the average SL and SG variations at pumping level decrease in the second part of cycles when the pumping reduced. Root-mean-square deviations for the average data amount from ±(100-50) % for the area of rather weak pumping when the SL buildup (or quenching) takes place and SG appears or disappears up to ±(30-15) % in the pumping range with amplitudes over $1.5 \cdot 10^5$ Pa. These fluctuations are not shown in the figure. The horizontal lines correspond: the lower one shows the multiplier photocell pulse rate in the dark current mode, the horizontal line above shows the noise level in the SG range at the selective voltmeter input and at the minimum level of pumping.

The difference between the pumping levels of occurrence and termination of SL and SG is also well found out by viewing histograms of SL and SG occurrence and termination moments, which characterize probabilities of occurrence and termination as a function of pumping pressure. These histograms are given in Fig. 2 where the results of all of 120 cycles of observations are summarized. The step of histograming is equal to the next two steps of pumping sweeping. In the common sweeping range the interval, shown in Fig. 2, amounts less than 25 %. In the top of Fig. 2 the histograms of occurrence (heavy line) and failure (thin line) of SG are shown, in the lower part of the figure are the histograms of buildup (heavy line) and quenching (thin line) of SL are given. From Fig. 2 it is well clear, that the maximums of histograms of SL buildup and SQ occurrence SG are connected to higher pumping levels, than the maximums of histograms of SL quenching and SG failure, and, i.e., these processes have a hysteresis.

SL buildup (or quenching) and SG occurrence (or failure) thresholds have been selected in accordance with a specified criterion. It was believed, that the threshold of SL buildup (or SG occurrence) was at that step of pumping level, when the SL pulse rate (SG amplitude) sharply increased (twice or more) in comparison with the previous step of pumping. For the SL quenching or SG failure threshold a pumping level was taken, when the SL pulse rate or SG level differed from the level of the dark current mode and SG noise level less than twice.

It is necessary to note, that even at a constant level of pumping SL and SG signals have trends: at each pumping increase in the characteristic ranges, following the moment of threshold overcoming, the signals rise on for a certain time, and at transition to lower step of pumping they often decrease gradually. Consequently the method of threshold definition is correct only within two next steps of pumping sweep. Except for that SL and SG arose sometimes not simultaneously. As a whole the threshold coincidence for SG occurrence and SL buildup was observed in 65 % of cases; in 20 % SG occurrence preceded by one sweeping step (in 2 cases the difference amounted two steps) to SL buildup, and in the rest 15 % cases SL inflamed earlier by a step, than the threshold of SG occurrence was fixed. The coincidence of the moments of SG failure and SL quenching at pumping decreasing occurred more often: in 92 % of cases. In 8 % cases the advancing or trailing of SG regarding the moment of SL quenching appeared practically equiprobable.

The obtained SL and SG characteristics show, that within the experimental error:

1) At pumping increase SG occurrence and SL buildup SL occur practically simultaneously and further both the SG amplitude, and SL pulse rate grow monotonic, tending to saturation;

2) SL quenching and SG disappearance at pumping decrease also occur practically simultaneously, but at smaller levels of pumping, than their occurrence during pumping increase.

Thus the effect of a hysteresis for occurrence and disappearance of SG and SL has been revealed.

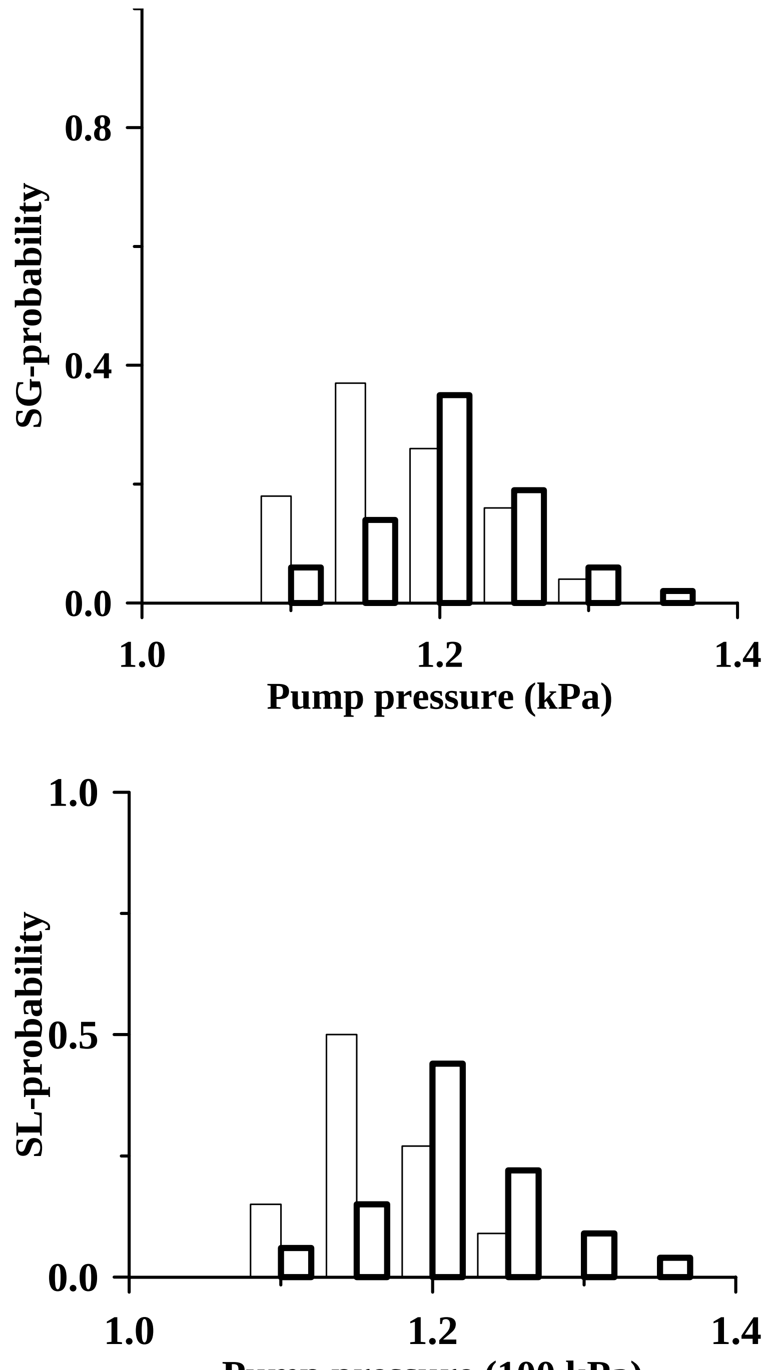

Fig.2 Histograms of the moments of SG occurrence (heavy line) and failure (thin line) and SL buildups (heavy line) and quenching (thin line), obtained from data collection SG and SL characteristics over all cycles of measurements.

Besides on the reactor taken from blacked out box visual observations with the simultaneous check of SG signals were carried out.

It turned out, that SG signals appear as separate impulses with duration about 2 ms (especially well it can be observed at rather low pumping levels). At pumping increase the frequency of occurrence of such SG pulses grows, but their duration and amplitude vary insignificantly. In general the occurrence of such SG pulses are well compounded with acts of occurrence of gas bubbles emerging through the water column. It was observed, that the sizes of such detached bubbles are close to the resonance size for the frequency of pumping. It follows also from direct visual observations of bubble sizes by a microscope, and also from the fact, that emerging bubbles practically do not change their velocity of emersion, crossing pressure and oscillatory velocity antinodes in the ultrasonic field of the reactor.

The events accompanied by SG occurrence can be qualitatively described as follows. Some gas cavities, necessarily remaining on the emitter surface after filling the resonator because of nonwettabillity, at a certain pumping level start to grow due to water degassing. At the moment when the cavity sizes become close to resonant for the pumping frequency, their volumes change significantly under ultrasonic field, and their oscillations become strongly nonlinear. In these conditions parametric oscillations at higher (non- spherically symmetric) modes on a surfaces of attached to the wall bubbles are raised. These modes are generated at the subharmonic frequency. The development of such oscillations up to high amplitudes comparable with bubble radius gives in division the gas cavity attached to the emitter surface, and occurrence in volume of water gas bubbles.

As on the emitter surface a "parent" gas cavity is still present, the size of the detached bubble is at the first moment less than the resonant one, and consequently the bubble remains in the pressure antinode area under the radiation force near to the emitter surface. But the bubble size is growing fast due to a rectified diffusion and when such separated bubble size becomes resonant to pumping, it leaves the area near to the emitter surface and emerges to the water surface.

In our interpretation of the experimental results regarding occurrence, development and SL quenching which occur at the average synchronously to SG occurrence and failure, we adhere to the point of view, that SL is a recombination luminescence of sonolisys products in water. In turn the sonolisys, i.e. the dissociation of water under sound, is basically a result of viscous losses [2-5]. In our previous papers it has been shown, that the sonolisys efficiency (the relation of the sonolisys output to viscous losses) is identical both at presence of bubbles, or without them. Therefore gas bubbles generally are not necessary for sonolisys. In the same time in the presence of bubbles a sharp SL buildup occurs. This effect is probably connected with higher mode excitation of a bubble surface at SG frequency [6,7]. The nature of the excitation of compound oscillations of a bubble surface is parametric: the phenomenon of a hysteresis is peculiar to such processes at excitation and failure of oscillations. In our experiments quadrupole oscillations of bubbles (the second mode) should be generated. Unfortunately it was impossible to observe oscillations on a surface of a small bubble. At excitation of capillary waves on bubble surface the viscous friction is located in shift currents in the layer with thickness comparable to length of a capillary wave. Accordingly, in the water layer, adjoining to the bubble dissociation of water most intensively occurs. Therefore concentration sonolisys products becomes much higher close to bubbles, than on the average over the entire volume of water. It seems to be a reason of SL buildup synchronously with bubble occurrence in water.

3. CONCLUSION

In this work we experimentally studied peculiarities of occurrence and termination of sonoluminescence and subharmonics. It was found that their characteristics are similar during cycles of acoustic pumping increase and decrease. It was also observed the hysteresis effect. Visual observations has revealed the process of bubble detaching from the acoustic emitter. Obtained results can be interpreted from the point of view of water sonolisys caused by bubble shape oscillations and recombination luminescence of products of water sonolisys.

ACKNOWLEDGEMENT

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