N.V. Dezhkunov

MECHANISMS OF THE ENHANCEMENT OF SONOLUMINESCENCE IN INTERACTING ULTRASONIC FIELDS HIGHLY DIFFERING IN FREQUENCY

Belarusian State University of Informatics
and Radioelectronics, P.Brovka St. 6, 220072 Minsk, Belarus
Tel.: (172) 39-8635; Факс: (172) 39-0914
E-mail: dnv@bsuir.edu.by

This work provides the results of investigation of the mechanism and patterns of sonoluminescence generation in the interacting ultrasonic fields. It is shown that nonadditive SL enhancement takes place not only under the simultaneous influence of the fields on the liquid but also in the high frequency (HF) field after preliminary liquid insonation by the low frequency (LF) field as well. The long term afteraction of the LF field on the cavitation generated by the HF field was discovered. The preliminary action of the HF field on the liquid makes no effect on the intensity of the SL generated by the LF field. The conclusion was drawn on the basis of the results received that prevailing mechanism of SL amplification is the generation of new cavitation nuclei upon the collapse of cavitation bubbles driven by the LF field.

In works [1-4], it has been shown that the action of a low-frequency (LF) field on the cavitation zone generated by a high-frequency (HF) is an efficient method for increasing the activity of cavitation. The SL intensity in a combined field generated by the simultaneously operating pulsed HF and continuous LF radiators exceeds many times the sum of the SL intensities produced by each of the fields individually [4].

This is in agreement with results of investigation of ultrasonic capillary effect [5], chemical activity of cavitation [6-10], erosion rate and drug delivery to the suspended cells [11-13].

Increase of energy introduced into the liquid by two transducers and interference of the fields are evident reasons of the increase of the intensity of sonoluminescence. Let us consider other possible mechanisms.

1. Decrease in the total quasistatic (in respect to the HF field) pressure during rarefaction phase of the LF which promotes increase of the sizes of cavitation nuclei and HF cavitation threshold decrease [14]. As a result the number of bubbles cavitating under HF field increases and causes corresponding increase of SL intensity. In the half - period of LF-field compression ($P_{LF}>0$), the total quasistatic pressure increases which corresponds, according to [14,15,16], to the quicker collapse of the bubbles formed during the half-period 0f rarefaction of the LF - field.

2. Suppression of forming the clusters of cavitation bubbles pulsing under HF field. The reason of this suppression may be action of shock waves and microjets generated by large (HF) bubbles. Interbubble interactions are considered to be main reason of the decrease of the efficiency of energy concentration in multibubble cavitation zone in comparison with single bubbles [17, 18]. The bubbles in clusters are held near each other [19,20,21] and therefore interact very strongly. It is possible that big bubbles generated by LF field hinder the formation of HF bubble clusters by seeding more uniformly the cavitation nuclei throughout the liquid volume. This may create better conditions for more efficient bubbles collapse under HF field.

3. Broadening of the spectral composition of the resultant field. The cavitation liquid is a substantially nonlinear medium. In interaction of the fields with frequencies f_{LF} and f_{HF}, in such a medium the waves of combined frequencies $f_{LF}+f_{HF}$ and $f_{HF}-f_{LF}$ can be generated [22]. Therefore, the resultant field covers a substantially wider set of the frequencies than the sum of the spectra of the initial fields. This must lead to broadening of the size range of the bubbles involved in the cavitation process and, consequently, to an increase in the total number of cavitating bubbles and intensity of SL [23].

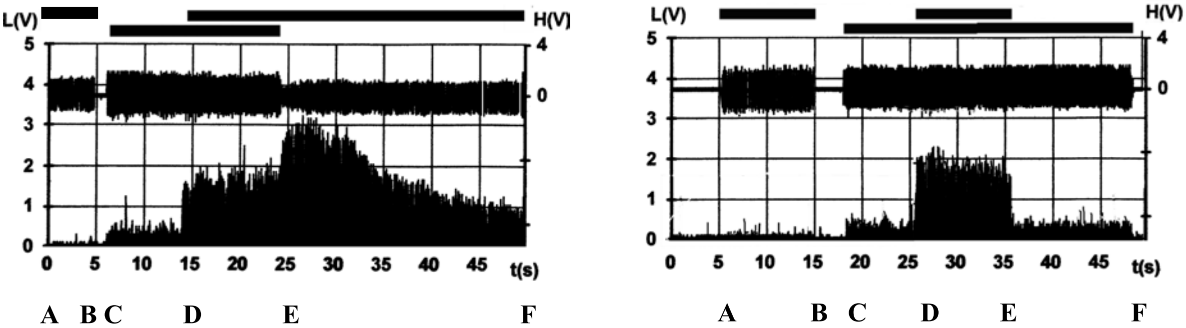
4. Generation of new nuclei of cavitation. As is known [24, 25], on collapse the cavitation bubbles break down into fragments. The seizes of these fragments, evidently, are much smaller, than that of initial bubbles. By this reason they may serve as nuclei for cavitation under HF field. These new nuclei contain less air than the initial bubbles. Therefore, they are likely to collapse in the HF field at a higher rate than the bubbles drown from the nuclei stably existing in the liquid. Thus, owing to this mechanism both the number of cavitation bubbles and the efficiency o their collapse can increase. The

last will entail an increase in the maximum pressures and temperatures attained in the vapor-gas mixture inside bubbles and, as a consequence, an increase in the SL intensity.

The results provided in the present work make it possible to evaluate the contribution of the indicated mechanisms to the effect of SL enhancement in interaction of the ultrasonic fields highly differing in frequency.

Experimental. The chamber represents a stainless cylinder with inner diameter of 120 mm and height 180 mm. The high frequency concave piezoceramic transducer having working frequency 880 кГц is mounted at the bottom of the chamber. The chamber has two windows on the lateral wall at the level of the focal spot of the HF transducer. The horn of the low frequency (19.9 kHz) transducer is introduced into the chamber through one of the window. Light guide of the photomultiplier is mounted on the other window. Hydrophone is introduced through the through the cover of the chamber in such a way that its sensor (2mm in diameter and 0,25 mm in thickness) is placed 50 mm over the focal spot of the HF transducer. For precavitation conditions sound pressure P at the focal point of HF transducer is linearly related to voltage U applied to the transducer: $P(10^5\,\text{Pa}) = k \times U(\text{Volt})$, and radiated power in all explored range of voltages is proportional U^2 with accuracy not lower than the accuracy of measurements. Here $k = 0.093$ Па/В.

Results. Figure 1, a shows results of recording simultaneously the output signal H of a hydrophone (the upper oscillogram) and of the photomultiplier signal L (the lower oscillogram).



a) b)

Fig. 1. Oscilloscope recordings of the hydrophone output *H* (upper record) and of the photomultiplier *L* (lower record). HF field parameters: pulse period – 100 ms, pulse duration – 2 ms, driving voltage $U_{HF} = 55$ V; vibration amplitude *A* of the LF transducer is 8 μm: a) after joint operation of both transducers (period DE) LF is stopped; b) HF is stopped.

Initially, HF and LF ultrasonic radiators were switched on in turn (AB and CD periods respectively, then they were switched on simultaneously for a certain time (period DE). Than LF transducer was closed and HF transducer proceeded to operate. The regimes of the transducers operation were chosen such that intensity of the HF field I_{HF} was not much higher that the threshold intensity I^*_{HF} at which SL appears, and intensity of the LF field I_{LF} was much higher than I^*_{LF}. So the photomultiplier averaged output was 17 mV for HF transducer working alone and 190 mV for LF transducer working alone. From this figure it is seen that the result of the joint action of the two fields (DE) is much higher than the sum of the results achieved by every field separately, i.e. nonadditive summation of the effects takes place.

Figure 1b shows the results of an experiment similar to the first one with the same modes of operation of the radiators. In the time intervals AB and CD, the HF and LF radiators were switched on alternately, in the interval DE both radiators simultaneously, and in EF only the LF radiator. Thus, the difference from Fig. 1, a lies in the sequence of switching off the fields after their simultaneous operation, namely; the HF radiator was switched off and the LF radiator continued to operate. As in the first case, at a concurrent operation of the radiators a considerable amplification of the SL was observed. However, two essential features compared to the previous experiment were revealed: 1) at the instant the high-frequency field was switched off there was no second jump of the SL intensity; 2) when the HF field was switched off the SL intensity dropped practically instantaneously to the initial value created by the LF field alone, i.e., the influence of the aftereffect of the switched-off (HF) field on the SL intensity generated by the operating (LF) field was absent.

Figure 2 presents the results of the SL recording in the course of the experiment performed according to a radically differing scheme, namely, first the LF field was switched on for a short time and then, some time **Δt** after termination of the LF radiation operation, the HF field was switched on. Thus, in this case, the actions of the LF and HF fields on the liquid are separated in time.

The modes of operation of the LF and HF radiators were the same as in the experiments whose results are given in Fig. 1. It is seen that If Δt is small or equal to zero (< 1--2 *c*), then at the instant the LF field is switched on the SL intensity abruptly increases to values close to the maximal attained under the conditions of the experiment presented in Fig. 1a. Upon the switch-on of the HF field at larger Δt L smoothly increases, reaches L_{max} and then slowly decreases, tending to some limiting value. With increasing Δt the attained value of L_{max} decreases, as does the growth rate of L. This is obviously due to the relaxation during the time interval Δt of the changes in the cavitation properties of the liquid caused by the action of the LF field. In some cases, the time of complete relaxation, i.e., the time scale of the memory, reaches several hours. During this period the liquid returns to the initial state and the value of L upon the switch-on of the HF field is practically equal to that measured prior to the action of the LF field, i.e., of the order of 17 mV for the conditions of the given experiment.

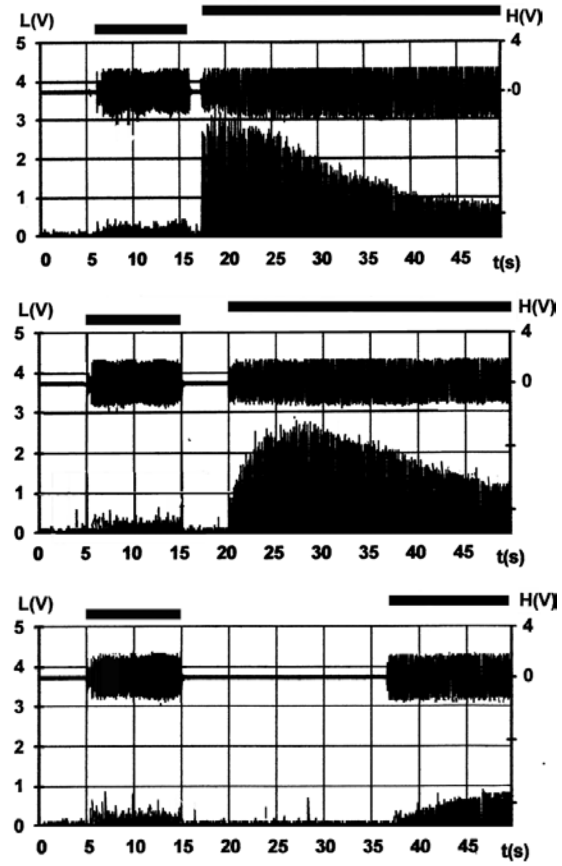


Fig. 2. *L* and *H* dependencies on time *t* with separate operatin LF and HF fields for different time intervals: Δt=1.5, 5, и 22.5 s (from up to down).

Figure 3 shows the time dependencies of the SL generated under the action of the HF field with a short-term superposition of the LF field for various intensities of the HF field with the LF field intensity greatly exceeding the SL threshold 3Л I^*_{LF}.The moments of switching on and off of the LF field are indicated by arrows pointing upwards and downwards, respectively. Averaging was performed over three experiments. If the HF field intensity is somewhat lower than or equal to the SL threshold, (figures 3, I, a, b) then under the action of the LF field L abruptly increases, reaches the limiting value, and then slightly changes with time. If the HF field intensity is higher than the threshold I^*_{HF} (Fig. 3, I, b), at the moment the LF field is switched off a second jump of the SL intensity is observed and only then does its decrease begin. If the HF field intensity is much higher than the SF threshold ($I_{HF}>>I^*_{HF}$), (Fig. 3, I, d), then after the LF field is switched on the total intensity of the SL does not increase, but decrease.

At a low intensity of the LF field (of the order of the cavitation threshold or lower, see Fig. 3, II, a-d) the character of the $L(t)$ differs as follows: when the LF field is switched on L increases not so much as in the previous case, and this increase is smooth, and not abrupt; when the LF field is switched off, the second jump of L is not observed whatever the HF field intensity. Moreover, the rate of decrease in L when the LF field is switched off (i.e., the relaxation rate of the cavitation properties of the liquid) is much higher compared to the relaxation rate after the action of an intense LF field (Fig. 2, 3). At a high intensity of the HF field (Fig. 3, II, d) the additional action of the LF field opearting at low intensity practically causes no increase in the SL intensity.

Discussion. As is seen from the above graphs, the result of the interaction of the two fields is strongly dependent on the mode of insonification. The mechanism of SL amplification associated with the direct interaction of the fields are obviously realised only if both fields are on. The fact that for the majority of modes of sonification at the instant the LF field is switched off the decrease in the SL intensity is not instantaneous but smooth permits suggesting that the contribution of these mechanisms to the effect being investigated is small and the main factor is likely to be the generation of new cavitation nuclei upon the bubble collapse. These nuclei are not stabilised by the surface film, or in the microcracks of particles in the liquid therefore, as soon as the LF field is switched off they rapidly reduce their sizes due to the gas diffusion from the bubble into the environment. As a result, the number of bubbles cavitating under the action of the HF field decreases, which leads to a decrease in the SL intensity. This hypothesis is verified by the fact that when the LF field is switched off the intensity of the acoustic signal received by the hydrophone (Fig. 1, a, upper oscillogram, portion EF) slowly increases. This means that the ultrasound absorption in the cavitation region decreases obviously due to the decrease in the bulk density of bubbles across the sonic wave.

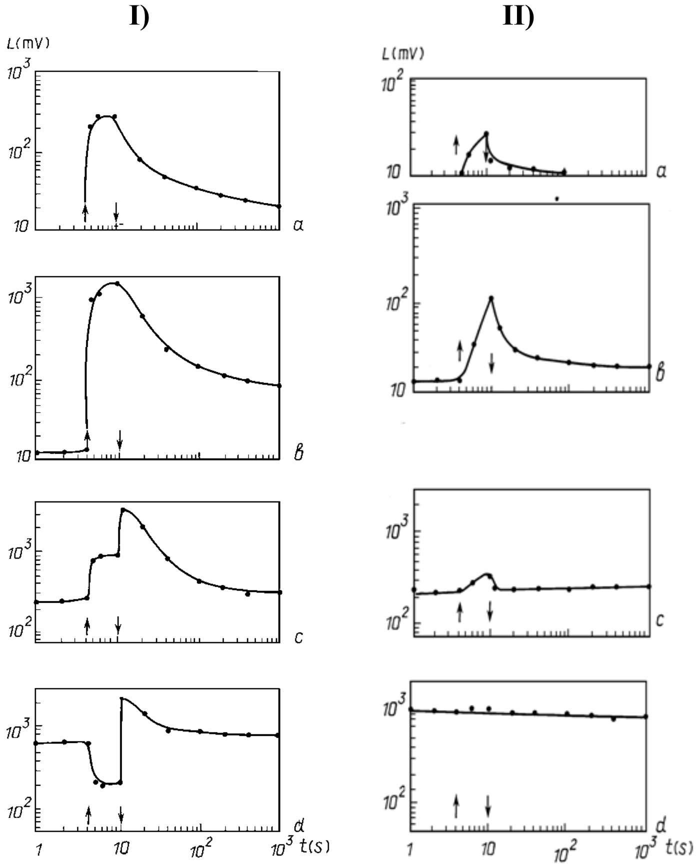


Fig. 3. Time dependensies of L on time t for different intensities of HF field at the intensity of the LF field much lower than SL threshold (A=12 micron) – I and at the intensity of LF field much higher than SL threshold (A=1,5 micron) - II. HF transducer voltage U, Volt = 55 (a), 75 (*b*), 125 (c) и 150 (d).

From the point of view of this model the influence of the aftereffect of the LF field on the SL intensity generated by the HF field with alternate switch-on of the LF and HF fields (Fig. 2) is explained as follows. When the LF field is switched off the new cavitation nuclei live for some time, providing a higher SL generation efficiency in the HF field. As is seen from Fig. 2, if the Δt is small, then L increases stepwise when the HF field is switched on, and the values obtained thereby are approximately equal to $L_{\max}$ (Fig. 2) achieved at the moment of switching off the LF field after joint operation of both transducers. Thus, in the absence of direct interaction between the fields, too, the resulting amplification of the sonoluminescence is not smaller than in the case of the simultaneously operating radiators.

Practically, the instantaneous fall of L as soon as the HF field is switched off (with the LF field on, Fig. 1, b) does not contradict the hypothesis about unstable nuclei either. Indeed, the bubbles cavitating under the action of the HF field are at least an order of magnitude (to be more precise, for the conditions of the experiments described above -- about 40 times) smaller than the bubbles cavitating under the action of the LF field. Therefore, neither the HF bubbles nor, the more so, their fragments can serve as cavitation nuclei for the LF field. This explains the absence of the aftereffect of the HF field on the cavitation generated by the LF field.

Attention is drawn to the jump of the SL intensity observed as soon as the LF field is switched off (Fig. 1, a and Fig. 3). To explain this feature, it is necessary to take into account that at a large concentration of cavitation bubbles, as a result of the strong interaction between them, the bubbles can lose their spherical form in the early stage of collapse and the rate of the latter can decrease. As a result, the conversion efficiency of the sound energy to the energy of shock waves and thermal energy decreases [26]. This can lead to a corresponding decrease in the intensity of the cavitation effects, including the SL. The screening action of the cavitation region, too, can have a negative influence on the cavitation activity at a large concentration of bubbles. When the LF field is switched off the number of bubbles in the cavitation region rapidly decreases and, possibly, at some instant, their density approaches the optimum density corresponding to the SL intensity maximum. Probably, this can cause an additional burst of L when the LF field is switched off.

The absence of the influence of fragments of HF bubbles on the LF-field-generated cavitation leads to the fact that such a burst is not observed with the HF field off and the LF field on (Fig. 1, b). Thus, if the intensity of the LF field is much higher than the cavitation threshold, then the prevailing mechanism of SL amplification is the generation of new cavitation nuclei upon the collapse of cavitation bubbles excited by the LF field. Then these nuclei cavitate under the action of the HF field, increasing the integrated SL intensity.

If the LF field intensity is lower than the SL threshold, then the LF bubbles, i.e., the bubbles pulsating under the action of the LF field, practically do not collapse and do not generate additional cavitation nuclei. In this case, a marked amplification of the SL is only observed for HF field intensities of the order of the SL threshold, i.e., $0{,}5\ I^*_{HF} < I_{HF} < 1{,}3\ I^*_{HF}$, and the aftereffect of the LF field either is absent or is short-term. In this case, the SL amplification is likely to be determined by the mechanisms associated with the direct interaction of the fields, i.e., by their combining, the periodic decrease in the quasi-static (with respect to the HF field) LF field pressure, and the widening of the spectral composition of the resulting field.

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