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The Evolution of a Cavitation Zone in a Focused Ultrasonic Field

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Abstract—We have experimentally studied the generation of sonoluminescence (SL) and cavitation noise in the field of a focusing ultrasonic radiator during a gradual smooth increase in the applied voltage. In addition to the SL signal, we have recorded the output signal of a hydrophone situated behind the focal region of the radiator and measured the cavitation-noise spectrum. Four stages in the development of a cavitation zone have been distinguished as manifested by the specific character of dependences of the measured parameters on the voltage applied to the radiator. Spectral signs of the cavitation-noise characteristic of each stage of development of the cavitation zone.

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In recent years, the effect of high-power (in particular, focused) ultrasound on processes in biological structures has been extensively studied [1–7]. The obtained results showed that many effects such as the improved penetration of drugs through cell membranes (sonoporation) [2, 3], suppression of tumor-cell growth [4, 5], enhancement of drug action [4, 6], and drug release from carriers [7] have a cavitation nature—that is, are related to the nucleation, growth, pulsation, and collapse of microbubbles in liquid media [8]. Hallez et al. [9] discussed the possibility of using focused ultrasound in the cavitation regime in sonochemistry. However, the laws of cavitation-zone development in the field of a focusing radiator are still insufficiently studied and methods of determining the state of a cavitation zone and measuring the cavitation activity have yet not been developed, which significantly limits the possible practical applications of focused ultrasound.

In the present investigation, we have for the first time separated four stages in the development of a cavitation zone, which are distinguished by characteristics of the cavitation effects, including sonoluminescence (SL), cavitation noise, and ultrasound absorption in the cavitation zone.

The experimental setup and methods have been described in detail elsewhere [10, 11]. The working cell comprised a stainless-steel cylinder with a diameter of 100 mm and a height of 160 mm with a piezoceramic focusing transducer (radiator) arranged in the bottom compartment. The resonant frequency of the piezoceramic element was $f_0 = 720$ kHz. The opposite edge of the cell was closed by a cone-shaped lid with the cone protruding inward the cell. A hydrophone was inserted through the lid center so that its receiving piezoceramic disk element with a diameter of 2 mm and a thickness of 0.25 mm was situated 25 mm behind the focal spot of the radiator. A photomultiplier (Philips XP1110) with 20-mm-diameter optic fiber was mounted on a side window made in the cell wall on the focal spot level.

The radiator had power supplied from a computer-controlled acoustic generator (UZG-08-01 BGUIR) equipped with a system of automated resonance frequency tuning. The maximum generator output power of 70 W was achieved at an applied voltage amplitude of 275 V. The experimental procedure was as follows. The cell was filled with distilled water and allowed to stand for 2 days, after which water was ultrasonically outgassed for 20 min at a transducer applied voltage of 170 V (~10 W/cm^2). Preliminary partial outgassing of the medium significantly improves the reproducibility of experimental results [10]. Upon outgassing, the cell was tightly closed by the inward-oriented cone lid. The output signals of the photomultiplier (L) and hydrophone (H) were preamplified and fed to a multichan- 1
nel digital oscilloscope (Hewlett Packard 54601A). The spectra of acoustic signals detected by the hydrophone were recorded by a Hewlett Packard E4411B spectrum analyzer.

Figure 1 shows the results of simultaneous monitoring of the output signals of photomultiplier (L) and hydrophone (H) as functions of time t during a gradual smooth increase in voltage U applied to the radiator. The voltage generator was switched on at $t = 5$ s. As can be seen, hydrophone output signal H at the first

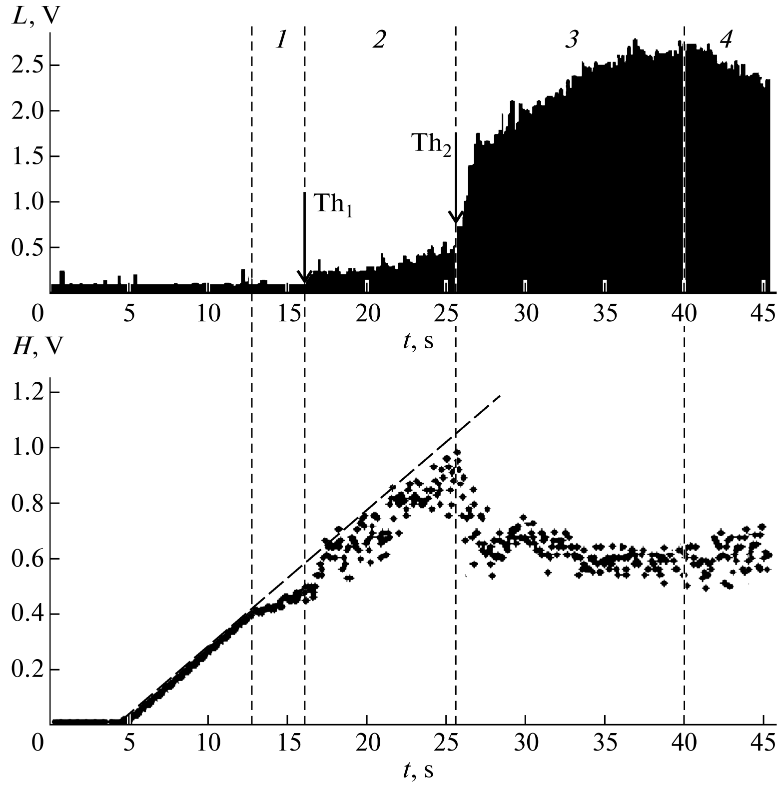

Fig. 1. Results of simultaneous ~~monitoring~~ of the output signals of the photomultiplier (L) and hydrophone (H) in pulsed focused ultrasound field during gradual smooth increase in the voltage applied to a radiator generating ultrasound pulses with duration $\tau = 3$ ms at a repetition period of $T = 100$ ms. Vertical dashed lines separate stages *1*–*4* in cavitation-zone development. The rate of computer-controlled voltage increase, 7 V/s; liquid temperature, 21 ± 1.5°C.

stage exhibits linear increase with time. Shortly before the SL onset (about $t = 13$ s), the hydrophone response deviates from the initial line toward the lower slope of the $H(t)$ plot, which is evidence for an increase in the absorption ultrasound energy—apparently due to the formation of bubbles in the focal region of the radiator.

In the example presented in Fig. 1, the SL emission with intensity L above the background level arises at approximately $t = 17$ ms, which is indicated by arrow Th_1 corresponding to the first SL threshold. The onset of SL is accompanied by a change in the character of the $H(t)$ plot, in which a significant scatter appears in the points recorded by the oscilloscope. At certain critical voltage U, the slope of $L(t)$ exhibits a jumplike change that corresponds to a sharp increase in the rate of SL intensity growth. This is indicated by arrow Th_2 corresponding to the second SL threshold. Rapid growth in the SL intensity is accompanied by a corresponding drop in hydrophone response signal H.

Total acoustic signal H represents a sum of the initial ultrasonic field and the acoustic emission from cavitation bubbles (cavitation noise). Stable pulsation and collapse of these cavities decrease the acoustic transparency of the focal region, especially at their growth stage, which can lead to screening of the field and a decrease in intensity of the hydrophone output signal (manifested by points situated below the average $H(t)$ line in Fig. 1). Collapsing cavities generate shock waves, which can give rise to pulses of increased intensity (manifested by points situated above the average $H(t)$ line). Rapid increase in the absorption of ultrasound in this regime is apparently related to the onset of multiplication of cavitation bubbles by the chain reaction mechanism described in [8]. After the rapid growth stage, the SL intensity tends to some limiting value and then begins to decrease, which is accompanied by visible formation of large stable bubbles.

Based on the results presented above, it is possible to separate four stages in the development of a cavitation zone in the focusing radiator field: (*1*) the appearance of bubbles and their pulsation without SL generation; (*2*) the appearance of emission and slow growth in the SL intensity at weakly increasing absorption of ultrasound; (*3*) fast (frequently jumplike) growth in the SL intensity accompanied by rapidly increasing absorption of ultrasound in the cavitation zone; and (*4*) saturation of the cavitation zone, at which the SL intensity decreases with increasing ultrasound intensity. Note that only two regimes of cavitation-zone development (precavitation and cavitation) have been discussed in the literature to date [8, 12].

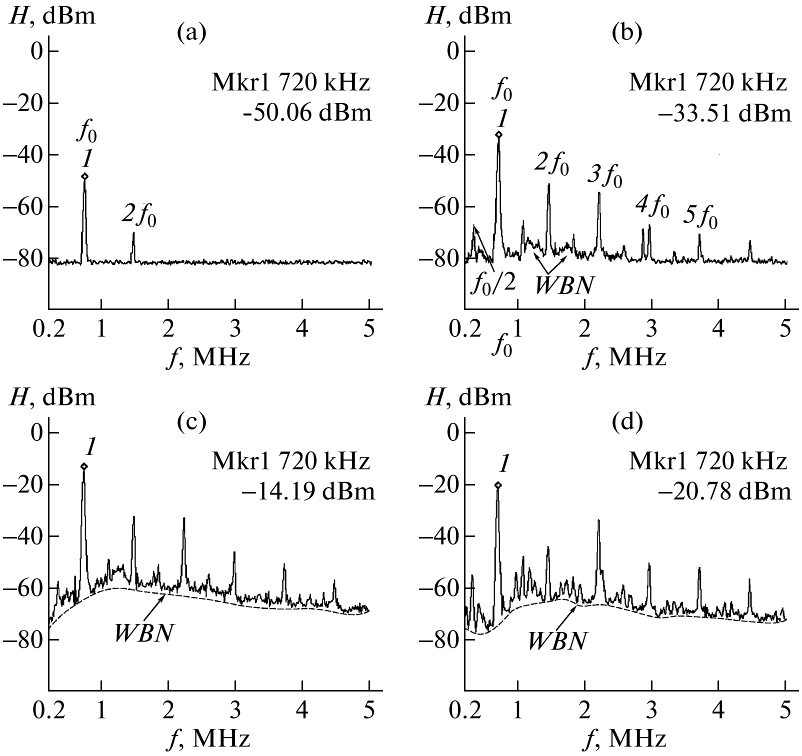

Fig. 2. Spectra of acoustic signal H at various stages of development of the cavitation zone: (a) stage *1*, (b) stage *2*, (c) stage *3*, and (d) stage *4*. Ultrasound pulses with duration $\tau = 3$ ms were generated at a repetition period of $T = 100$ ms; all measurements were performed at a liquid temperature of 21 ± 2°C; peak 1 corresponds to the main resonance frequency of $f_0 = 720$ kHz; the dashed curve shows the wide-band noise (WBN) level.

Figure 2 shows the spectra of cavitation noise for various intensities of ultrasound corresponding to the aforementioned stages of development of the cavitation zone. For ultrasound intensities below the cavitation threshold, the spectrum contains only the main resonance frequency of $f_0 = 720$ kHz (this case is not presented in the figure). The $2f_0$ harmonic appears before the SL onset (Fig. 2a). The corresponding acoustic signal is probably generated due to nonlinear pulsations of the cavitation bubbles with relatively small amplitudes. The low intensity of the first harmonic and the absence of higher harmonics suggests that the volume number density of bubbles in the focal region of the radiator is insignificant and the character of pulsations differs but little from linear.

The appearance of $3f_0$, $4f_0$, and higher harmonics in the acoustic signal spectrum (Fig. 2b) is probably related to the formation of nonlinearly pulsating cavities and a significant increase in their volume concentration (Fig. 1, stage *2*). This is accompanied by the appearance of $f_0/2$ subharmonic and half-integer frequencies $nf_0/2$ ($n = 2, 3, 4, ...$). For ultrasound intensities above the SL threshold (i.e., at stage *2* of cavitation-zone development), higher harmonics nf_0 with $n \geq 5$ also appear in the spectrum. Subsequent increase in the intensity of ultrasound leads to further expansion of the spectrum toward high-frequency components and to the appearance of wide-band noise (WBN).

The third stage is characterized by rapid growth in the intensity of high-frequency harmonics and the WBN component (Fig. 2c). In a regime corresponding to the maximum SL intensity, the WBN intensity also reaches its maximum. At the fourth stage, the spectrum shows a significant increase in the intensity of the $f_0/2$ subharmonic (Fig. 2d) and the appearance of additional peaks on the left and right from $nf_0/2$ frequencies.

An important feature in evolution of the cavitation zone during the passage from stage *3* to *4* is manifested by a decrease in intensity of the main harmonic f_0 despite increasing radiator power. This behavior is indicative of a significant increase in the absorption of ultrasound in the cavitation zone. Assuming (in accordance with [13, 14]) that the subharmonic appears as a result of the pulsation of bubbles with dimensions above the resonant size, the observed increase in its intensity at stage *4* can be interpreted as related to an increase in the concentration of large bubbles and/or in the size of a cavitation cluster (or clusters). The increase in the volume concentration of bubbles in the cavitation zone above an optimum value and the formation of large bubbles are two factors that favor a decrease in the efficiency of conversion and concentration of the ultrasound energy upon collapse of these bubbles [11], which is just what leads to a decrease in the SL intensity at stage *4* of cavitation-zone development.

Thus, the above data show that the spectral characteristics of cavitation noise are substantially different at various stages of development of the cavitation zone. This circumstance allows these sages to be reliably identified.

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SPELL: 1. preamplified