Laser-induced shock-wave-expanded nanobubbles in spherical geometry

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ABSTRACT

The secondary cavitation generation following laser-induced breakdown in aqueous media in spherical geometry, mimicking the geometry of the frontal part of the human eye, was studied. A numerical simulation of the shock wave propagation was performed, yielding peak-pressure maps, correctly predicting the location of the secondary cavitation onset for different shock wave source positions. The comparison between the simulation results and the experiments, performed with a high-precision, multiple-illumination technique, supports the suggested description of the nature of the secondary cavitation onset. It is shown that large transient negative pressures are created at the location of the acoustic image of the shock wave source, which is different from the optical focus. After the passage of the shock wave, abundant secondary cavitation is generated there. Additionally, the existence of an important contributing factor to the reduction of the secondary cavitation threshold is supported by the experimental results, namely the pre-illumination of the water by the breakdown-generating laser pulse, playing a crucial role in conditioning the medium. There is strong experimental evidence of the existence of another mechanism of pre-conditioning the water for the secondary cavitation onset, namely in the form of repetitive negative pressure pulse passage through the same volume, an indication of a possible two- or multiple-stage process.

1. Introduction

After laser-induced breakdown in aqueous media, following the primary expansion of the plasma-induced cavity (bubble) and emission of a shock wave, the following events can in continuation lead to the creation of the so-called secondary cavitation, provided that certain conditions are met.

The conditions for secondary acoustic cavitation inception in pre-illuminated, non-degassed distilled water were studied. This secondary cavitation is triggered by the shock waves that are emitted during the laser generated breakdown and also by the consequent collapse(s) of the corresponding primary cavitation bubble [1]. The initially spherically expanding compressional shock wave, which soon after its emission propagates sonically as a monopolar pressure pulse [2,3], is partially reflected from the concave surface and refocused at a location, different from the breakdown spot [4]. In the process of the acoustic focusing, a nearly spherically converging compression is transformed into a diverging rarefaction with a large negative peak pressure [5,6]. This transient tensile stress causes the existing nanobubbles in the liquid to expand, in the regions where the transient tensile stress is sufficiently large. The primary, optically-induced cavitation, generated at the optical focus [7] is therefore accompanied by a secondary, acoustically-induced cavitation which is triggered near the acoustic focus [8,9]. In general, the two foci are located at different positions which enables spatially as well as temporally separated visualization of the cavitation structures characteristic for each type of generation mechanism.

The above-described secondary cavitation is studied here by comparing the numerical simulation results to experiments that are designed to mimic the transient response of the human eye tissue in the first few hundred microseconds after the introduction of the laser pulse into the eye through an ophthalmic lens. It can be regarded as a simulation of two laser-based procedures: posterior capsulotomy [10] and vitreolysis [11], depending on the position of the optical breakdown. In the actual medical procedure, the ophthalmic lens is brought in contact with the human cornea and serves as a focusing lens for the treatment laser pulses [12]. Using such a lens reduces the spherical optical aberration and thus enables tighter focusing and more precise photo-disruption of the selected tissues, i.e., the opacified posterior capsule in capsulotomy and vitreous strands in vitreolysis. Apart from its primary purpose to serve as the optical element (converging optical lens), it also possesses an adverse property which is generally not very known or at least not discussed extensively. Namely, the ophthalmic lens acts also as...
an acoustic element (concave mirror), refocusing the mechanical energy, carried by the laser generated shock waves, somewhere inside the eye, potentially causing mechanical or chemical damage to tissues. In this way, the treatment that is intended at the location of the optical focus, may bring about the concentration of mechanical energy by acoustic focusing at a different, undesired location, the event and its effects being concealed from the operator’s attention.

The negative phase of an intense pressure transient may induce visible cavitation in the liquid in which it propagates when its value $p$, is comparable to the acoustic cavitation threshold (ACT), the negative pressure $p$, being defined as the negative pressure at which there is 50% probability for the acoustic cavitation occurrence [16,17].

Predictions for the acoustic cavitation threshold (ACT) for homogeneous nucleation in water span from $\approx 120$ MPa [18,19] up to $\approx 192$ MPa [9]. However, the value of the ACT does not depend only on the magnitude and the spatiotemporal distribution of the pressure pulse but also on the conditions of the liquid, such as temperature, pre-sonification, pre-illumination [20], dissolved gas content, contaminants, pH etc. In view of this, one can well understand the comment in [19] that the values of the cavitation pressure threshold in literature are “amazingly scattered, even between similar experiments”. It is generally assumed that the cavitation bubbles begin their growth from the pre-existing microscopic pockets of undissolved gas and/or vapor that are either stabilized on solid surfaces or float freely in the bulk. There is controversy about the existence and nature of stabilized nanobubbles in bulk water, which is currently receiving a lot of attention [20–23].

The experiments in the present research show that the secondary cavitation occurs almost exclusively in the anterior laser cone, i.e. in the volume of water which differs from the rest by the fact that it was previously illuminated by the excitation laser pulse, as observed already before [4,24]. Since then, an unambiguous confirmation that the preconditioning of water in the form of illumination shortly before the passage of the negative pressure transient plays a key role in secondary cavitation bubble generation, came in the work of Rossello et al. [20]. In view of this, together with the results of the shock wave pressure evolution calculation compared to the experimental results, we are able to further confirm the understanding of the phenomena occurring during the first few microseconds after the laser induced bubble formation in eye-like geometry. It can be presumed that the light energy is being deposited inside the bulk of the water either by heating the liquid or by being absorbed by the nonscopic impurities, resulting in local tiny hot spots from which water vapor-filled pockets are formed. But, as pointed out in [20], the exact mechanism is not known. The images of these submicron voids on the camera sensor are much smaller than the camera pixel size. As such, prior to the passage of the negative pressure pulse they are not detected by the visualization technique used in the experiments, but their presence must be assumed, as they must be acting as the seeds for secondary cavitation.

Possible explanations of secondary cavitation initiation in water offered by other authors do not seem to be valid in our case. Their reported cavitation threshold values for the tensile stress amplitude range over a substantial interval, from $-20$ MPa to $-60$ MPa, depending on the conditions of the experiments [10,22,23]. The authors explain that in their case the homogeneous seed population of water vapor microbubbles was produced by the preceding laser breakdowns in the same water tank [25] or, stable gas nanonuclei were present in the bulk water beforehand [9,26]. Interestingly, in the work of Ando et al. [27], where authors made special effort to suppress heterogeneous nucleation, the cavitation threshold pressure for homogeneous nucleation in water was estimated to be $-60$ MPa.

Both explanations fail to describe the situation when the formation of the secondary cavitation predominantly occurs on the laser path, for example when the laser breakdown is generated in water close to its free surface [28,29] and in [20].

Both, in [20] and in our experiments, the reported negative pressure cavitation threshold value is considerably smaller compared to the experiments in [10,22,23], namely $-5.0$ MPa [20] and here estimated to be in the range of $-5$ MPa to $-9$ MPa. Also, in both experiments, the secondary cavitation is observed exclusively in the volume of water which was illuminated by the laser pulse. A similar effect was observed by Supponen et al. [28]. The reported value of the pressure in reference [28] was estimated to be 0.3 MPa, measured 44.5 mm away.

2. Methods

2.1. Multiple illumination technique

The experimental system (Fig. 1) and its principle of operation is similar to the one used in [4]. It consists of an excitation laser (Q-switched Nd:YAG) providing 10 ns laser pulses to initiate optical breakdown inside a water-filled experimental cell, and a highly adaptable laser-diode-based illumination system which is capable of generating any specified train of pulses having variable duration, adjustable energies per pulse, with a variable and adjustable idle time within the train, with a temporal resolution of 12.5 ns [30]. The illumination system is synchronized with an arbitrary external trigger, in this case a high-speed camera. The capabilities of the illumination system are central to the design of the experiment. The setup is used to capture multiple snapshots of the same region at several times during a single exposure.

An ophthalmic lens (Katena, [12]) which is designed to be placed in contact with the human eye in laser-based medical procedures such as capsulotomy or vitreolysis, is here mounted in the wall of the container filled with distilled water.

The shock wave which is created by the plasma during the laser-induced dielectric breakdown at the optical focus is captured at several key instances during its propagation. The highly adjustable illumination system allows capturing the images with very high temporal and spatial resolution during a long exposure time of a single event. The principle is illustrated in Fig. 2, explaining how to interpret the combined images acquired by the multiple illumination technique. In the left-hand column of Fig. 2, the constituent images are presented. The second row in Fig. 2 is the triple exposure image recorded around the time when the reflected pressure wave arrives at the acoustical focus, namely (i) 0.75 μs before, (ii) exactly at the time and (iii) 0.75 μs after. This sequence takes place approximately 10 μs after the initial breakdown. The second image (third row in Fig. 2) is taken 4 μs later than the triple illumination. By then, the wave has moved past the bubble and the secondary cavitation bubbles are already visible at the site of the acoustical focus. The image presented in the bottom row in Fig. 2 is a superposition of four images, generated during a single event, using four illumination pulses of the adaptable illumination system, revealing all the above explained features in a single image.

The collapse of the primary bubble which follows after a few

![Fig. 1. a) Experimental configuration of multiple illumination technique principle and b) a typical train of illumination pulses, used during the acquisition of a single exposure image.](image-url)
hundred microseconds is also accompanied by the emission of the shock wave. In this case, the capabilities of the technique are even more apparent. Namely, in the final image (bottom of second column in Fig. 2) we can see a combination of sharp images of primary bubble at approximately 6 µs after breakdown, the triple exposure around the second focusing at 360 µs and the third image after another 4 µs have passed, revealing the freshly formed (second) secondary cavitation cloud.

2.2. Numerical procedure

A computational model was built to determine the time-dependent pressure field, capturing the propagation, reflection, and conversion of the laser-induced shock wave, to contextually interpret the experimental results described in the next section. The intention was to extend the approach and the procedures described in [6,31], in the search for detailed description of the secondary cavitation onset in the spherical, eye-like geometry.

The model geometry is axisymmetric, consisting of three different media: water, polymethyl methacrylate (PMMA) and air/vacuum as depicted in Fig. 3. The pressure pulse source is on the axis of symmetry, at five different locations, corresponding to possible real situations occurring in various eye treatments.

For the PMMA, which is considered a homogeneous and isotropic linear elastic solid, the formulation includes Navier–Lamé equations from elastodynamics, involving equilibrium equations, strain–displacement relations and constitutive equations of linear elasticity. Water is considered an inviscid and compressible acoustic medium. In our case, the response is assumed to follow a classical linear wave equation extended with a frequency-independent attenuation model. The gov-

Fig. 2. Multiple illumination principle for visualization of secondary cavitation. Below each image the timing of the illumination pulse sequence is indicated. Left column: triple illumination image (second row) around the time when the shock wave is focused after reflection and secondary cavitation cloud (third row), taken 3.76 µs after that (single illumination). Bottom row: events recorded using multiple illumination during a single frame, at approximately the same times as the above two images. Right column: same as the left one, but showing events after the collapse of the primary bubble, several hundreds of microseconds after the initial breakdown. Top row: primary bubble. Bottom row: single exposure image of a single event, illuminated by five corresponding short pulses. The arrows point at the same shock wave, visible at different illumination times. The times of the illumination are (in microseconds): (left column: 10.08, 10.83, 11.58 and 15.34, right column: 6.57, 358.70, 359.45, 360.20 and 364.0).

Fig. 3. Problem geometry. Top: full. Bottom: reduced. The points $a_i$ on the symmetry axis are the five different shock wave source locations (laser dielectric breakdown positions), used in the simulation as well as in the experiment. The gray circles around the sources represent the sizes of the primary cavitation bubbles at the time when the reflected shock wave arrives back. The boundaries of the computational domain are modeled in the simulation as nonreflecting.
The acoustic attenuation is given by:

\[ \Delta p(r, t) = \frac{2\pi}{c} \frac{\partial p(r, t)}{\partial t} - \frac{1}{c^2} \frac{\partial^2 p(r, t)}{\partial t^2} = 0 \]  

(1)

where \( r \) is the spatial coordinate, \( t \) is time, \( \Delta \) is the Laplacian operator, \( c \) is the wave speed in the fluid and \( \kappa \) denotes the acoustic attenuation coefficient. In this formulation, the acoustic losses are accounted for by the mid-term. For instance, the equation can be derived simply by assuming the basic Maxwell’s viscoelastic constitutive equation with bulk modulus and bulk viscosity as parameters in material behaviour [32]. No dissipative effects due to heat transfer mechanisms were assumed.

The interaction between the acoustic and the solid medium is modelled via consistency condition, where the pressure at the acoustic medium boundary equals the normal surface traction loading at the solid medium interface. Except for the water–PMMA interaction, the outer boundaries of water are non-reflecting. The computational domain is discretized by 28 million nodes associated with 10 million axisymmetric acoustic and axisymmetric solid finite elements (FEs), to accurately model spatiotemporal pressure distribution at an instant. Quadratic FE formulation is adopted. The Newmark-beta algorithm is used for time integration, where the maximum time increment is set to 1 ns.

To minimize the computational burden, a reduced geometry (Fig. 3, bottom) is employed, so that the lens is truncated at the cross-section where it is rigidly clamped in the annular surface located at \( z = -2.01 \) mm, marked in the cross-section by two yellow lines. The cut surface is modelled as nonreflecting. In the reduced geometry, the computational volume is a cylinder with a radius of 10 mm. Except for the rigid clamping, where the displacement is set to 0 at all times, the remaining boundaries of the computational volume are nonreflecting.

In the computation, the pressure pulse was assigned 100 ns duration (cca 150 \( \mu \)m length) and peak value at 1 mm from the source to be 60 MPa, both being representative values for typical conditions [33,34]. The pressure pulse temporal profile is shown in Fig. 4.

The resulting pressure pulse has a rise time of 10 ns and full width at half maximum (FWHM) of 30 ns. Such a pressure pulse in the far field has 0.63 mJ of energy, corresponding to the near field shock wave energy of about 4.2 mJ, which in turn is about twice the bubble energy of \( 2 \) mJ [35].

From the acquired pressure field \( p(r, z, t) \), a simulation of the corresponding experimental image using schlieren technique was calculated, emulating the local intensity of light in the image as:

\[ \nabla^2 p \cdot \frac{\partial p}{\partial t} + \frac{\partial^2 p}{\partial r^2} = 0 \]

(2)

A numerical schlieren image is constructed by displaying \( \nabla^2 p \) in a nonlinear scale to expose the weak non-uniformities of the flow density as:

\[ D(x, z, t) = 0.5 \left[ 1 - \text{erf} \left( \frac{\kappa \nabla^2 p}{\text{max}(|\nabla^2 p|)} \right) \right] \]

(3)

with \( \kappa \) chosen in such a way that relevant features are presented adequately (here \( \kappa = 100 \)). \( D \) is then displayed in grayscale, with values between 0 (black) and 1 (white). To a very good approximation, the density of water \( \rho_1 \) and its refractive index \( n_1 \) vary linearly with the overpressure \( p \) in the pressure range of interest \((-10^2 \text{ MPa} \text{ to } 10^2 \text{ MPa})\), thus the same numerical schlieren image \( D \) is obtained if \( p \) is substituted by \( \rho_1 \) or \( n_1 \).

3. Results and discussion

3.1. Computational results

The positions of laser induced breakdowns which act as shock wave point-sources are located on the system optical axis (Fig. 3), at \( a_1 = 6.48 \) mm, \( a_2 = 8.06 \) mm, \( a_3 = 10.13 \) mm, \( a_4 = 11.06 \) mm and \( a_5 = 14.25 \) mm. The focal length of the lens when it is acting as the acoustical mirror is \( 8.06 \text{ mm, } a_1 = 11.06 \text{ mm and } a_5 = 14.25 \text{ mm.} \)

The animation (Video 1) shows the simulation results of the pressure field evolution for the case when the source of the pressure pulse is positioned at point \( a_5 \), that is 10.13 mm from the lens vertex and approximately 2 mm from the lens center of curvature (8.1 mm from the lens vertex). A cross-section containing the cylindrical symmetry axis of the system is shown in the animation. Positive pressure areas are red, while blue denotes negative pressure. The first part of the propagation from the point source is spherically symmetric, outbound in radial direction, uniform and uneventful and is not shown in the animation, which follows the pulse propagation and evolution from the time 6 \( \mu \)s from the creation onwards, when the reflection of the wave from the optical lens (here acting as an acoustical mirror) begins.

When the compressional, monopolar pulse reaches the lens (PMMA), which has higher acoustic impedance than water, it is reflected as a compression, except for the effects at the edge. The part of the wave transmitted to PMMA is not visualized in the animation video. Apart from the details of the wave scattering pattern at the somewhat complex structure of the rim of the lens, which is not of interest in this work, the reflected pulse is comprised of the central part of the wave and the toroidally shaped edge wave.

The structure of the reflected wave is similar to and in accordance with the shape of the acoustic field of a pulsed circular piston [36]. Here, the shape of the central part of the reflected wave is slightly modified in comparison to the flat piston case, because of the curvature of the reflecting surface [37]. Nevertheless, the main features can still be well recognized: as in the case of the flat piston the reflected wave consists of (i) the central portion, propagating in the cylinder representing the geometrical shadow of the reflecting surface and (ii) the toroidally shaped edge wave, emanating from the rim of the lens. The outer part of the edge wave has the same phase as the central part (here compression, denoted red in the animation and in all the figures), while the inner part of the edge wave has opposite phase (here rarefaction, blue). Because of the concavely curved boundary, both parts of the reflected wave, the central part and the edge wave, are focused. In the particular case, presented in the animation, the reflected wave focuses at approximately 6.6 mm from the lens (mirror) vertex. At that point, the central part of the wave undergoes a Gouy phase shift, a feature which is characteristic of waves passing through a focus [38,39]. In the present case, the Gouy phase shift is manifested as a transition from a monopolar compression to a monopolar rarefaction pulse [37], indicating a phase shift of \( \pi \), characteristic for spherical wave focusing. On the contrary, the edge wave continues unaffected after its passage over the symmetry axis, indicating its diverging nature. The moment of phase transition is not
clearly observable in the animation, because of the simultaneous passage of the edge wave through the same region. But the transformed (phase shifted) central part, now a rarefaction, can be clearly resolved as a feature of the edge wave through the same region. But the transformed -

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tion are calculated as:

\[
\Omega = 2\pi(1 - \cos(\theta))
\]

where \(\theta\) is the plane angle subtended by the lens, as viewed from the source. The relative values of the solid angles for the five source locations are given in Table 1 (third column), denoted \(\xi\) and normalized so that full solid angle is 1 (unity).

The pressure in a wave spherically propagating from a point source falls off with distance as \(1/r\) and the energy as \(1/r^2\). The distances that each wave travels from its source \(a_i\) to the lens (mirror) are different, so the portions of the initial energy at the time of the reflection are different for each of the five cases. The corresponding relative factors of \(1/a_i^2\) on the symmetry axis are given in the fourth column in Table 1. Multiplied values \(\xi_i \times 1/a_i^2\) account for both factors, the relative solid angle and the relative propagation distance (on the axis), yielding estimated relative energy upon reflection for the five different source locations. In the sixth column this value is normalized to the smallest of the five energies (case of source at \(a_5\) which is farthest of the five sources). The wave pressure amplitude is proportional to the square root of the energy (seventh column in Table 1). These ratios give a general orientation for the expected relative pressure amplitudes in the five cases, provided that other conditions are the same. For example, for the same initial energy input, in the case of source at point \(a_1\) the overall maximum pressure amplitude upon reflection at the lens is 4.35 times larger than for the case of source at \(a_5\). Material attenuation was not considered. The relative solid angle and the expected relative pressure amplitude are presented graphically in Fig. 7 (solid curve).

The detailed shape of the transient pressure field, presented in the contour maps of Fig. 5 is influenced by these general geometrical considerations, but at the same time it is a consequence of other effects such as focusing, conditions at the boundaries, and most of all, the presence of the primary cavitation bubble in the shock wave path. The bubble is a large obstacle for the wave, being roughly an order of magnitude larger than the dominating wavelength dimension. The computed maximum pressure amplitudes presented in Fig. 5 and Fig. 6 should be viewed in this light and the results interpreted accordingly. The estimation presented in Table 1 and Fig. 7 gives the general orientation as to what is the relative available energy for the creation of the secondary cavitation bubbles.

The five individual data points in Fig. 7 represent the negative pressure amplitudes for shock wave sources at points \(a_i\) as computed in the simulation. For ease of comparison, both types of data are normalized with respect to the case of the source at point \(a_5\). As expected, the relative pressure amplitude is substantially affected when the source is located closer to the lens, in the vicinity of the center of the lens curvature (sources at \(a_1\) and \(a_2\)). In these two cases a lot of the shock wave energy is reflected by the primary bubble in directions other than the location of the acoustic image of the source. As the source is positioned farther away from the lens, the amplitude reduction with distance approaches the expected course.

3.2. Experimental results and discussion

All the images in Fig. 8-11 are multiple illumination single exposure photographic schlieren images – snapshots at various times after the optical breakdown was initiated at five different locations \(a_i\) from the lens vertex. The black rectangle on the left side in every image is the lens. Only the edge (rim) can be observed, while the curved surface is hidden from view. The important feature, namely the shock wave reflecting surface, is depicted in Fig. 9 by the black arcs on the far left of each individual image, to indicate its position, its curvature and the overall scale. The position of the optical focus is determined by additional optics mounted in front of the lens which is built into the wall of the container. At the position of the optical focus, the primary bubble (black circle) is created by the laser generated breakdown. It acts as the source of the shock wave pressure pulse which propagates spherically
(caption on next page)
Fig. 5. Numerical simulation results: largest pressure at a given point at any time after the reflection of the shock wave from the lens (red: positive, blue: negative), for five different source positions. Center: the contour field lines. Left column: largest negative pressures on the axis. Right column: largest positive pressures on the axis. The white circle represents the size of the primary bubble at the time the returned wave is reflecting from it. The closer the primary bubble to the reflective surface, the more it influences the transient pressure field. The contour of the negative pressure field tends to have a spike-shaped projection towards the reflecting surface, while the positive pressure fields have the spike in the opposite direction. This shape originates from the edge wave inner and outer parts, propagating and focusing in this concave geometry, as can be understood by observing the animation of the wave propagation (Video 1). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Fig. 6. The largest negative pressures on the axis at any time after the shock wave reflection, for the five different source locations. Where the reflected wave encounters the primary bubble, the pressure is 0. This occurs at different intervals of z coordinates for each case.

from the site in all directions.

Direct comparison between the simulation results and the experiment can be seen in Fig. 8, showing the calculated pressure (left column) and numerical schlieren image (right column) for the source located at point a5, at approximately the same times as the observed volume was illuminated in the experiment, during a single exposure (Fig. 8, bottom). The technique enables the recording of many waves, associated with the same event, in a single experimental image, which in this way conveys more information than individual images would. Because the waves are well spatially resolved, we can directly observe their relative positions and other characteristics, for example, the creation of secondary bubbles.

The top row images in Fig. 8 are showing the calculated shock wave after the reflection from the concave surface (located to the left, not drawn). The central part of the wave, propagating along the axis, is compression (red). At the sides two arcs can be observed, propagating at an angle towards the axis and belonging to the edge wave. In the second row, the central, converging part of the wave has reached its focal point and is thus very small. What is seen is the edge wave, with its outer part being a compression and the inner part a rarefaction. It should be pointed out again, that the pictures are the cross-sectional representations of the actual spherical (3D) focusing. Since the edge wave is propagating at an angle with respect to the symmetry axis, in the third row of Fig. 8 its rarefaction part is now on the outside and the compression is on the inside, near the axis, overtaking the central part. After having passed the focus, the central part is now Gouy phase-shifted to become a rarefaction and is spatially connected to the rarefaction portion of the edge wave. The evolution of the wave shape can best be understood observing the animated version in Video 1. The images in the fourth row are of a situation a few microseconds later, when the amplitude of the central part is greatly diminished due to the scattering from the bubble and spreading (red circle). The same applies to the compression part of the edge wave, while the rarefaction part of the edge wave did not encounter the bubble at all.

The same kind of images as the one explained in detail in Fig. 8 are presented in Fig. 9 for five different source locations: after the primary breakdown (left column) and after collapse of the primary bubble (right column). The distance from the lens vertex increases from a1 in the top row to a5 in the bottom row.

The radius of curvature of the lens from which the shock wave is reflected is 8.1 mm, so the pressure wave in the second row of Fig. 9, where the source is 8 mm from the lens vertex, is reflected approximately back to the same position, where the primary bubble has grown in the meantime. As expected, in the top row (source at a1) the reflected wave is focused to the right of the bubble position, and in the bottom

![Fig. 7. Estimation of the relative portion of the solid angle subtended by the reflecting surface of the lens (dashed curve), the relative reflected shock wave pressure amplitude (solid curve) and the computed pressure amplitudes (individual orange data points), representing the simulation results (see Fig. 6) for the five source positions. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)](image)

**Table 1**

| Source position a1 | Distance from the lens [mm] | Portion of the total solid angle $a_i$ | $1/a_i^2$ [$10^{-8}$m$^{-2}$] | $s_i \times 1/a_i^2$ [$10^{-5}$m$^{-2}$] | Relative reflected energy | Relative reflected pressure (p $\propto E^{1/2}$) | Acoustic image position $q_i$ |
|-------------------|-----------------------------|---------------------------------------|-------------------------------|-------------------------------|------------------------|-----------------------------|------------------------------|
| a1                | 6.48                        | 0.307                                 | 2.38                          | 7.32                          | 18.90                  | 4.35                        | 10.80                        |
| a2                | 8.06                        | 0.227                                 | 1.54                          | 3.50                          | 9.03                   | 3.00                        | 8.14                         |
| a3                | 10.13                       | 0.154                                 | 0.97                          | 1.50                          | 3.88                   | 1.97                        | 6.75                         |
| a4                | 11.06                       | 0.131                                 | 0.82                          | 1.07                          | 2.76                   | 1.66                        | 6.39                         |
| a5                | 14.25                       | 0.079                                 | 0.49                          | 0.39                          | 1                      | 1                           | 5.66                         |
three images the acoustic image of the source lies between the lens and the bubble.

The general positions of the acoustic images of the sources (that is, the positions where the reflected pressure waves are focused) are in accordance with the general expressions known from the geometrical optics, namely the position $b$ of the image formed by a concave mirror having curvature radius $R$ is calculated as $b = \frac{2}{R} - \frac{1}{a}$, where $a$ is the location of the object from the mirror. A more detailed, non-paraxial analysis of geometric focusing that includes spherical acoustic aberration can be found in [1]. The generalized result of such analysis says that the farthest the source position is from the center of curvature of the lens, the longest the focal interval due to spherical aberration. This means that the acoustic focus is sharpest when the source is closest to the lens’s center of curvature and thus more acoustical energy is directed to a smaller volume.

Quantitatively, the present numerical simulation yields the same result: the pressure peaks on the axis broaden spatially (Fig. 6) for cases from source position $a_1$ to $a_5$. The cases $a_1$ and $a_2$ should be excluded from this comparison since the influence of the bubble is substantial. Data for the acoustic image positions for the five source locations used in the experiment is given in the last column of Table 1. Using the rulers in Fig. 9, it can be verified that the acoustic images positions, marked with yellow arrows, are in all cases in accordance with the calculation. The source positions are marked by orange arrows. Furthermore, the same locations are predicted by the simulations results: the positions of the peaks of large pressure in Fig. 6 are in accordance with the experimental results and with the simplified calculation (Table 1, last column).

The secondary cavitation bubbles which are regarded to be brought on by the tensile stress of the propagating rarefaction are not formed yet when the shock wave is near its focal point (after 10 µs and again 360 µs after the breakdown-generating laser pulse) but are in both cases clearly observable soon after the wave has passed the bubble. This can be verified by inspecting the individual images in the second and the third row of Fig. 2, which are approximately 4 µs apart from one another.

We expect the secondary cavitation to be most abundant where the negative pressure amplitude is largest. In general, this occurs in the vicinity of the position of the acoustical image of the source (in accordance with the estimation in Table 1) but for details of each particular case one should refer to the calculated pressure field in Fig. 5. According to the calculated pressure contour maps, the negative pressure amplitude is highest when the shock wave source is positioned in $a_1$ and $a_2$, also in $a_4$. Source position $a_2$ is namely characterized by distinct interference and scattering of the wave by the primary bubble, while in the case $a_3$ the source is already quite far. One would therefore expect approximately equal amount of secondary cavitation bubbles in cases $a_1$, $a_3$ and $a_4$, but in the topmost row of Fig. 9, namely case $a_1$, the expected secondary cavitation cloud is missing noticeably. The possible reason is discussed below.

On the other hand, the absence of the secondary cavitation bubbles in the first column, second row of Fig. 9 (source $a_2$) is not surprising and can be explained by the position of the breakdown bubble at the location of the acoustic image of the source, interfering with the creation of the secondary cavitation bubbles, which is expected to take place there. This is also reflected in the fact that, for the same reason, the predicted pressure contour maps do not exhibit strong pressure amplitude there, compared to other source positions. In this respect the source position (2) differs from other cases. Therefore, for the cases $a_1$ and $a_2$ strongest interference by the bubble presence is observed.

For the source positions $a_3$ to $a_5$, the secondary cavitation bubbles appear in the vicinity of the acoustic image (Table 1 and Fig. 9), where also the contour maps show largest negative pressures. The number of the secondary bubbles seems to be roughly proportional to the calculated amplitude of the negative pressure.

The marked inconsistency, namely the remarkable absence of secondary cavitation bubbles in the $a_2$ case despite large tensile stress amplitude ($\sim 70$ MPa) should be commented. The absence of secondary...
cavitation bubbles contradicts the expected and apparently established proportionality between the negative pressure and the amount of secondary bubbles. It should be noted that, except maybe for a very thin volume near the axis, the calculated values of the pressures are roughly an order of magnitude smaller than the values, reported in most other works to be the threshold values for the cavitation onset, in different conditions, namely from −6 MPa up to −60 MPa, most of them around 20–30 MPa. So instead of asking, why there are secondary bubbles missing in the case, one should be wondering why the bubbles in the cases a3 to a5 are observed at all! We believe that another effect should be considered, namely the fact that the volume of the water under consideration was illuminated by the laser pulse very shortly before it experienced the passage of the tensile transient. As mentioned in the introduction, it was confirmed that such pre-illumination of the water is crucial for the creation of the cavitation bubbles [20] at such relatively small amplitude pressures. In [20] the illumination creates seed bubbles that are smaller than 500 nm in diameter and with a lifetime of typically less than 1 ms. The presence of invisible nanobubbles which are expanded by the tensile pressure wave importantly lowers the apparent cavitation threshold. Illumination intensity in [20] was reported to be 500 times smaller than the cavitation threshold. The description of our findings is consistent with that scenario. The illumination in our case is provided by the cavitation-generating laser pulse itself. It is incident from the left, illuminating the water between the lens and the breakdown site, its energy finally being used up in the dielectric breakdown and subsequent phenomena, like the creation of plasma, the bubble and the shock wave. The water to the right of the optical focus (bubble position) is therefore not illuminated by the laser pulse. This explains why in the case of the source at point a1 there is little secondary cavitation present despite largest negative pressure amplitudes of the shock wave.

It remains to clarify why, contradicting the above explanation, there still are a few bubbles observed to the right of the optical focus in the case of source position at point a1 (top row of Fig. 9). Most probably the reason lies in the nature of breakdown formation: when a laser pulse is focused to create a dielectric breakdown, in absence of other hot spots in the pulse spatial profile, the threshold for breakdown is first exceeded at

Fig. 9. Multiple illumination images of the shock wave propagation and secondary cavitation formation for five different source locations: after the primary breakdown (left) and after collapse (right). The shock wave source position from the lens vertex increases from a1 in the top row to a5 in the bottom row. The blue thin lines depict the formation of the acoustic image of the source according to the paraxial geometrical considerations for two cases. The positions of the source a1 are marked with orange arrows, while the acoustic image positions b1 are marked with yellow arrows. The ruler is in millimeters. C (blue point) is the center of curvature, while $\frac{1}{2}C = F_a$ (white point) is the acoustic focal point. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Fig. 10. The spatial distribution of illumination (red shaded area) and the volume where, by simulation results, the absolute value of the negative pressure exceeds 5 MPa (10 MPa). Left: after breakdown; right: after collapse. While the shape of the pressure field contour areas is expected to be the same in both cases, the pressure values are obviously smaller during the second passage (after bubble collapse). Source position: a3. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)
the smallest cross section of the laser beam, at the waist. The light absorption of the plasma is high, resulting in plasma growth towards the incoming light (to the left in the images), creating the so-called moving breakdown distributed shielding model [40,41]. The regions downstream are effectively shielded from the incoming light by the plasma. However, since this shielding is spatially and temporally distributed, a portion of the laser pulse energy is still transmitted, reportedly for tightly focused 6 ns laser pulses (similar to the ones used here) the transmission at pulse energies several times larger than the breakdown threshold is still a few percent (i.e. 2.8% at 6.1 mJ of pulse energy), and around 20% for energies that are one to four times larger than the threshold energy [41]. The volume to the right of the optical focus in the top row of Fig. 9 can therefore still be regarded as being illuminated, but to a significantly lesser extent than the regions to the left of the optical focus. This creates conditions for secondary cavitation formation, consistent with the above explanation. Also, according to the simulation results, in a very thin volume (cca. 0.5 mm diameter) near the axis, the negative peak pressure even exceeds −60 MPa which is considered sufficient to cause cavitation on its own, without prior illumination.

After the first expansion, the primary bubble collapses and another shock wave is emitted upon this collapse, in all aspects similar to the first one [2]. The images in the right column of Fig. 9 are images of the pressure transient propagation after the bubble collapse, as the one illustrated in Fig. 2 (right column), showing also the primary bubble image. The recorded events take place a few hundred microseconds after the creation of the first bubble. The whole description of the different cases for the different source positions can be repeated along the same lines. Namely the pressure transient propagates in a similar fashion as the first one, as expected, and the observed phenomena follow suit.

Besides the fact that the images after the collapse of the primary bubble very much resemble the first case (after breakdown), two additional general observations are worth mentioning: (i) the bubble after collapse consistently grows a little closer the boundary (length) and (ii) the new secondary cavitation cloud appears to be more prominent than the original one. The markedly larger quantity of the freshly created secondary bubbles is generated in approximately the same volume as after breakdown and in all the cases under consideration. This was briefly observed already before [1], namely that the dominant secondary cavitation cloud is not generated by the passage of the first shock wave, but rather by the one released during the first collapse of the main bubble.

As can be confirmed by inspecting the constituting single images (Fig. 2), there are no residual bubbles visible at the time of the passage of the new, second shock wave, which takes place a few hundred microseconds after the passage of first one, but the fresh secondary bubbles appear immediately (a few microseconds) after that second shock wave passage. This means that there is no (additional) illumination immediately prior to this shock wave passage. In the work by Rosello et al. [20], the lifetime of the seed bubbles that are supposedly created by the laser light, is up to 1 ms and is larger for larger bubbles [4]. It is therefore not unexpected to have the seed bubbles still present after approximately half a millisecond, considering additionally that the intensity of the pre-illuminating light pulse in our case is at least an order of magnitude larger. However, the larger quantity of the secondary cavitation bubbles after the second shock wave cannot be explained by this. We believe that a possible mechanism is another pre-conditioning of the observed volume, this time by the first negative pressure passage. In a sense, this second expansion of the secondary cavitation bubbles can therefore be regarded as a two-stage process. The first negative pressure transient expands the seed nanobubbles to a certain volume – a fraction of these bubbles, previously all smaller than one image pixel, can now be observed. The second shock wave, after a few hundred microseconds, continues the expansion of the same bubbles, of which many now have a larger initial volume and, as a consequence, more of them grow to the observable size for this experiment. With this, a lot of new questions open about the secondary cavitation cloud bubble, apart from its location, namely about the bubble size distribution, its evolution, whether the bubbles are stable or quasi-stable within this volume etc.

The spatial distribution of both the conditions that presumably contribute to the lowering of the threshold for secondary cavitation is schematically depicted in Fig. 10 (left: after breakdown, right: after collapse), for the case of the shock wave source in position a. Namely, the conical illuminated volume of water (red shaded area in Fig. 10) and the volume where the absolute value of the negative pressure exceeds 5 MPa (10 MPa) overlap exactly in the volume where most of the secondary cavitation cloud is created. By comparison of the two images in Fig. 10 it is evident that, although it experiences identical propagation characteristics, the shock wave after bubble collapse is considerably weaker than after breakdown. This is expected, since the available energy is dissipated through many channels. But, it should be mentioned again that despite this fact, surprisingly the intensity of the secondary cavitation cloud is noticeably stronger, suggesting the presence of another contributing factor, as already discussed above.

The value of the laser pulse intensity was not considered in detail and its role not examined any further, other than what was already mentioned: that in the volume of interest it exceeds the reported values for more than an order of magnitude and most likely acts as the cavitation threshold lowering factor.

Finally, the (micro)location and the shape of the secondary bubble cloud is observed, in combination with the experimental results at different excitation laser pulse energies. Both, the illumination and the

![Image](image-url)
The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability
Data will be made available on request.

Appendix A. Supplementary data
Supplementary data to this article can be found online at https://doi.org/10.1016/j.ultrasono.2021.106160.

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