1. Introduction

The phenomenon of laser-induced cavitation is well known [1], but still attracts attention due to the complexity of the processes, accompanying a cavitation bubble evolution. The cavitation bubble’s rapid growth and collapse can cause shock waves, rapid jets and sonoluminescence [2]. The aggressive nature of the laser-induced cavitation has found a broad range of applications, such as cell lysis [3], cell membrane poration [4] and ocular surgery [5]. Cavitation is used for mixing, pumping, switching and moving objects in microfluids. The laser-induced bubbles are used for the creation of highly focused supersonic microjets. Due to their excellent controllability, high velocity and relatively low power requirements, these jets are an attractive option for a needle-free drug injection [6].

Briefly, the physics of ultrashort high intensity laser pulse propagation in water is as follows. A femtosecond laser pulse with an even microjoule level of energy tightly focused into the bulk of the transparent dielectric leads to extreme intensity ($I \sim 10^{13}$ W cm$^{-2}$) in the microvolume and plasma formation (mean energy of electrons $\sim 10$ eV, electron density up to $10^{20}$ cm$^{-3}$). The nonlinear medium properties, laser beam parameters and focusing geometry have a violent effect on the plasma density distribution [7]. On the picosecond time scale the energy transferred from the plasma electrons to the medium leads to the generation of a thin vapor layer, which consequently transforms into a cavitation bubble [8]. Just after plasma recombination the cavitation bubble replicates the plasma shape, thus the initial spatial distribution of the laser intensity in the medium defines the spatial and temporal evolution of the cavitation bubble [8]. The Rayleigh model (or its more correct modifications) describes the dynamics of isolated laser-induced cavitation

\[
\rho \ddot{R} + \left( \frac{3}{2} \right) \rho \dot{R}^2 = p_i - p_o,
\]

where $R$ is the radius of the cavitation bubble, $p_i$ is the pressure inside the bubble and $p_o$ is the outer pressure [1, 7, 9–11]. The model can also be applied to cylindrical or semi-spherical cavitation bubbles with some simple modifications [9, 12].

---

Dynamics of multiple bubbles, excited by a femtosecond filament in water

F V Potemkin and E I Mareev

Faculty of Physics and International Laser Center M.V. Lomonosov Moscow State University, Leninskie Gory, bld.1/62, 119991, Moscow, Russia

E-mail: potemkin@automationlabs.ru

Received 15 September 2014, revised 19 November 2014
Accepted for publication 23 November 2014
Published 11 December 2014

Abstract

Using shadow photography we observed the evolution of multiple cavitation bubbles, excited by a femtosecond laser pulse in water, up to the microsecond time scale. In the tight focusing geometry a single filament is formed. The filament becomes the center of the cavitation region formation. When aberrations were added to the optical scheme, aberration hot spots along the filament axis are formed. At high energies (more than 40 $\mu$J) the filaments in the aberration hot spots are fired. Thereby a complex pattern of the cavitation bubbles is created. The bubbles can be isolated from each other or can form an exotic ‘drop-shaped’ cavitation region, whose evolution at the end of its ‘life’, before the final collapse, contains the jet emission. The dynamics of the cavitation pattern were investigated from the pulse energy and the focusing. An increase of the numerical aperture of the focusing optics leads to an increase of the cavitation area length. A strong interaction between the bubbles was also found. This leads to a significant change in the bubbles’ evolution, which is not yet in accordance with the Rayleigh model.

Keywords: femtosecond filament, laser-induced cavitation, aberrations, tight focusing

(Some figures may appear in colour only in the online journal)
But when two or more bubbles begin to interact with each other, or when the shape is strongly non-spherical, the cavitation bubbles’ evolution considerably differs from the one described by the Rayleigh model [6, 12, 13].

Laser pulses with power above the critical level lead to filament formation in the medium and the self-channeling of the laser radiation, which limits the intensity, the plasma electron density and the energy transfer to the medium [14]. The high linear absorption, tight focusing (NA > 0.3) and supercritical power enables the production of the unique regime, when one continuous plasma channel is formed. The plasma channel generates multiple overlapped cavitation bubbles, which construct one cylindrical cavitation area. There are several works on the thermodynamics and the hydrodynamics of the laser filament in the microsecond time scale in gases [15–19], but, to our knowledge, there are no works describing the cavitation bubble formation stimulated by the fs-filament. It is interesting to point out that the fundamental wavelength of the Cr:Forsterite laser (1240 nm), which was used in our experiments, lies within the peak of the water absorption spectra [20]. This peak is formed by the superposition of three vibration modes of the water molecule. Thereby the strong resonant absorption, the supercritical power regime and the tight focusing sufficiently complicate the processes of the laser-matter interaction, adding new features to the mechanical post-effects and dramatically revealing the theoretical description of the processes [12, 14]. The next thing affecting the plasma distribution is the aberrations, which play a significant role especially in the tight focusing geometry [21]. Such aberrations can create additional maxima in the intensity distribution and decrease the energy in the focal spot [21, 22]. In our case it leads to the generation of the ‘drop-shaped’ cavitation area, the evolution of which consists of a strong jet emission at the end of the bubble life. It is principal to emphasize now that there is no complete theory of laser energy redistribution in solids taking into account the processes of laser filamentation, energy transfer from excited electrons to the ion core and subsequent laser-induced shock wave launching and cavitation bubble generation [23]. Thus, our studies provide new information that will help in the development of theoretical approaches in this area, which certainly makes them stand out and also makes them extremely relevant.

2. Experimental setup

To observe the dynamics of cavitation bubbles on the microsecond timescale, a shadow photography technique was applied. In this technique the probe pulse passes through the sample and creates the uniform illumination on the CCD camera. The perturbations in the refractive index, induced by the pump pulse, act as scattering centers and are seen as dark areas on the CCD matrix. The experimental setup is sketched in figure 1. The second harmonic of the Nd:YAG nanosecond laser (the wavelength is 532 nm, the pulse duration 10 ns, the repetition rate is 10 Hz) was used as a probe pulse.

The cavitation bubbles were excited by a femtosecond filament, which was produced by a Cr:Forsterite femtosecond pulse (the wavelength is 1240nm, the pulse duration is about 140 fs, the intensity contrast is about 10^7, the repetition rate is 10 Hz). The energy of the pump pulse can be varied up to 190 μJ, corresponding to the peak power of 900 MW (taking into account a linear absorption in water), which is approximately one hundred times above the critical power for water. The pump laser pulse was focused into the water cell, creating plasma, shock waves and a sequence of cavitation bubbles. The plasma radiation and the probe beam, which was scattered on the diffuser plate, were collected on the CCD matrix by the microscopic objective. The time delay between the pump and the probe pulses was controlled electronically by the delay generator. It could be electronically tuned up to 80 μs with a 10 ns step. The camera acquired pictures with a 10 Hz frame rate and collected all the illumination between the laser pulses. In the absence of the probe pulse only plasma radiation could be observed. When the probe pulse passes through the water cell, the classical shadow pictures can be made. The plasma radiation could still be observed in the center of the dark area, which corresponds to the cavitation area. Outside the dark region the illumination was much greater than the plasma luminesce and it couldn’t be observed. The light emitted from the plasma gives information about the plasma distribution and the filament location and size.

The experiments were carried out with two sets of focusing parameters: a tight focusing with a numerical aperture NA = 0.4 and a focal length of 3.3 mm (lens L1- Philips CAY033), and a loose focusing with an effective numerical aperture NA = 0.3 and a focal length of 8 mm (lens L2- Rochester A240TM-C). Turning the focusing lens 180 degrees with respect to the laser radiation dramatically changed the optical path of the beam passing through the lens, and created spherical aberrations in the scheme. In order to characterize the focusing conditions the experiments were carried out in air and in water. When the aspheric lens L1 was placed in air a bright spark 4 μm in diameter and about 30 μm in length was observed, which corresponds well to the estimates of the diffraction limited focal spot size. No distortion of the plasma shape was observed indicating the absence of aberrations with the experimental scheme resolution [22]. In water a plasma channel (length greater than 100 μm) was created instead. To avoid aberrations in the other cases, the aspheric lens was placed inside the water cell.

3. Results and discussions

When an ultrashort laser pulse with a peak power much greater than the critical power propagates through a medium, a dynamical balance between the Kerr self-focusing, plasma defocusing and diffraction occurs, and a laser filament is fired [14]. Instead of collimated and loosely focused beams in the tight focusing geometry the laser intensity in the focal spot can reach a value up to 10^{15} W cm^{-2} (it is an upper limit of a rough estimate). Accordingly the electron density can reach a value of \( n_{el} = m_0 \omega_0^2/4 \pi e^2 \approx 7.3 \times 10^{20} \text{ cm}^{-3} \) and the energy of the plasma electrons is sufficiently larger than in other cases [24]. In our conditions the energy is localized in the microvolume
with 4 µm in diameter. As a result only a single filament can be formed. To determine the contribution of the processes taking place during the filament propagation (plasma defocusing, Kerr self-focusing and diffraction) simple estimates can be made. The length of the self-focusing can be calculated as \( L_{sf} = \frac{\lambda}{2\pi n_2 I} = 1 \mu m \) \((n_2 = 1.6 \times 10^{-16} \text{cm}^2 \text{W}^{-1})\), the length scale for the plasma defocusing \( L_{defoc} = L_{pl} n_{at} n_e = n_0\lambda n_{cr}/\pi n_e = 5 \mu m \) and the diffraction length is a Rayleigh length, which is about 15 µm [14]. Therefore the Kerr self-focusing does not allow the laser radiation to leave the optical axis and one continuous filament with an approximately uniform distribution of the electron concentration along the optical axis can be formed [25]. This can be confirmed by the fact that the diameter of the cavitation bubble is uniform along the filament axis, because these diameters are proportional to the plasma electrons’ energy (see figure 2). When enough energy dissipates through the linear and non-linear absorption, the filament stops.

We compared the results obtained with different focusing lenses and found that greater focal length leads to greater filament length. For example, when the lens L1 was used, the filament length was about 1.5 times greater than in the case of the lens L2. Two main processes lead to this fact. The first process is the linear absorption (the distance between the lens and the geometrical focus is greater in the case of the lens L2). The second process is the intensity decrease in the focal plane of the lens L2 in comparison with the lens L1, therefore the Kerr self-focusing is smaller and can’t efficiently keep energy near the optical axis. The filament
location is practically indistinguishable from the cavitation bubble region, because the bubble formation is the result of water evaporation by laser-induced plasma. The filament length has a logarithmic dependence on the laser pulse energy (see figure 3(a) [26]). If loose focusing with the same power of laser radiation is used, the filament breaks into multiple filaments [14], because the role of diffraction is much smaller than in the case of the tight focusing. In this case cavitation bubbles can’t be formed, because there is not enough energy localization characterized by the electron density not exceeding $3 \times 10^{18} \text{ cm}^{-3}$ [27]. The authors do not exclude that there could be nm-size cavitation bubbles, which are much smaller than the resolution of the experimental setup (about $2 \mu\text{m}$ [28]).

When the aberrations were added to the optical scheme, the cavitation bubbles were formed in hot spots, which are located in the aberration foci. When the laser radiation was focused by the L2, the cavitation bubbles in the hot spots are sufficiently separated from each other (see figure 4). These bubbles are initially spherical, but with the energy increase their shape becomes closer to a cylindrical one. The shape of the bubble, as was mentioned above, strongly depends on the initial plasma distribution. The energy increase leads to filament formation in each hot spot. The length of the filament is a logarithm of the laser pulse energy, therefore the length of each cavitation bubble depends logarithmically on the laser energy (see figure 3(b)). At high energies (more than $40 \mu\text{J}$) the diameter of neighbor bubbles flattens, therefore the energy transferred to each bubble is equal and the intensity in each point of the cavitation area isn’t changed along the filament, due to the intensity clamping [29]. When tighter focusing geometry is used (see figure 5), the role of the Kerr self-focusing is less than the diffraction, therefore the filament can be fired only in the first bubbles
at high intensities (more than $10^{13}$ W cm$^{-2}$). In other cases the cavitation bubbles have a spherical shape. Now let us concentrate on the cavitation region evolution. Just after formation the cavitation region replicates the shape of the laser-induced plasma. Then the cavitation region rapidly grows. The growth of the cavitation region’s diameter is in good agreement with the Rayleigh model. After reaching the maximal size it starts to collapse (see figure 2), because the pressure inside the cavitation region is much smaller than the outer pressure. The energy, conserved in the cavitation bubble is determined by the bubble maximal diameter $E \sim D^3$ [1].

When the bubble pattern was created instead of one spherical bubble, the evolution of the cavitation area becomes more complicated. The collapse of the cavitation region induced by laser radiation, which is focused by the L2 lens, starts at the outer bubbles (see figures 2 and 4). The laser pulse which propagates in an initially isotropic medium breaks this symmetry and creates the preferred direction coinciding with the optical axis. Therefore the speed of collapse along the optical axis is larger than in the perpendicular direction. The rapid collapse of the outer bubbles also leads to the momentum and energy flux, which is forwarded to the center of the cavitation region (see figure 5), which significantly increases the speed of the bubble collapse. This flux is symmetric about the center of the cavitation region. Therefore the cavitation bubbles collapse to the small area in the center of the initial cavitation region.

When one continuous filament is formed the bubbles are completely overlapped (see figure 2). The overlapping leads to an energy exchange between the cavitation bubbles. The exchange becomes stronger, when the difference between the bubble diameters (energies) grows. The evolution of the cavitation region in the case when aberrations were added to the optical scheme, is in good agreement with the Rayleigh model, despite the energy exchange between the cavitation bubbles (see figures 4(a) and (b)). This can be easily explained by the fact that the cavitation bubbles have approximately uniform diameter and energy, therefore the exchange is insignificant. At small energies, when aberrations were added to the optical scheme, the cavitation bubbles are isolated from each other and the energy exchange is negligible (see figure 4(a1)). When a single filament is formed, only the growth of the cavitation region diameter can be described by the Rayleigh model. During the cavitation bubbles’ growth, the pressure and energy gradients are directed from the bubble. Therefore the forces, acting on the bubble boundary, do not sufficiently differ from the forces that arose in the case of the spherical bubble growth. An attempt to approximate the cavitation area evolution, assuming that it has a cylindrical shape, based on the Rayleigh, gives a result that is much worse than in the case of the bubble with a spherical shape [12]. Accordingly, the cavitation area can be considered as a superposition of overlapped spherical bubbles, which interact with each other. The speed of the collapse sufficiently differs from the one predicted by the Rayleigh model because of the small oscillation times of the outer bubbles. They collapse first and transfer the conserved energy to the nearest bubble increasing its lifetime. This leads to the energy flux transmitting the energy from bubble to bubble.

For the lens L1 with aberrations all cavitation bubbles are spherical and the diameter of each consequent bubble is less than the previous one (figure 5(a)). The evolution of the cavitation bubbles is in good agreement with the Rayleigh model for laser energy below $14 \pm 2 \mu J$ and for the bubbles distant from the first ‘hot spot’, because in the first order of approximation such bubbles are spherical and their interactions with each other can be neglected. The bubbles begin to overlap at energies above $14 \pm 2 \mu J$. The overlapping leads to an energy exchange between the bubbles and a significant...
difference to the Rayleigh model occurs (figure 5(a)). At long time delays (more than 2 μs) and when the pump energies are above 80 ± 8 μJ (see figure 5) the bubbles are completely overlapped and indistinguishable from each other. They form one ‘drop-shaped’ cavitation region, whose length is greater than the maximal diameter. The diameters of the first bubbles are approximately equal, therefore they have an equal energy. Due to this fact the energy exchange between the first bubble and its neighbors is insignificant. When the ‘drop’ collapses, we have a significant difference in the energy and pressure gradient on the opposite sides of the structure (see figures 5(d)–(f)), which is similar to the bubble collapse in the case of the lens L2. Thus, the length of the cavitation region decreases much greater than the diameter (figure 5(a)). Such a rapid decrease of the length leads to the energy and momentum flux that is directed to the center of the cavitation area. The smaller bubbles collapse first, therefore the flux directed toward the laser radiation and the jets appears (see figures 5(g)–(j)). After averaging shadow photographs over twenty pictures, we also found that the average cavitation region shape is a modified mirror image of the initial bubble. This is because the energy is transmitted towards the laser, which leads to the formation of the secondary cavitation bubble before the geometric focus. The conservation of the cavitation bubble shape on the microsecond time scale still makes it possible to determine the initial spatial plasma density distribution. One can say that the cavitation ‘remembers’ the initial energy distribution on the filament axis up to the microsecond time scale.

4. Conclusion

In this letter we investigated the evolution of multiple cavitation bubbles excited by the femtosecond filament fired by tightly focused near-IR Cr:Forsterite laser radiation with high linear absorption in water under a supercritical power regime. We established that in our experimental conditions a single filament can be formed. If aberrations are added to the optical scheme multiple filaments will be fired in aberration foci. Overlapping of the filaments can efficiently increase the length of the cavitation area. The filaments, whose lengths logarithmically depend on the laser energy, become the centers of the cavitation bubble formation. At different regimes of the filament formation cavitation bubbles can be isolated from each other, form one cylindrical cavitation region or construct one ‘drop-shaped’ cavitation region. We found that the bubbles’ evolution is completely described by the Rayleigh model when cavitation bubbles are completely isolated from each other. The overlapping causes the energy exchange between the bubbles. The influence of the exchange on every bubble’s formation depends on the energy difference between neighboring bubbles. When the diameters (and energies) of the bubbles are sufficiently different from each other the Rayleigh model still could not be applied to that bubble’s behavior description. During the collapse the outer bubbles collapse first and transfer their energy to the inner bubbles or to the surrounding medium, emitting jets.

Acknowledgment

This research has been supported by the Russian Foundation for Basic Research (Project No. 14-02-00819a) and partly by the M.V. Lomonosov Moscow State University Program of Development.

References

[1] Lauterborn W and Vogel A 2013 Bubble Dynamics and Shock Waves ed C F Delale (Berlin: Springer)
[2] Suslick K S and Flannigan D J 2008 Inside a collapsing bubble: sonoluminescence and the conditions during cavitation Annu. Rev. Phys. Chem. 59 659–83
[3] Rau K R, Guerra A, Vogel A, Venugopalan V, Ili A G and Weg P M 2004 Investigation of laser-induced cell lysis using time-resolved imaging Appl. Phys. Lett. 84 2940–2
[4] Dijkstra R, Le Gac S, Nijhuis E, van den Berg A, Vermes I, Poot A and Ohl C-D 2008 Controlled cavitation-cell interaction: trans-membrane transport and viability studies Phys. Med. Biol. 53 375–90
[5] Hammer D X, Thomas R J, Nooij D G, Rockwell B A and Vogel A 1995 Ultrashort pulse laser induced bubble creation thresholds in oculair media Proc. SPIE 2391 30–40
[6] Peters I R, Tagawa Y, Oudalov N, Sun C, Prosperetti A, Lohse D and van der Meer D 2013 Highly focused supersonic microjets: numerical simulations J. Fluid Mech. 719 587–605
[7] Lim K Y, Quinto-Su P A, Klaseboer E, Khoob B C, Venugopalan V and Ohl C-D 2010 Nonspherical laser-induced cavitation bubbles Phys. Rev. E 81 016308
[8] Schaffer C, Nishimura N, Glezer E, Kim A and Mazur E 2002 Dynamics of femtosecond laser-induced breakdown in water from femtoseconds to microseconds Opt. Express 10 196–203
[9] Plessert M S 1977 Bubble dynamics and cavitation Annu. Rev. Fluid Mech. 9 145–85
[10] Ohl C-D, Kurz T, Geisler R, Lindau O and Lauterborn W 1999 Bubble dynamics, shock waves and sonoluminescence Philos. Trans. R. Soc. A Math. Phys. Eng. Sci. 357 269–94
[11] Quinto-Su P, Lim K and Ohl C-D 2009 Cavitation bubble dynamics in microfluidic gaps of variable height Phys. Rev. E 80 047301
[12] Godwin R P, Chapyak E J, Noack J and Vogel A 1999 Aspherical bubble dynamics and oscillation times Proc. SPIE 3601 225–36
[13] Tinne N, Schumacher S, Nuzzo V, Ripken T and Lubatschowski H 2009 Dynamic and interaction of fs-laser induced cavitation bubble for analyzing cutting effect Proc. SPIE 7373 73730L
[14] Couairon A and Mysyrowicz A 2007 Femtosecond filamentation in transparent media Phys. Rep. 441 47–189
[15] Yu J, Mondelain D, Kasparian J, Salmon E, Geoffroy S, Favre C, Boutou V and Wolf J-P 2003 Sonographic probing of laser filaments in air Appl. Opt. 42 7117–20
[16] Zheng Z-Y, Zhang J, Hao Z-Q, Zhang Z, Chen M, Lu X, Wang Z-H and Wei Z-Y 2005 Paper airplane propelled by laser plasma channels generated by femtosecond laser pulses in air Opt. Express 13 10616–21
[17] Wahlstrøm J K, Jhajj N, Rosenthal E W, Zahedpour S and Milchberg H M 2014 Direct imaging of the acoustic waves generated by femtosecond filaments in air Opt. Lett. 39 1290
[18] Cheng Y-H, Wahlstrøm J K, Jhajj N and Milchberg H M 2013 The effect of long timescale gas dynamics on femtosecond filamentation Opt. Express 21 4740–51
Laser Phys. Lett. 12 (2015) 015405

[19] Jhajj N, Rosenthal E W, Birnbaum R, Wahlstrand J K and Milchberg H M 2014 Demonstration of long-lived high-power optical waveguides in air Phys. Rev. X 4 011027

[20] Bayly J G, Kartha V B and Stevens W H 1963 The absorption spectra of liquid phase H2O, HDO and D2O from 0.7 to 10μm Infrared Phys. 3 221–2

[21] Marcinkevičius A, Mizeikis V, Juodkazis S, Matsuo S and Misawa H 2003 Effect of refractive index-mismatch on laser microfabrication in silica glass Appl. Phys. A 76 257–60

[22] Vogel A, Nahen K, Theisen D, Birngruber R, Thomas R J and Rockwell B A 1998 Influence of optical aberrations on laser-induced plasma formation in water, and their consequences for intraocular photodisruption Proc. SPIE 3246 120–31

[23] Manenkov A A 2014 Fundamental mechanisms of laser-induced damage in optical materials: today’s state of understanding and problems Opt. Eng. 53 010901

[24] Liu W, Kosareva O, Golubtsov I S, Iwasaki A, Becker A, Kandidov V P and Chin S L 2003 Femtosecond laser pulse filamentation versus optical breakdown in H2O Appl. Phys. B 76 215–29

[25] Dergachev A A, Ionin A A, Kandidov V P, Seleznev L V, Sinitsyn D V, Sunchugasheva E S and Shlenov S A 2013 Filamentation of IR and UV femtosecond pulses upon focusing in air Quantum Electron. 43 29–36

[26] Gordienko V M, Makarov I A, Mikheev P M, Savel’ev A B, Shashkov A A and Volkov R V 2004 Self-channeling of femtosecond laser radiation in transparent two-component condensed medium Proc. SPIE 5399 96

[27] Minardi S, Gopal A, Tatarakis M, Couairon A, Tamosauskas G, Piskarskas R, Dubietis A and Di Trapani P 2008 Time-resolved refractive index and absorption mapping of light-plasma filaments in water Opt. Lett. 33 86–8

[28] Vogel A, Linz N, Freidank S and Paltauf G 2008 Femtosecond-laser-induced nanocavitation in water: implications for optical breakdown threshold and cell surgery Phys. Rev. Lett. 100 038102

[29] Bowden C M and Chin S L 2002 Intensity clamping of a femtosecond laser pulse in condensed matter Opt. Commun. 202 189–97