Near threshold nucleation and growth of cavitation bubbles generated with a picosecond laser

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ABSTRACT

The nucleation and growth of cavitation bubbles few micrometers in size in water generated by a 60 ps 515 nm fiber laser is observed and visualized near nucleation threshold. The study is performed by monitoring the plasma size, the cavitation bubble size and the emitted shock waves. The latter two aspects are supported by the Gilmore model using a Noble-Abel-stiffened-gas (NASG) equations of state. For the first time, two types of cavitation events are identified and visualized that exhibit a difference of more than two orders of magnitude in the excitation energy converted to mechanical effects with minimal change in excitation laser pulse energy. The result is localized cavitation and reduced mechanical stress on water-based media with potentially positive implications for laser treatments of biological tissue.

1. Introduction

Cavitation and accompanying phenomena such as shock waves have been extensively studied because of their important role in various technological and medical processes [1], either to mitigate their negative effects (e.g., erosion [2]) or to use them beneficially, e.g., to treat water-based media [3] or to solidify materials [4]. A well-established mechanism for generating controlled cavitation bubbles is laser-induced breakdown (LIB) [5]. It enables precise and repetitive formation of single or multiple cavitation bubbles suitable for a variety of applications, from the study of cavitation phenomena in different media and at interfaces to direct applications such as laser surgery, which can be extended to other methods of cavitation generation (e.g., acoustic [6]).

A particular interest is the generation of micrometer-sized cavitation bubbles for sonoporation [7] and nanosurgery [8] research. The mechanism of LIB-assisted laser nano-surgery of cells and tissues was presented in [6], where the formation of cavitation bubbles was explained in detail by the interplay of multiphoton and cascade ionization of water by the femtosecond excitation laser. When a certain free electron threshold [9] is reached, a luminous plasma [10] is generally formed with a rapid temperature rise [11], followed by shock wave emission and cavitation [12,13]. This converts some of the laser energy into mechanical energy [14], which affects the area outside the laser focal point. To limit the disturbed area, femtosecond lasers with high intensities are advantageous because they allow for the generation of cavitation bubbles in the sub-micrometer range. The reason behind is that pulses from femtosecond lasers have sufficient intensity to cause LIB while the pulse energy is still low due to short pulse duration and can be tuned down to limit the plasma generation around the focal volume. On the contrary, nanosecond laser pulses carry much higher energies at the same threshold intensities and when the LIB process starts, exhibit much brighter and usually elongated plasmas and shockwaves [15]. A recent study [16] investigated the LIB generation of micrometer-sized cavitation bubbles in water and two lower viscosity solutions using a femtosecond laser. For the tested energies between 30 nJ and 80 nJ, a transfer of more than 10 % of the excitation laser energy into the shock wave was shown, and the dependence of the generated cavitation bubble radius and the emitted shock wave on the excitation energy was almost linear in the whole energy range.

To initiate cavitation with minimal effects to the surrounding medium it is necessary to minimize the generated plasma intensity and thus the emitted shock waves. Previous studies show that laser pulses with a duration in the low picosecond range generate shock waves with significantly lower shock pressures than nanosecond or femtosecond pulses by more than a factor of eight [14]. This corresponds to the local minimum in plasma absorption exhibited for picosecond pulses [17], suggesting that weaker shock waves are emitted when the plasma cannot be sufficiently pumped by the excitation laser pulse.

In this work, the properties of cavitation bubbles a few micrometers

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in size are investigated. The bubbles are generated by a MOPA-based fiber laser with 60 ps pulse duration at 515 nm wavelength. The energy of the excitation laser pulses is varied on the microjoule scale up to 3 μJ. We observe a twofold threshold behavior for the microbubble-generating cavitation process. Laser pulse energies just above the first threshold resulted in cavitation effects confined to an area with a radius of less than 5 μm and weak conversion of laser energy into mechanical effects. Pulse energies above the second threshold resulted in detectable plasma, well defined cavitation bubble evolution and emitted shock waves. We explain the phenomena experimentally by monitoring the amplitude of the shock waves and the size of the plasma.

2. Material and methods

2.1. Experimental setup

The experimental setup for observing laser-induced cavitation bubble formation at the micrometer scale consisted of an excitation fiber laser source coupled to a 3D-printed experimental chamber through a microscope objective. The excitation laser was a custom fiber laser source based on a gain switched laser diode master oscillator power amplifier architecture with a maximum average power of 15 W at fundamental wavelength of 1030 nm. It was frequency doubled to 515 nm and generates 60 picosecond pulses. An external modulator was used to produce one pulse per second and to adjust the excitation laser pulse energy. A time interval of one second was chosen to allow for a steady state condition to be reached before each LIB event in water. For the experiments we used distilled water with the specified conductivity of 0.8 μS/cm. The excitation laser (z direction) was focused through a microscope objective immersed in water located on the top of the experimental chamber (Fig. 1a) which has a cover-glass visualization ports (x direction), pressure sensor fiber port (y direction) and a volume of 20x20x20 mm³. The objective was a Nikon CFI Apo NIR 40x W with 40x magnification, a nominal numerical aperture (NA) of 0.8, and a working distance of 3.5 mm. In the experiment the numerical aperture was changed between 0.43 and 0.8, by varying the excitation laser beam size in front of the objective. The 1/e² diameter of ideally focused laser beam would be approximately 0.8 μm at maximum aperture of NA = 0.8.

The example image of the cavitation bubble together with the accompanying shock wave and fiber of the fiber optic probe hydrophone (FOPH) is shown in the Fig. 1b. The fiber was moved away from the breakdown region not to disturb the time evolution of the cavitating bubble when measuring the maximum radius dependence on the excitation laser energy.

2.2. Detection

2.2.1. Visualization

For visual cavitation bubble and shock wave detection a short pulsed illumination system [18] was used. Based on the fast driven laser diodes it enabled illumination pulse duration tuning and was already applied in the monitoring the shock wave propagation near a concave surface [19] and shock wave expanded nanobubbles [20]. For optical visualization of the shock waves using 10x magnification, an illumination pulse duration was set to 3 ns. The camera used was a type of an externally triggered standard industrial monochrome camera (Basler acA1920-40 μm) with 1920x1200 resolution and pixel size of 5.86 μm × 5.86 μm. The camera has a monochromatic CMOS sensor with the size of 11.3 mm × 7.1 mm and can be operated at the framerate up to 41 fps. In our case it was operated in a single frame per trigger. A stable TTL trigger signal to the camera was provided by the laser source driver. After each excitation laser pulse fired in 1 s interval the image was captured on the camera by the single illumination pulse. The delay between the excitation laser pulse and the illumination pulse was adjusted to record the cavitating bubble size at the desired time. For the given excitation laser energy and illumination pulse time delay fifty images were captured at 1 s intervals. The plasma measurements were performed by switching the illumination pulse off and setting the camera exposure to 500 μs centered around the time of laser induced breakdown.

2.2.2. Pressure measurement

Transient shock wave pressure was measured using a custom-build fiber optic probe hydrophone (FOPH) operating at a 1030 nm wavelength, guiding the light in a single-mode fiber with 5 μm core and 125 μm cladding diameters. The FOPH has been tested and validated in measuring of a sub-nanosecond shock wave pressure rise times [21] and in combination with the multi-frame multi-exposure shock wave imaging for velocity and pressure measurements [22].

The FOPH was positioned perpendicular to the incident excitation laser beam (Fig. 1b) at a distance of 45 μm from the breakdown. The light reflected from the FOPH tip was detected by a 5 GHz photodiode (Thorlabs DET08CFC), which displayed a shock wave induced voltage drop from a steady-state signal of 500–1000 mV on an oscilloscope. For the given excitation laser energy thirty traces were recorded at 1 s intervals between laser shots.

3. Theory

The nucleation and growth of cavitation bubbles generated by the laser induced breakdown (LIB) are strongly influenced by the laser pulse duration, intensity, and laser wavelength. Near threshold, the LIB generated plasma is not fully developed, which influences the laser excitation energy conversion to mechanical energy in form of cavitation...
bubble potential energy and the pressure amplitude of the accompanying shock.

3.1. Cavitation bubble nucleation by the laser induced breakdown

Cavitating bubble nucleation happens around the laser generated plasma and transfer of free electrons energy to the water leading to phase explosion. To describe the process of free electron generation in water, it can be treated as an amorphous semiconductor with a band gap of 6.5 eV [23]. Therefore, a number of photons \( k \) is required to generate a free electron by a multiphoton process (Table 1a), where \( k \) is at a laser wavelength of 515 nm. Important aspects of free electron generation in water-based media are explained in [17,24] and are summarized in Table 1, where \( q \) is the free electron density and \( \dot{q} \) the laser pulse intensity. The interaction of the processes during the duration of the excitation laser pulse causes the density of free electrons to increase up to the critical value \( (q_c)_{\text{crit}} \), above which the plasma becomes strongly absorbing and reflecting. Thus, different temporal and spatial profiles of free electron density can be produced, heavily influencing the conversion from laser excitation energy into mechanical energy.

During the excitation laser pulse, the absorption of laser energy ranges from less than 10 % for femtosecond pulses (multiphoton absorption only) to more than 90 % at the end of the nanosecond pulse (strong cascade ionization). The energy balance has been studied previously for large irradiances and different pulse lengths [26]. It was found that for bright plasma production, the deposited mechanical energy also increases in the form of the cavitation bubble size and the emitted shock wave pressure amplitude. Laser absorption in the generated plasma is lowest at 3 ps pulses [17]. This is attributed to the specifics of the processes involved in free electron generation (Table 1), in particular the temporal overlap between excitation laser intensity and free electron density (Table 1b), which is crucial for strong cascade ionization and bright plasma generation. This subsequently leads to a strong temperature rise of several thousand degrees in water at the focal point. At a pulse length of 60 ps, there is still a weak temporal overlap between the population of free electrons and the laser pulse, which limits the cascade ionization.

3.2. Cavitation bubble temporal evolution and pressure

After bubble nucleation and phase explosion the cavitation bubble starts to grow. To describe the bubble evolution on given time (several hundred nanoseconds) and spatial scales (several micrometers bubble radius) an established Gilmore model [27] for viscous compressible liquid was used. It enables an insight in to the initial bubble wall velocity right after the explosion and can be used to fit the measured data of bubble temporal evolution. From the bubble wall velocity right after the explosion we can also approximate the shock wave at the pressure sensor. The model describes bubble radius \( R \) by the second order ordinary differential equation:

\[
RR\left( 1 - \frac{R}{C_i} \right) + \frac{3}{2} \dot{R}^2 R \left( 1 - \frac{R}{C_i} \right) = H \left( 1 - \frac{R}{C_i} \right) + \dot{H} \frac{R}{C_i} \left( 1 - \frac{R}{C_i} \right)
\]

(1)

where \( C_i \) is the speed of sound in liquid at the bubble wall, \( H \) is the enthalpy difference of the liquid at the bubble wall and the infinity. The implementation of the model is given in [28] with the Noble-Abel-stiffened-gas (NASG) equations of state (EOS). In the reference [28], the model is applied on the initially small (micrometer scale) bubble at rest, meaning the gas pressure inside the bubble \( p_0 \) is in a steady state defined by the initial bubble radius \( R_0 \) surface tension with the coefficient \( \sigma \) and liquid pressure \( p_l \):

\[
p_l = p_0 + \frac{2\sigma}{R_0}
\]

(2)

For water the surface tension is \( \sigma = 0.073 \text{ N/m} \) and the liquid pressure is set to static ambient pressure of \( p_l = 100 \text{ kPa} \).

The surface tension starts to influence the initial conditions when working with the bubble nucleuses of the micrometer scale or less in water. The model is adapted to the LIB and phase explosion case by modifying the initial gas pressure \( p_0 \) to value associated to the laser energy deposited into the plasma and exceeding the steady state condition given in Eq.2. In literature [29,30] one can find the calculations of the pressure evolution during laser pulse, but it is for the laser pulse energies (millijoule range) well above threshold and the model is validated for the longer nanosecond pulses, where plasma is strongly absorbing. For picosecond pulses with microjoule energies that were used in our experiments we are close to the bubble nucleation threshold and as such the initial conditions are far from those of highly energetic pulses. Thus, the initial gas pressure was a free parameter in fitting the model to the measurements implemented as sharp rise in its value from steady state.

The calculation of the pressure variation in time at the FOPH distance from the breakdown was based on Kirkwood-Bethe hypothesis described in [13] and used to simulate shock waves produced after LIB [29] with the introduction of invariant named kinetic enthalpy \( y = r(h + u^2/2) \) that propagates outward from the breakdown region and connects local liquid velocity \( u \) and enthalpy difference \( h \) at given distance \( r \). The value of \( y(r) \) at the bubble wall with the radius \( R(t) \) and moving with a speed \( U(t) \) can be written as:

\[
y = R \left( H + \frac{U^2}{2} \right)
\]

(3)

According to Eq. (3) the bubble wall radius \( R \) and speed \( U \) at the given time present an initial conditions to calculate the kinetic enthalpy which can be than used to calculate the velocity profile of the disturbance propagating away from the bubble wall with the set of differential equations [29]:

\[
\dot{u} = -\frac{1}{c - u} \left[ (c + u) \frac{y}{r^2} \frac{2c^2u}{r} \right]
\]

(4)

\[
\dot{r} = u + c
\]

(5)

with the local speed of sound expressed as:

\[
c = c_0 \left( \frac{p + B}{p_0 + B} \right)^{(e-1)/2e}
\]

(6)

---

Table 1

| Process of LIB in water. | Rate q | Onset | Effect |
|------------------------|--------|-------|--------|
| a) Multiphoton ionization | \( \propto k^a \) | Start | Producing free electrons. Seeding process for the LIB. |
| b) Cascade ionization | \( \propto q \cdot I \) | Free electrons needed | Dominant for fs pulses. Inverse Bremsstrahlung absorption (IBA) \( \rightarrow \) free electrons gain kinetic energy |
| c) Recombination | \( \propto 2 \times 10^{-9} \text{s}^{-1} \) | During pulse | Impact ionization \( \rightarrow \) collisions produce new free electrons |
| d) Electron-ion energy transfer | / | Characteristics: \( t < 10 \text{ ps} \) [25] | Plasma process, limits plasma growth |

* Assisted by Tunneling ionization for \( I > 10^{13} \text{W/cm}^2 \) significant for femtosecond pulses (<100 fs).
and the transient pressure \( p \) at the \( r = r(t) \) written as:

\[
p(t) = (p_{\infty} + B) \left( \frac{y}{r^2} \right)^{(n-1)} \frac{1}{n(n+1)} \left[ \frac{1}{n} \right]^n (p_{\infty} + B) + 1 \]  

(7)

The equation of state coefficients are \( B = 6.22 \times 10^8 \) Pa and \( n = 1.19 \) \([31]\), density of water is \( \rho_{\infty} = 997 \) kg/m\(^3\) and pressure at infinity is equal to static ambient pressure \( p_{\infty} = 100 \) kPa. The Eq.7 can thus be used to calculate the acoustic wave pressure variation in time at a given distance from the LIB.

4. Results and discussion

The laser induced cavitation bubble nucleation and evolution on a scale of a few micrometers was observed by varying energy of the excitation laser pulse energy \( (E_L) \) focused in to the experimental chamber at three different numerical apertures. The breakdown region was recorded on the camera when the illumination pulse was on for bubble and shock wave visualization and when the illumination pulse was off for plasma inspection.

The image sequence of the cavitation process recorded in the case of the excitation laser pulse energy set to \( E_L = 0.5 \) \( \mu \)J is shown in Fig. 2a and in the case of \( E_L = 2.8 \) \( \mu \)J in Fig. 2b. In both cases, the laser beam was focused at \( \text{NA} = 0.58 \). The Illum. OFF image shows the breakdown region with the illumination pulse turned off and an electronic camera shutter open for 500 \( \mu \)s centered around the time of laser induced breakdown. Additional images illuminated with the 3 ps pulse show the bubble at different time points. By the visual inspection no shock waves or plasma could be detected at low excitation laser energies. In addition, the maximum radius shown in Fig. 2a is more than two orders of magnitude smaller than that shown in Fig. 2b, which was produced with an excitation laser pulse energy only three times higher, indicating a large difference in energy conversion from laser pulse to the potential energy of the bubble.

To record the evolution of bubble radius in time the time delay between the laser excitation pulse and the illumination pulse was varied at a given excitation laser pulse energy. For each combination of energy and time delay fifty shots were fired in 1 s intervals into the chamber. Fig. 3 shows the occurrence plot of radii (discretized in 1 \( \mu \)m bins) plotted versus time (time delay) at the excitation laser energy \( E_L = 0.54 \) \( \mu \)J where mixed nucleation and bubble growth were detected, supporting the difference in the cavitation effect intensity for small excitation energy variation.

![Fig. 2. The image sequence of the cavitation process recorded in the case of a) the excitation laser pulse energy set to \( E_L = 0.5 \) \( \mu \)J and b) set to \( E_L = 2.8 \) \( \mu \)J. The laser beam is focused at \( \text{NA} = 0.58 \). The Illum. OFF image shows the breakdown region with the illumination pulse turned off for plasma visualization. The scale bar is equal to 50 \( \mu \)m.](image)

![Fig. 3. Mixed process bubble nucleation and growth observed at \( E_L = 0.54 \) \( \mu \)J and \( \text{NA} = 0.58 \)](image)
Turning the illumination pulses off at the set excitation pulse energy the plasma image was captured on camera in $\vec{y} - \vec{z}$ plane. The measured intensity plasma profile ($\vec{y}$ direction) perpendicular on the excitation laser path is shown in Fig. 4 for five excitation laser pulse energies at $NA = 0.58$. The corresponding images of plasma are shown in the Fig. 4 insets (a-e).

The results of the FOPH pressure measurement at the distance of 45 $\mu$m from breakdown are shown in Fig. 5. The results are shown for three different numerical apertures ($NA = 0.43$, $NA = 0.58$ and $NA = 0.80$), with the first row (Fig. 5a–c) showing the representative measured pressure traces for excitation pulse energies $E_L = 0.56 \mu J$, $E_L = 0.66 \mu J$ and $E_L = 2.6 \mu J$ at $NA = 0.58$. Fig. 5e shows the probability of the detected shock waves. It can be seen from the Fig. 5a that measured pressure is still above the noise level of the system (~2 MPa) but shock waves below the excitation laser energies of $E_L = 0.56 \mu J$ at $NA = 0.58$ could not be detected while the cavitation bubble was still captured by the camera. Similar sharp cutoff was exhibited at $NA = 0.43$ and $NA = 0.80$.

To better illustrate the difference between the cavitation effects near the LIB threshold the combined plots of data collected with the camera and FOPH are shown in Fig. 6. The values of the maximum bubble radius (Fig. 6a), plasma size (Fig. 6b), shock wave pressure (Fig. 6c) and shock wave probability (Fig. 6d) are plotted as a function of the excitation laser pulse energy at $NA = 0.58$. The maximum bubble radius for given excitation energy was determined by numerically fitting the model (Eq. (1)) to the measured data as shown in Fig. 6e. The resulting graph (Fig. 6a) shows two thresholds. The first threshold applies when only the bubbles are detected and the second, indicated by the blue dashed line, applies when the bubble is accompanied by the shock wave emission.

The second threshold is also confirmed by the shock wave emission probability plot (Fig. 6d). In the case of the LIB process after the second threshold the plasma can be detected at excitation pulse energies higher than $E_L = 0.9 \mu J$ in contrast to $E_L = 0.54 \mu J$ when the shock waves can be first detected. In addition to the sensitivity of our system, this can be attributed to the low temperature of the plasma, which was previously reported for bubble formation using nJ femtosecond lasers [32]. According to this report, the bubbles can be formed at plasma temperatures on the order of a few hundred degrees Celsius. At these temperatures, the peak of plasma emission is far outside of the detection range of the detectors used ($\geq 1 \mu m$).

Depending on the laser energy conversion to the mechanical energy (bubble potential energy, shock wave amplitude and plasma intensity) two event types can be identified. Low energy conversion (LEC) event where only the bubble is detected and high energy conversion (HEC) event where the bubble is detected together with the shock wave and plasma. The bubble potential energy can be calculated from the cavitating bubble maximum radius [26]:

$$E_{\text{pot}} = \left(\frac{4}{3}\pi\right)(p_v - p_i)R_{\text{max}}^3$$

where $p_v$ is the vapour pressure inside the bubble ($p_v = 2330$ Pa at 20 $^\circ$C). Plotting the bubble volume, which is directly proportional to the bubble potential energy, gives a clear representation of these two types of events (Fig. 7). In the example of $NA = 0.58$ the LEC and HEC events have similar probability around excitation energy of 0.54 $\mu J$, but exhibit a large difference in transferred energy from the laser pulse to the cavitation mechanical effects with the HEC producing visible plasma, seven times larger bubble than LEC (resulting in a more than 300–times greater bubble energy), and detectable shock waves.

Fig. 7 shows the value of the bubble volumes on the logarithmic scale to clearly distinguish the two events. When excitation laser energy exceeds the second threshold (between LEC and HEC events) the conversion efficiency rapidly increases. While the LEC events bubble volume and in consequence the bubble potential energy exhibit almost exponential dependence on the excitation laser energy, the HEC events volume exhibits linear trend from which slope a slope efficiency ($\Delta E_{\text{pot}}/\Delta E_L$) can be calculated. The slope efficiency for HEC events is shown in Table 2 second column for given numerical aperture with the

![Fig. 4. Measured plasma perpendicular on the excitation laser path (\vec{y} direction) shown for five excitation laser pulse energies at NA = 0.58. The insets from a) to e) show the measured plasma intensity in the \vec{y} – \vec{z} plane of the experiment. The horizontal white dashes on the insets marks the line where the profiles were taken. The size of each inset is 21 x 21 μm.](image-url)
The tighter focusing exhibits higher slope efficiency which is attributed to stronger plasma formation and absorption of laser light during the laser pulse. For comparison the third column of Table 2 shows the values of measured maximal conversion efficiency for the LEC events.

With the measuring equipment at the disposal no shock waves were detected for the LEC events. To get the insight on the acoustic wave emission accompanying the HEC events, a Gilmore model (Eq. (1)) was fitted to the cavitation bubble radius evolution in time by taking the initial pressure jump as a free parameter and fixing the starting bubble nucleus to the measured plasma size. The model returned the bubble wall velocity which was used in accordance with the equations Eq. (3) to Eq. (7) to calculate the HEC events pressure trace measured at the distance of FOPH (45 μm). The risetime of the 1 GHz oscilloscope was accounted for in the initial slope. The three illustrative examples are shown in Fig. 8. The result of Gilmore model fitted to the measurements of cavitation bubble radius evolution is shown in subfigures (a, b and c) while the corresponding fits to shock wave traces are shown in subfigures (d, e and f). Each row in the Fig. 8 corresponds to the excitation laser energy $E_L = 0.66 \mu J$ (Fig. 8a and d), $E_L = 1.81 \mu J$ (Fig. 8b and e) and $E_L = 2.81 \mu J$ (Fig. 8c and f) from top to bottom at $NA = 0.58$.

In comparison to the measured bubble radii in HEC events, which are well defined at given energy, the radii of the LEC event are better visualized with the occurrence plot as the small instabilities in the process (mainly the excitation laser pulse energy on the order of 2 %)

### Table 2

| NA  | HEC event. eff. (%) | LEC event. eff. (%) |
|-----|---------------------|---------------------|
| 0.43| 10.8                | 8.5×10^{-3}         |
| 0.58| 12.7                | 3.7×10^{-3}         |
| 0.80| 13.5                | 0.3×10^{-5}         |

absolute error of 0.5 %. The tighter focusing exhibits higher slope efficiency which is attributed to stronger plasma formation and absorption of laser light during the laser pulse.
Celsius, at which water in breakdown region is vaporized and temperature is expected to be on the order of few thousand degrees when the vapor expands and the pressure drops. In the dred degrees Celsius, we see only the phase change of the -

Fig. 9. LEC events: Gilmore model fitted to the measurements of cavitation bubble radius evolution (a, b, c) with the shock wave traces acquired from the fitted model (D, E, F). Each row corresponds to the excitation laser energy $E_L = 0.49 ~\mu J$, $E_L = 0.51 ~\mu J$, and $E_L = 0.54 ~\mu J$ at $NA = 0.58$.

causes different evolutions of the produced bubbles (Fig. 9a–c). Further the LEC events show the decrease in the number of the detected bubbles when we observe them at times when they are expected to start to collapse.

The difference in the behavior can be attributed to the temperature of plasma before bubble nucleation. In the LEC case, the temperature and pressure are low enough (expected to be on the order of few hundred degrees Celsius) that we see only the phase change of the water from the vapor after the explosion and back to the liquid state when the vapor expands and the pressure drops. In the HEC case the temperature is expected to be on the order of few thousand degrees Celsius, at which water in breakdown region is vaporized and partly dissociated into gaseous products ($H_2$ and $O_2$). The mixture can thus overgo the complete growth and collapse of the cavitation bubble.

The model verified on the HEC events is again run on the LEC events with the exception that no measured pressure traces are available for the model to be compared to. Three illustrative examples are shown in Fig. 9. The result of Gilmore model fitted to the measurements of cavitation bubble radius evolution is shown in subfigures (a, b, c) with the shock wave traces acquired from the fitted model shown in subfigures (d, e, f). Each row in the Fig. 9 corresponds to the excitation laser energy $E_L = 0.49 ~\mu J$, $E_L = 0.51 ~\mu J$, and $E_L = 0.54 ~\mu J$ from top to bottom at $NA = 0.58$.

The model fit of the LEC events was set to depict the upper limit on the produced acoustic wave during the bubble formation. For this we again assumed instant pressure increase due to laser pulse excitation. The fitted bubble radius evolution in shown with the red dashed line on Fig. 9(a–c). In can be seen that the instant pressure increase is indeed the upper limit of the nucleation and growth process and some of the LEC events shows slower bubble growth which would reflect in smaller acoustic pressures. In the case of the upper limit the model returns the pressures below 60 kPa and perturbation duration on the scale of several tens of nanoseconds Fig. 9(d–f). The pressure transient duration is in the LEC case more than 10–times longer (approx. 50 ns) than in the HEC case with the three orders of magnitude smaller amplitude (less than 60 kPa). The modeling results confirm the assumption that the LEC events produces the acoustic waves with low amplitude on a kilopascal scale.

The observed cavitation phenomena and two populations of LEC and HEC events can be attributed to the excitation laser with the pulse duration comparable to the characteristic time of the electron-ion energy transfer (Table 1d), while the pulse intensity can control the rate of the free electron generation and consequently the overlap between the free electron population and the laser pulse intensity for cascade ionization (Table 1b). In the case of LEC the cascade ionization is weak, leading to low energy conversion to the mechanical effects. Increasing the energy enables the increase of the free electron density during the laser pulse to such levels that cascade ionization becomes dominant leading to avalanche ionization and high electron density with bright plasma.

5. Conclusions

This paper shows the results of a study on the generation of cavitation bubbles with a diameter of a few micrometers by an excitation fiber laser with a pulse duration of 60 ps at a wavelength of 515 nm. A twofold threshold process is observed by monitoring the plasma size, cavitation bubble size, and generated shock waves. The identified low energy conversion (LEC) and high energy conversion (HEC) events show a large difference in the energy transferred to the cavitation bubble potential energy (greater than 300-fold) with minimal change in excitation energy. The result is reduced mechanical stress on the water-based medium, which may be advantageous in laser treatments of biological tissue where highly localized perturbations are desired. Such a degree of localization has previously only been achieved with femtosecond pulses and is now being demonstrated for the first time with picosecond pulses. This is made possible by a combination of wavelength selection, pulse duration, and strong focusing, as well as by energetically stable laser pulses that exhibit a smooth intensity distribution in the time domain. Although events similar to the LEC events presented in this work have been produced previously with femtosecond laser pulses, this is the first time a sharp transition between LEC and HEC has been visualized and confirmed by plasma and shock wave measurements, as well as explained and validated by numerical model results.

CRediT authorship contribution statement

Vid Agrež: Conceptualization, Methodology, Visualization, Data curation, Investigation, Formal analysis, Writing – original draft, Writing – review & editing. Jaka Mur: Conceptualization, Methodology, Validation, Visualization, Data curation, Investigation, Writing – original draft, Writing – review & editing. Jaka Petelin: Conceptualization, Methodology, Software, Validation, Visualization, Resources, Writing – review & editing. Rok Petkovsek: Conceptualization, Methodology, Validation, Investigation, Writing – review & editing, Funding acquisition, Supervision.

Declaration of Competing Interest

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.
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