Pre-print of a published manuscript

Comparison of the overall temporal behavior of the bubbles produced by short- and long-pulse nanosecond laser ablations in water using a laser-beam-transmission probe

Ayumu Matsumoto, Ayaka Tamura, Atsushi Kawasaki, Takaya Honda, Peter Gregorčič, Naoya Nishi, Ken-ichi Amano, Kazuhiro Fukami, Tetsuo Sakka

DOI: <https://doi.org/10.1007/s00339-016-9755-x>

Published in: Applied Physics A, March 2016, 122:234

Received: 2 October 2015

Accepted: 12 February 2016

This is a pre-print of an article published in Applied Physics A. The final authenticated version is available online at: <https://doi.org/10.1007/s00339-016-9755-x>.

Please cite this article as: A. Matsumoto, A. Tamura, A. Kawasaki, T. Honda, P. Gregorcic, N. Nishi, K. Amano, K. Fukami, T. Sakka, Comparison of the overall temporal behavior of the bubbles produced by short- and long-pulse nanosecond laser ablations in water using a laser-beam-transmission probe, Appl. Phys. A, 122 (2016), DOI: <https://doi.org/10.1007/s00339-016-9755-x>

Title

Comparison of the overall temporal behavior of the bubbles produced by short- and long-pulse nanosecond laser ablations in water using a laser-beam-transmission probe

Authors

Ayumu Matsumoto^{1,a}, Ayaka Tamura¹, Atsushi Kawasaki¹, Takaya Honda¹, Peter Gregorčič², Naoya Nishi¹, Ken-ichi Amano¹, Kazuhiro Fukami³, Tetsuo Sakka^{1,b}

¹ Department of Energy and Hydrocarbon Chemistry, Graduate School of Engineering, Kyoto University, Kyoto 615-8510, Japan

² Faculty of Mechanical Engineering, University of Ljubljana, Aškerčeva 6, 1000 Ljubljana, Slovenia

³ Department of Materials Science and Engineering, Graduate School of Engineering, Kyoto University, Kyoto 606-8501, Japan

^a fax: +81 75 383 2490, e-mail: matsumoto.ayumu.78w@st.kyoto-u.ac.jp

^b fax: +81 75 383 2490, e-mail: sakka.tetsuo.2a@kyoto-u.ac.jp

PACS numbers

47.55.dd (Bubble dynamics)

47.55.dp (Cavitation and boiling)

52.38.Mf (Laser ablation)

Abstract

We investigate the overall temporal behavior of the bubbles produced by 20-ns- and 100-ns-pulse-duration laser ablations in water using a laser-beam-transmission probe (LBTP). This technique gives the transmission signal attributed to the whole bubble dynamics including the secondary oscillations with a single laser shot. Comparing the signals obtained for both pulse durations, the periods of the first oscillation of the bubble are almost the same. Nevertheless, the periods of the subsequent oscillations are significantly different depending on the pulse duration. Such results are obtained by virtue of the LBTP technique.

1 Introduction

Laser-induced breakdown and laser ablation in water can be applied to underwater elemental analysis [1, 2] and synthesis of nanoparticles [3, 4]. However, those are very complex phenomena which are characterized by various interactions [1–4]. It is well known that a cavitation bubble is generated along with a plasma, as well as a shock wave, by laser-induced breakdown in water and laser ablation of submerged solids. The laser-induced bubble repeats expansion and contraction in the scale of a few hundred microseconds, which can be well-described by the Rayleigh-Plesset equation [5–9], while the plasma disappears in a few microseconds. The bubble dynamics is quite interesting for fundamental research, while it plays an important role for the underwater elemental analysis [10–17] and the synthesis of nanoparticles [18–25].

The effects of the laser parameters on the laser-induced bubble in bulk water (Vogel et al. [26, 27]) and on the bubble produced by the laser ablation in water (Cristoforetti et al. [10]) have been studied. The behavior of the laser-induced bubble has been mainly studied in the pulse duration ranging from femtoseconds to short-nanoseconds so far [10, 26, 27]. On the other hand, it has been clarified that the relationship between the plasma and the bubble is very important to obtain intense and narrow optical emission spectral lines for the underwater elemental analysis, and the relationship can be controlled by changing the pulse duration in the range from ten to hundred nanoseconds [16, 17]. It has also been clarified that nanoparticles are ejected from the bubble in the liquid at the timing of the bubble collapse during the multiple oscillations [4, 24]. However, the whole bubble dynamics including the secondary oscillations in the case of long nanosecond pulse has not been studied, although some bubble images at the first oscillation have been observed [28].

The bubble dynamics has been investigated mainly by shadowgraph imaging [9–12, 14–21, 24, 25, 28]. Other approaches have also been used, such as Schlieren imaging [29], X-ray radiography [30], high-speed laser stroboscopic videography [31], pressure transducer or piezoelectric hydrophone [9, 32], beam-deflection probe [33–35], and optical transmission techniques [36–38]. In the case of shadowgraph imaging, multiple events are required to capture the whole bubble dynamics. Therefore, the shot-to-shot fluctuation makes it difficult to evaluate the temporal behavior of the bubble accurately. In the case of laser-beam-transmission probe (LBTP), the information of the whole bubble dynamics including the secondary oscillations can be obtained with a single laser shot [36], which makes it easy to evaluate the temporal behavior of the bubble accurately.

In the present work, we examine the bubbles produced by the laser ablation of an Al target in water with the laser-pulse durations of 20 ns and 100 ns using the LBTP

technique. We aim at the investigations of the overall temporal behavior of the bubble produced by the long nanosecond pulse irradiation and the applicability of the LBTP technique for monitoring the laser ablation in liquid.

2 Experimental

Figure 1 shows the experimental setup for the LBTP measurement. A home-built Q-switch Nd:YAG laser (pulsed laser) with the wavelength of 1064 nm, the pulse energy of 6.0 mJ (fixed), the pulse duration of 20 ns (short pulse) or 100 ns (long pulse), and the repetition rate of 0.1 Hz was used for the ablation. The pulsed laser was focused onto a flat edge of an Al target in ultrapure water in the direction normal to the surface by a plano-convex lens with the focal length of 70 mm. The distance between the water surface and the target surface was 10 mm. The beam waist of the irradiation laser was $\sim 50 \mu\text{m}$. The ablation spot was adjusted to be the center of the flat edge. The thickness of the target was 1.0 mm. A He-Ne laser (Uniphase, 1103P-1367) (continuous-wave (CW) laser) with the wavelength of 632.8 nm was used as a probe light. The probe light was expanded 10 times using a beam expander (Sill Optics, S6ASS0107/121) and transmitted through the interaction area in the direction parallel to the flat edge from a side of the target. The center of the probe light was intersected with the ablation spot. The whole probe light transmitted through the interaction area was introduced to a photodiode (Thorlabs, Si amplified detector, PDA10A-EC) by another plano-convex lens. The bandwidth of the detector is from DC to 180 MHz. The distance between the ablation spot and the detector was 2.0 m. The signal of the detector was recorded by an oscilloscope (Iwatsu, ViewGo II, DS-5654). The bandwidth, the rising time, and the sampling rate of the oscilloscope were 500 MHz, 750 ps, and 1 GS/s, respectively. A trigger signal was taken from another photodiode which detects the reflected light from a beam splitter placed on the optical path of the pulsed laser. The trigger signal was also recorded by the oscilloscope to monitor the pulse duration.

Figure 1b shows the cross sectional view of the interaction area. The center of the probe light was intersected with the ablation spot. The spatial beam radius of the expanded probe light was $\sim 3.0 \text{ mm}$, which is larger than the maximum radius of the bubble ($\sim 1.0 \text{ mm}$ [28]). The transmission signal detected by the photodiode reflects the total amount of the probe light passed through the region near the target surface. Since the bubble scatters and refracts the probe light, the transmission light decreases with the bubble expansion. Therefore, we can obtain the transmission signal containing the information of the expansion and the contraction of the bubble. Note that the detection system of the transmission light is located far away from the interaction area. Thus, the

light scattered or refracted by the bubble is not detected as the transmission signal.

3 Results and discussion

Figures 2a and 2b show typical transmission signals obtained for 20-ns and 100-ns pulse irradiations, respectively. The signals were normalized by the signal voltage acquired before the pulse irradiation. The normalized signals show the typical behavior of the laser-induced bubble. Mark A in Fig. 2a corresponds to the timing of the pulse irradiation. The positive peak is attributed to the plasma emission. Mark B, i.e., the negative peak, appears due to the first oscillation (expansion and contraction) of the bubble. The LBTP signal does not reach zero value, since the spatial beam radius of the probe light (~ 3.0 mm) is larger than the maximum radius of the bubble (~ 1.0 mm [28]). Mark C shows the timing of the collapse and the re-expansion of the bubble. The time interval between marks A and C is the oscillation period of the first oscillation. Mark D shows the region with several negative peaks that correspond to multiple oscillations of the bubble. The signal did not return to the original position, which is 1.0 in the figure, after the multiple oscillations are over. This is due to residual matters on the target surface, which diffuse slowly in water and attenuate the probe beam. Thus, we consider that such a direct current (DC) part of the LBTP signal reflects the particle concentration around the ablation spot after the bubble disappeared. The positive peak attributed to the plasma emission does not disturb the rest of the LBTP signal. In practice, the plasma emission disappears in a few microseconds [39]. Since the detection system is located far from the point of the plasma generation, the positive peak lasts only a few hundred nanoseconds. Such a timescale is extremely shorter than that of the bubble oscillations. Note that we tried to perform similar measurements with a band-pass interference filter which transmits only the light with the wavelength of the probe light. In this case, the detection of the plasma emission was completely avoided. However, the filter also decreases the intensity of the transmission signal, which made it difficult to observe the multiple oscillations of the bubble. Therefore, the filter was not used in the present experiments.

We cannot discuss the bubble radius straightforwardly from the LBTP signal. This is because the spatial intensity of the probe light is not uniform and the shape of the bubble at the secondary oscillations is different from an ideal hemisphere. However, the LBTP signal is strongly correlated to the bubble radius. The signal intensity can be theoretically converted to the bubble radius, if the probe laser is well defined [38]. The absolute radius of the bubble, however, is not essential to study the temporal behavior of the bubble. Although the bubble radius is difficult to be measured accurately, the LBTP

signal reflects the oscillation period.

The overall signals in Figs. 2a and 2b look very similar at the first glance, while the signals attributed to later oscillations seem to be different. Here, we compare the signals obtained for 20-ns- and 100-ns-pulse irradiations in details. It should be noted that we did not change the pulse energy. Figure 3 shows the signals attributed to the first oscillation. Small negative peaks can be observed at the falling edges of signals attributed to the first and second oscillations (see two arrows in Fig. 3). Such peaks are attributed to shock waves, which are induced at the timing of the plasma generation or the bubble re-expansion and propagate in the surrounding water with supersonic speed. Surprisingly, the signal attributed to the first oscillation did not depend on the pulse duration, although the relationship between the plasma and the bubble during the pulse irradiation is significantly different [17].

Basically, the bubble dynamics is determined by the initial conditions of the bubble. However, it is difficult to determine the initial conditions due to some reasons, e.g., the shape of the nascent bubble is rough and the transient plasma exists in the bubble. Comparing the picosecond and nanosecond pulses, the bubble dynamics varies depending on the presence or absence of the plasma excitation via inverse bremsstrahlung, i.e., a longer pulse duration results in a longer period of the bubble oscillation [10]. Such a trend is due to a high-temperature plasma in the bubble, which gives a momentum for the bubble expansion. In the case of picosecond pulse, all the energy is deposited to the target surface before the plasma generation. In the case of nanosecond pulse, the plasma is generated during the pulse irradiation and is excited in the bubble by absorbing the later part of the laser pulse, which increases the temperature of the plasma. Comparing the short and long nanosecond pulses, the plasma is rapidly quenched in the case of short pulse, while the high-temperature plasma remains for a longer time in the case of long pulse [17]. Therefore, the oscillation period of the bubble in the case of long-pulse is expected to be longer than that of short pulse. However, the period of the first oscillation did not change with the pulse duration in the present experiments. We consider that the first oscillation is dominated by the pulse energy rather than the pulse duration, regardless of whether the energy is consumed for heating the target surface or the plasma as follows. In the case of short pulse, a larger number of species are explosively ablated from the target surface and the species are instantaneously excited when the bubble is still small. This gives a higher momentum for the bubble expansion, although the plasma is rapidly quenched. On the other hand, in the case of long pulse, a smaller number of the species are ablated and the species are slowly excited in the expanding bubble. Although the momentum given for the bubble expansion must be smaller immediately after the plasma

generation, the rapid quenching of the plasma is suppressed. Thus, the high-temperature plasma persists for a longer time, which expands the bubble more. Therefore, the periods of the first oscillation can be almost the same. The bubble expansion may be simply controlled by the pulse energy in the present range of the pulse duration.

Here, we refer to the results of shadowgraph measurements that the bubble sizes at 600 ns after the pulse irradiation do not depend on the pulse duration (30, 50, 100 ns) [17]. The observation of similar bubble sizes at 600 ns supports the present result that the transmission signals attributed to the first oscillation obtained for short and long pulses are almost the same. It is of great interest to observe a nascent bubble during the pulse irradiation using the present technique. But unfortunately, the initial bubble cannot be measured in the present system. Actually, the decrease of the transmission signal attributed to the bubble expansion was observed at several microseconds after the pulse irradiation. Immediately after the pulse irradiation, the positive peak attributed to the plasma emission lasts a few hundred nanoseconds, and successively, the negative peak attributed to the shock wave lasts a few microseconds. Although the band pass filter enables us to block the plasma emission, the shock wave signal cannot be avoided. As for the bubble at the early stage, therefore, we have referred to the previous results of shadowgraph measurements [17].

Figure 4 shows the signals attributed to the second and later oscillations of the bubble. Unexpectedly, the signal attributed to the secondary oscillations significantly depends on the pulse duration, especially from the third oscillation, although the signals attributed to the first oscillation are almost the same. Figure 5 shows the oscillation period as a function of the bubble oscillation. The oscillation periods of the later oscillations in the case of long pulse are shorter than that of short pulse. Additionally, we show the averaged signals of five measurements in Fig. 6 to confirm that this observation is systematic. The bubble oscillations at the later stage can be clearly seen in the case of short pulse. On the other hand, the bubble oscillations at the later stage in the case of the long pulse are averaged out and not seen in this figure, suggesting that the shot-to-shot fluctuation is significant..

Here, we discuss the factors which may affect the later oscillations of the bubble. It has been considered that nucleation of particles occurs in the bubble [4, 20, 22, 23]. The particles diffuse with the bubble expansion and aggregate with the bubble contraction. Those are released from inside to outside of the bubble during the bubble collapse and dispersed in water [4, 24]. In the present experiments, we can monitor the particle concentration around the ablation spot after the bubble disappeared from the DC part of the LBTP signal, i.e., the signal after the multiple oscillations of bubble are over. In the

case of short pulse, the intensity of the DC part is relatively low (e.g., see Fig. 6), which means a higher concentration of the particles. This indicates that a greater amount of the particles exists in the bubble during the multiple oscillations. The dynamics of the first oscillation seems to be determined by the pulse energy, regardless of whether the energy is consumed for heating the target surface or the plasma as described above. On the other hand, the dynamics of the secondary oscillations should be determined by the behavior of the collapsing bubble at the previous oscillation. The particle concentration in the bubble will strongly affect the behavior of the collapsing bubble. Therefore, the secondary oscillations can be different depending on the pulse duration. Changes in the surface geometry of the target after the pulse irradiation can also affect the behavior of the bubble. Short-pulse irradiation produces a deep crater [40]. The bottom of the crater can be the re-expansion point of the bubble, which can give reproducible oscillations of the bubble. In the case of long pulse, a rough surface without a deep hole is formed [40]. Therefore, the re-expansion of the bubble may be unstable, which causes the shot-to-shot fluctuation. Such a difference may be caused by the relatively high ablation efficiency of the Al target. The bubble size and the oscillation period become smaller and shorter with the bubble oscillation, respectively. This can increase the effect of the above factors on the behavior of the bubble.

It should be noted that the shape of the bubble at the secondary oscillations is irregular [31] compared to that at the first oscillation. The bubble at the later oscillations may be mainly composed of the residual matters rather than the gaseous phase. In addition, the particle cloud is suspended over the oscillating the bubble. Thus, the later oscillations of the LBTP signal may be due to the particles. In the case of short pulse, however, we consider that the later oscillations are not due to the particles. This is because the multiple oscillations are clearly observed even in the averaged signal in Fig. 6, which means that the shot-to-shot fluctuation is not significant. Also, when comparing the data obtained for shot-by-shot, the secondary oscillations are correlated with the first oscillation, i.e., when the period of the first oscillation is long, the periods of the secondary oscillations are also long. In the case of long pulse, the LBTP signal may be dominated by the particles rather than the bubble, since the multiple oscillations are averaged out as shown in Fig. 6. The particles can disturb the periodic behavior of the transmission signal.

4 Conclusions

In the present work, we investigated the effects of the difference in a hundred nanosecond-scale pulse duration on the overall temporal behavior of the bubble, which has not been studied enough so far. Especially, the difference in the secondary oscillations of the

bubble has been successfully observed, which is achieved by virtue of the LBTP technique. The following results are very interesting that the signals attributed to the secondary oscillations are considerably different depending on the pulse duration although those attributed to the first oscillation are almost the same, which cannot be explained only by the conventional idea, i.e., the plasma excitation via inverse bremsstrahlung. The present results also indicate that the non-repeatable conditions can be overcome by the LBTP technique. Moreover, the LBTP technique is a very appropriate technique for monitoring the laser ablation in liquid, since the signal attributed to the particle concentration has been successfully observed, which is worth to be the subject of further investigations.

Acknowledgements

This work was supported by JSPS KAKENHI Grant Number 14J02461.

References

1. A. De Giacomo, M. Dell'Aglio, O. De Pascale, M. Capitelli, *Spectrochim. Acta, Part B* **62**, 721 (2007)
2. V. Lazic, S. Jovićević, *Spectrochim. Acta, Part B* **101**, 288 (2014)
3. G.W. Yang, *Prog. Mater. Sci.* **52**, 648 (2007).
4. M. Dell'Aglio, R. Gaudiuso, O. De Pascale, A. De Giacomo, *Appl. Surf. Sci.* **348**, 4 (2015)
5. Lord Rayleigh, *Philos. Mag.* **34**, 94 (1917)
6. M.S. Plesset, *J. Appl. Mech.* **16**, 277 (1949)
7. A. Casavola, A. De Giacomo, M. Dell'Aglio, F. Taccogna, G. Colonna, O. De Pascale, S. Longo, *Spectrochim. Acta, Part B* **60**, 975 (2005)
8. W. Soliman, T. Nakano, N. Takada, K. Sasaki, *Jpn. J. Appl. Phys.* **49**, 116202 (2010)
9. A. Tamura, T. Sakka, K. Fukami, Y.H. Ogata, *Appl. Phys. A* **112**, 209 (2013)
10. G. Cristoforetti, M. Tiberi, A. Simonelli P. Marsili, F. Giammanco, *Appl. Opt.* **51**, B30 (2012)
11. T. Sakka, A. Tamura, T. Nakajima, K. Fukami, Y.H. Ogata, *J. Chem. Phys.* **136**, 174201 (2012)
12. V. Lazic, J.J. Laserna, S. Jovićević, *Spectrochim. Acta, Part B* **82**, 42 (2013)
13. V. Lazic, J.J. Laserna, S. Jovićević, *Spectrochim. Acta, Part B* **82**, 50 (2013)
14. B. Thornton, T. Sakka, T. Takahashi, A. Tamura, T. Masamura, A. Matsumoto, *Appl. Phys. Express* **6**, 082401 (2013)
15. B. Thornton, T. Sakka, T. Masamura, A. Tamura, T. Takahashi, A. Matsumoto, *Spectrochim. Acta, Part B* **97**, 7 (2014)
16. T. Sakka, A. Tamura, A. Matsumoto, K. Fukami, N. Nishi, B. Thornton, *Spectrochim. Acta, Part B* **97**, 94 (2014)
17. A. Tamura, A. Matsumoto, K. Fukami, N. Nishi, T. Sakka, *J. Appl. Phys.* **117**, 173304 (2015)
18. T. Tsuji, Y. Tsuboi, N. Kitamura, M. Tsuji, *Appl. Surf. Sci.* **229**, 365 (2004)
19. T. Tsuji, Y. Okazaki, Y. Tsuboi, M. Tsuji, *Jpn. J. Appl. Phys.* **46**, 1533 (2007)
20. W. Soliman, N. Takada, K. Sasaki, *Appl. Phys. Express* **3**, 035201 (2010)
21. A. De Giacomo, A. De Bonis, M. Dell'Aglio, O. De Pascale, R. Gaudiuso, S. Orlando, A. Santagata, G. S. Senesi, F. Taccogna, R. Teghil, *J. Phys. Chem. C* **115**, 5123 (2011).
22. S. Ibrahimkuty, P. Wagener, A. Menzel, A. Plech, S. Barcikowski, *Appl. Phys. Lett.*

- 101**, 103104 (2012)
23. P. Wagener, S. Ibrahimkutty, A. Menzel, A. Plech, S. Barcikowski, *Phys. Chem. Chem. Phys.* **15**, 3068 (2013)
 24. A. De Giacomo, M. Dell'Aglio, A. Santagata, R. Gaudiuso, O. De Pascale, P. Wagener, G. C. Messina, G. Compagnini, S. Barcikowski, *Phys. Chem. Chem. Phys.* **15**, 3083 (2013)
 25. M. Dell'Aglio, R. Gaudiuso, R. ElRashedy, O. De Pascale, G. Palazzo, A. De Giacomo, *Phys. Chem. Chem. Phys.* **15**, 20868 (2013)
 26. A. Vogel, S. Busch, U. Parlitz, *J. Acoust. Soc. Am.* **100**, 148 (1996)
 27. A. Vogel, J. Noack, K. Nahen, D. Theisen, S. Busch, U. Parlitz, D.X. Hammer, G.D. Noojin, B.A. Rockwell, R. Birngruber, *Appl. Phys. B* **68**, 271 (1999)
 28. T. Sakka, S. Masai, K. Fukami, Y.H. Ogata, *Spectrochim. Acta, Part B* **64**, 981 (2009)
 29. V. Lazic, S. Jovićević, M. Carpanese, *Appl. Phys. Lett.* **101**, 054101 (2012)
 30. S. Ibrahimkutty, P. Wagener, T. dos Santos Rolo, D. Karpov, A. Menzel, T. Baumbach, S. Barcikowski, and A. Plech, *Sci. Rep.* **5**, 16313 (2015)
 31. R. Tanabe, T. T.P. Nguyen, T. Sugiura, Y. Ito, *Appl. Surf. Sci.* **351**, 327 (2015)
 32. S.J. Shaw, W.P. Schiffrs, T.P. Gentry, D.C. Emmony, *J. Phys. D: Appl. Phys.* **32**, 1612 (1999)
 33. R. Petkovšek, P. Gregorčič, J. Možina, *Meas. Sci. Technol.* **18**, 2972 (2007)
 34. R. Petkovšek, P. Gregorčič, *J. Appl. Phys.* **102**, 044909 (2007)
 35. P. Gregorčič, R. Petkovšek, J. Možina, *Appl Phys A* **93**, 901 (2008)
 36. P. Gregorčič, M. Jamšek, M. Lukač, M. Jezeršek, *J. LA&HA* **2014**, 14 (2014)
 37. M.H. Mahdich, M. Akbari Jafarabadi, *Appl. Phys. A* **116**, 1211 (2014)
 38. L. F. Devia-Cruz, S. Camacho-López, V. R. Cortés, V. Ramos-Muñiz, F. G. Pérez-Gutiérrez, and G. Aguilar, *Appl. Opt.* **54**, 10432
 39. H. Oguchi, T. Sakka, and Y. H. Ogata, *J. Appl. Phys.* **102**, 023306 (2007)
 40. T. Sakka, H. Oguchi, S. Masai, Y.H. Ogata, *Chem. Lett.* **36**, 508 (2007)

Figure captions

Fig. 1 Experimental setup for laser-beam-transmission probe (LBTP)

Fig. 2 Typical transmission signals obtained for (a) 20-ns- and (b) 100-ns-pulse irradiations. The pulse energy is the same (6.0 mJ)

Fig. 3 Comparison of the signals attributed to the first oscillation of the bubble

Fig. 4 Comparison of the signals attributed to the subsequent oscillations of the bubble

Fig. 5 Comparison of the oscillation period of the bubble as a function of the bubble oscillation. The error bars correspond to the standard deviations of five measurements

Fig. 6 Comparison of the averaged signals of five measurements

Figures

Fig. 1

Fig. 2

Fig. 3

Fig. 4

Fig. 5

Fig. 6