

Article

Observation of Laser-Induced Bubbles in Glycerol–Water Mixtures

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Abstract: This work presents a method of directly imaging the growth and collapse of laser induced-bubbles in glycerol and water mixtures. The direct optical imaging is augmented with interferometric measurements of the vibration spectrum of the bubble-vessel system. Experimentation confirms the expectation that fluid viscosity affects the bubble formation and lifetime. During the experiment, deviation from the Rayleigh–Plesset equation is observed. Given this deviation of the observed bubble dynamics from the expected results, it is possible that the limited size of the sample volume and the walls of the container impact the bubble dynamics. The optical observations are supported by the observations of the system's vibration spectrum.

Keywords: laser-induced plasma; time-resolved imaging; cavitation bubble; viscosity

1. Introduction

Plasmas and bubbles formed during laser radiation of liquids are a complicated physical system with applications ranging from plasma medicine, synthesis, and catalysis of reactions, improvements in laser induced breakdown spectroscopy, and online process monitoring [1]. This work evolved from methods of quantifying and observing fertilizers in liquid environments using both Raman and laser-induced breakdown spectroscopy. As discussed in O'Neill et al., it is difficult to observe the atomic emission spectra of liquid samples used in laser-induced breakdown spectroscopy [2]. During attempts at observation, bubbles were observed, leading to this work.

In this work, observations of the growth and collapse of laser-induced cavitation bubbles in fluids of varying viscosity with hard walls as boundary conditions are described. Similar to the work of Fu et al. and Zhou et al., this work observes the formation of laser-induced bubbles with boundaries [3,4]. In contrast to the aforementioned works, this work looks at the effects of viscosity as opposed to laser intensity. This data has the potential to increase the body of knowledge beneficial to plasma medicine and online process monitoring using laser-induced breakdown spectroscopy or any situation in which the sample volume is well contained. This work expands on the work of Xiu-Mei et al. and Englert et al. on cavitation bubbles in viscous fluids [5,6]. Xiu-Mei et al. and Englert et al. observed bubble formation by measuring the variation in received power of a beam transmitted through the bubble process, while this work directly images the bubbles using a high-speed camera. To that end, mixtures of water and glycerol are illuminated with a pulsed doubled Nd:YAG laser, and the bubbles are directly imaged to measure their size. Vibration measurements of the sample vessel provide insight into the potential impact of the effect of the container and sample boundaries. In this work, the cross-sectional area of bubbles is directly measured with a high-speed camera, and the vibrational frequency of the sample volume is measured using interferometric methods.



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2. Description of the Bubble Process

The initial formation of the bubbles generated in the experiments are consistent with the processes that occur during nanosecond laser-induced breakdown spectroscopy as described in Lazic et al. and Kennedy et al. as well as the observed early bubble formation presented in the work of Jia et al. [7–9]. Figure 1 graphically illustrates this process. A plasma is created with the leading edge of the laser pulse through a combination of cascade ionization and multiphoton absorption. This plasma is then heated by the rest of the pulse, and the plasma expands. The high temperature of the plasma causes a bubble to form, and the plasma expansion leads to bubble expansion. This expansion continues, creating a cavitation bubble. As the plasma decays and cools, the interior pressure of the bubble drops, causing its radius to decrease. With enough energy, the bubble oscillates, rapidly increasing and decreasing in radius. This continues until the energy is dissipated and a steady state is reached. In addition to Figure 1, images of the process are shown in Section 3.4. The full bubble collapse and expansion process observed in this experiment occurs in the span of 1–2 ms.

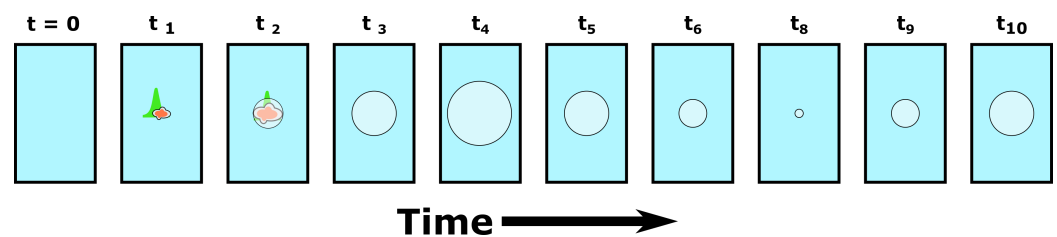


Figure 1. Bubble formation and expansion process. ($t = 0$) shows the cuvette at its initial conditions, (t_1) shows the initial laser heating and plasma formation, (t_2) shows the plasma expansion and bubble formation, and (t_3 : t_{10}) show the expansion and contraction of the bubble. This entire process occurs over approximately 1–2 ms.

3. Methods

The methods used to observe the bubble formation and decay are outlined in the following section, with a table containing relevant material properties. The bubble area is directly imaged with a high-speed camera, and the vibration spectrum is measured with interferometric methods.

3.1. Bubble Area

The optical setup and data processing steps are outlined in the following sections. Details regarding the specific components used are given.

3.1.1. Optical Setup

To generate the bubbles, an 8mJ 532 nm Nd:YAG laser with a 10 ns pulse width is used. The beam is expanded using a $5\times$ beam expander and then focused by a $10\times$ infinity corrected objective into a $12\text{ mm} \times 12\text{ mm} \times 45\text{ mm}$ sealed cuvette. To measure the bubbles created by the laser pulse, a Photron Mini AX200 high-speed camera (Photron, Tokyo, Japan) is coupled with a homemade infinite conjugate microscope with a spherical singlet lens. The Photron Mini AX200 has a 1024×1024 pixel resolution at 6000 fps, with reduced resolution at higher speeds. The maximum frame rate of is 540,000 fps. For this investigation, a moderate frame rate of either 50,000 fps or 80,000 fps is used when imaging the bubble formation. A 14 mW red LED (Thorlabs LEDD-630, Thorlabs, Newton, NJ, USA) is used for bright field illumination. Figure 2 shows the experimental setup, and components used. Contrast in the image is due to the difference in the index of refraction between the gas in the bubble and the water surrounding it.

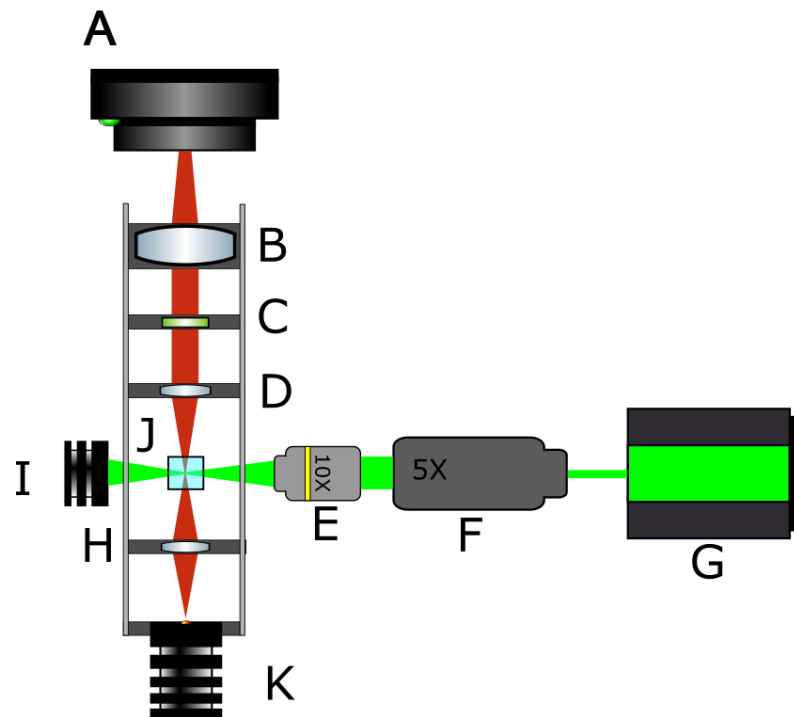


Figure 2. Diagram of the optical system used to measure bubble size. (A) Photron Mini AX200 high-speed camera, (B) Thorlabs TTL200-S8 tube lens, (C) 532 nm lowpass filter, (D) $f = 125$ mm bi-convex lens, (E) 10 $\times$ objective, (F) 5 $\times$ beam expander, (G) 8 mJ doubled Nd:YAG laser, (H) $f = 50$ mm plano-convex lens, (I) beam dump, (J) cuvette, (K) 635 nm LED.

3.1.2. Data Processing

Under bright field illumination of the sample, the bubble itself appears as a dark region with a bright spot in the center where the light has focused through it. A spatial Gaussian filter with a standard deviation of 8 in pixel intensity and kernel size of 33×33 pixels is applied to the images. This is implemented using the built-in function *imgaussfilt()* in MATLAB 2021. This has the effect of removing noise, the bright center spot, and background subjects that are not of interest. After filtering, the image is binarized; summing the black pixels and applying a calibration of pixels to the area allows the cross-sectional area of the bubble to be determined. The calibration used is an absolute calibration determined by imaging two targets of known size. Two precision irises, 100 μm and 200 μm , respectively, are imaged, and from that a system calibration is determined. Figure 3 shows the image acquired by the camera and the intermediate processing steps.

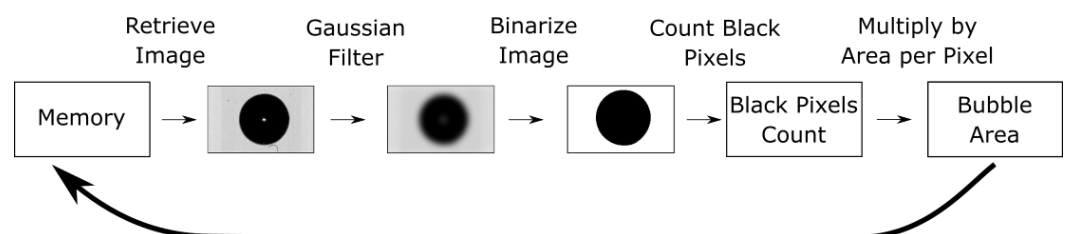


Figure 3. Raw image of the bubble captured by the camera, and the intermediate steps in processing it. The image is loaded from memory, a Gaussian filter is applied, the image is then binarized, and the number of black pixels in the image is counted. Given the calibration, the area is then calculated.

3.2. Vibration Measurements

Given the small sample volume, one cannot rule out the impact of the vibration of the vessel and feedback from the sample volume walls. To investigate the potential impact of

sample volume and cuvette vibrations, the vibration spectrum of the irradiated cuvette is measured with both deionized water and glycerol.

Optical Setup

In contrast to the work of Fu et al., this work measures the wall vibration spectrum optically as opposed to directly with a pressure transducer [10]. The sample volume is excited with the Nd:YAG laser used in the previous sections, and the vibration spectrum is measured with an extended Michelson interferometer as described in [11]. The output of the photodiode is sent to the input of a spectrum analyzer. At this time, only the frequency of oscillation is under investigation, not the direction; to that end, a homodyne interferometer is used. The experimental setup is shown in Figure 4.

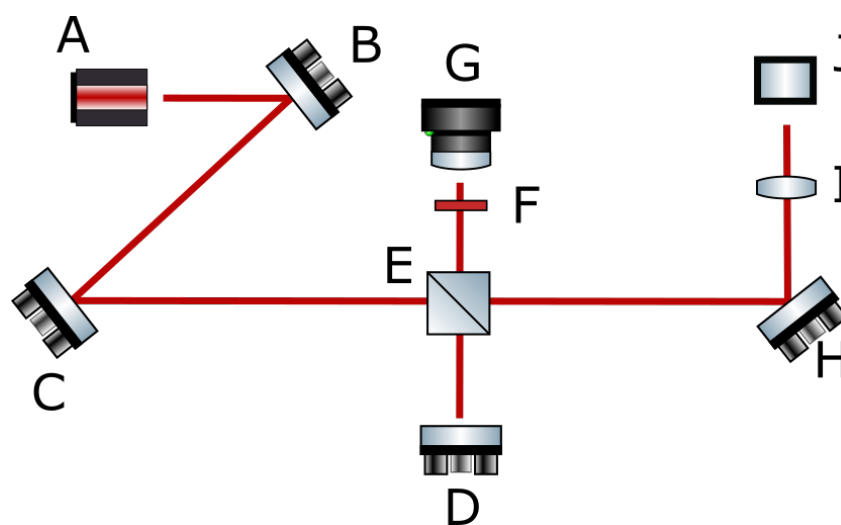


Figure 4. Michelson interferometer assembly: (A) HeNe laser, (B) fold mirror, (C) fold mirror, (D) reference mirror, (E) cube beam splitter, (F) 632 nm bandpass filter, (G) Thorlabs PDA36A silicon photodiode, (H) fold mirror, (I) spherical singlet lens, (J) cuvette.

3.3. Relevant Material Properties

Table 1 shows the material properties relevant to this experiment. Materials used in this experiment include deionized water and 99% anhydrous glycerol. To vary the viscosity of the solution investigated, water and glycerol were mixed. The viscosity of each sample and its density are calculated using the relations outlined in Cheng [12] et al. and programs developed by Volk and Kähler [13].

Table 1. Selected material properties of water and glycerol Mixtures.

Mass Concentration of Glycerol	Density (kg/m ³)	Dynamic Viscosity (Ns/m ²)	Absorption Coefficient (m ^{−1})	Speed of Sound (m/s)
0	996.89	0.001	0.0498 [14]	1500
0.25	1.06×10^3	0.002		
0.667	1.17×10^3	0.014		
0.75	1.19×10^3	0.027		
1	1.27×10^3	0.906	0.05 [15]	1964

3.4. Bubble Area

Selected images of the bubble expansion and collapse process taken with the microscope are displayed in the following figures. The full collection of images can be provided upon request. Figure 5 shows the expansion and collapse of a bubble in DI water, filmed at 50 K frames per second. Figure 6 shows the growth and eventual steady-state status of the

bubble formed in glycerin, filmed at 80 K frames per second. Figure 7 shows the measured area of the bubbles using the method outlined in Section 3.1.2. Using the data shown in Figure 7, the first derivative is taken to estimate the rate of growth of the bubbles; this is shown in Figure 8. All bubbles follow the general trend of a large expansion followed by smaller oscillations in size. There is variation in the area of bubbles and the number of oscillations. Figure 7C shows a prolonged collapse similar to that seen in Fu et al., and the behavior of the bubbles broadly matches what they reported [10].

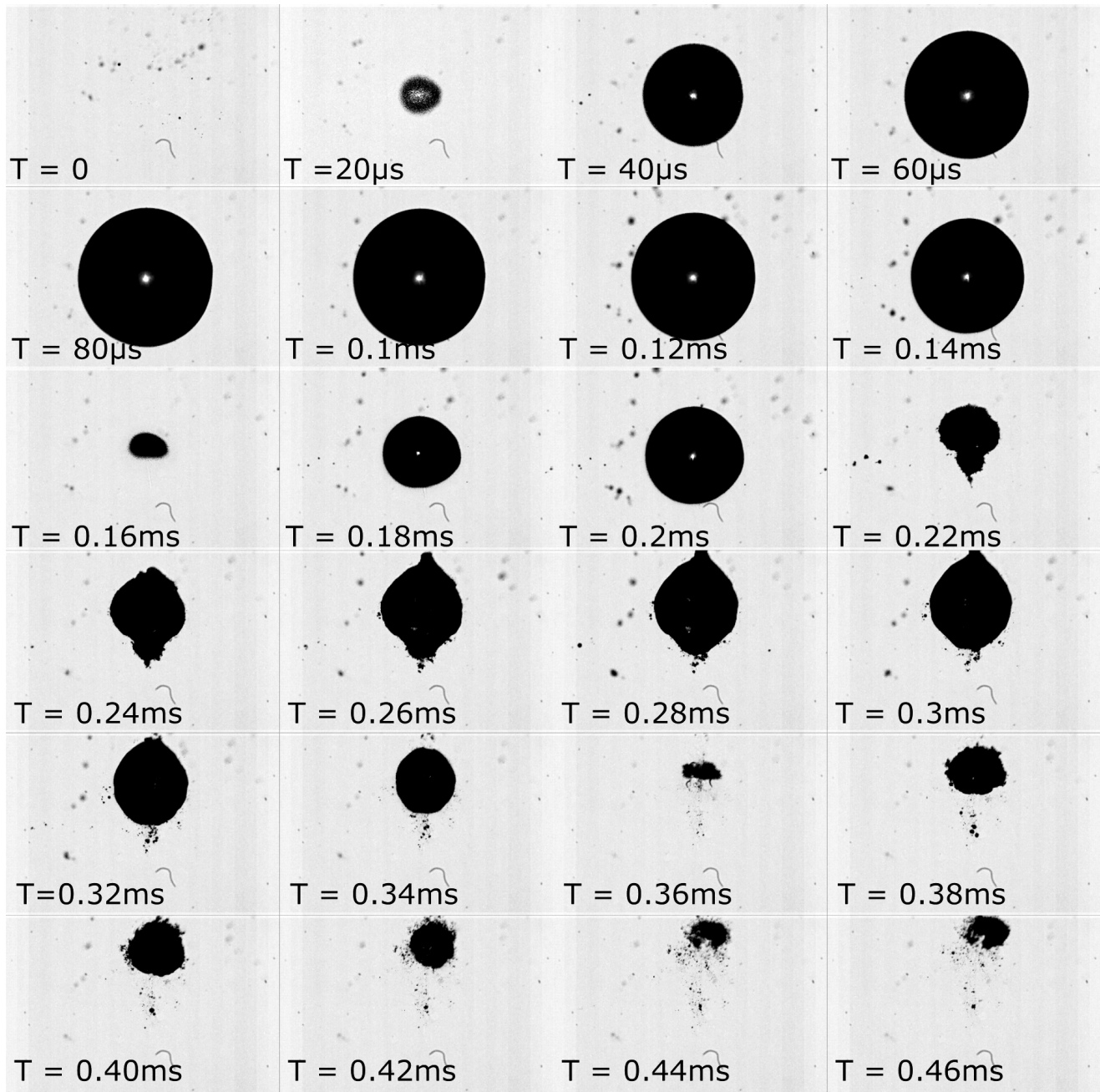


Figure 5. Captured images of the bubbles formed in water, filmed at 50 k frames per second.

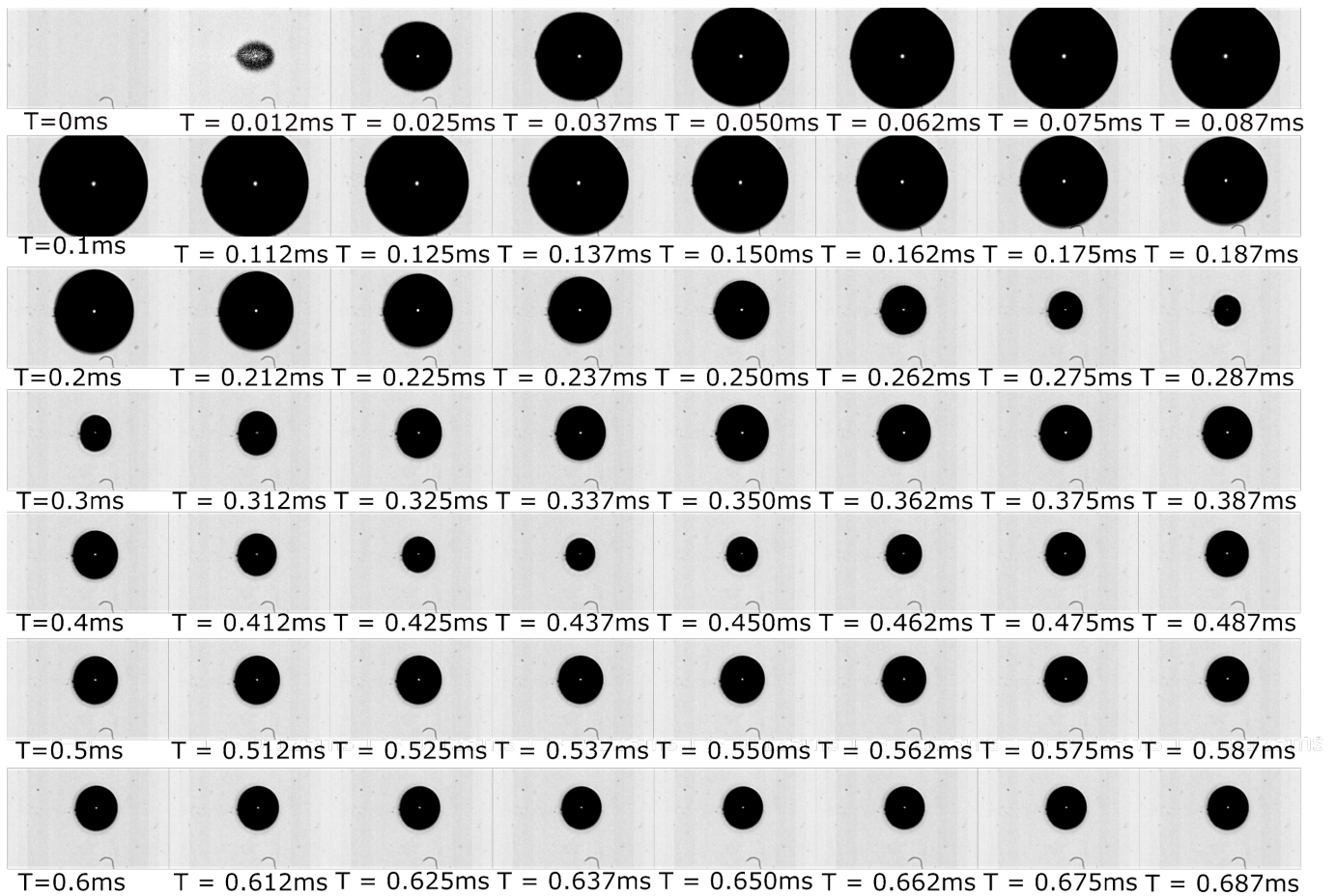


Figure 6. Captured images of the bubble formation in glycerol, filmed at 80 K frames per second.

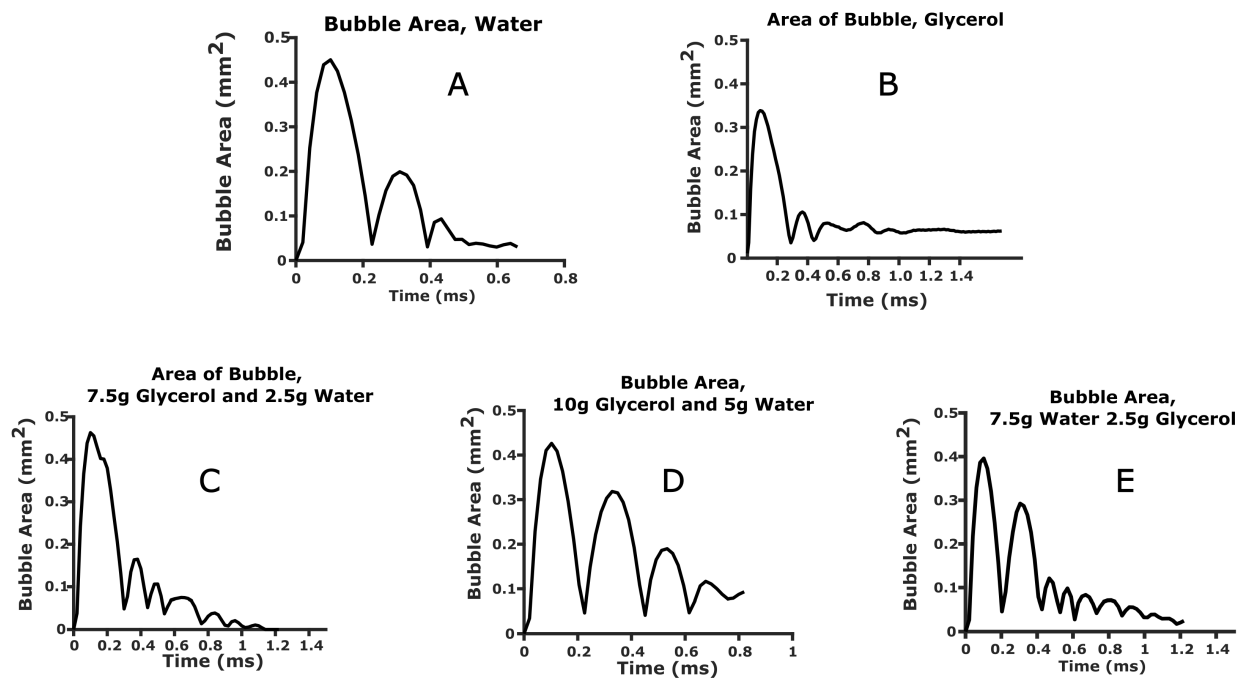


Figure 7. Plots of the area of each bubble with respect to time. (A) Area of the bubble in water. (B) Area of the bubble in 95% anhydrous glycerol. (C) Area of the bubble in a mixture of 7.5 g glycerol and 2.5 g water. (D) Area of the bubble in a mixture of 10 g glycerol and 5 g of water. (E) Area of the bubble in 7.5 g water and 2.5 g glycerol.

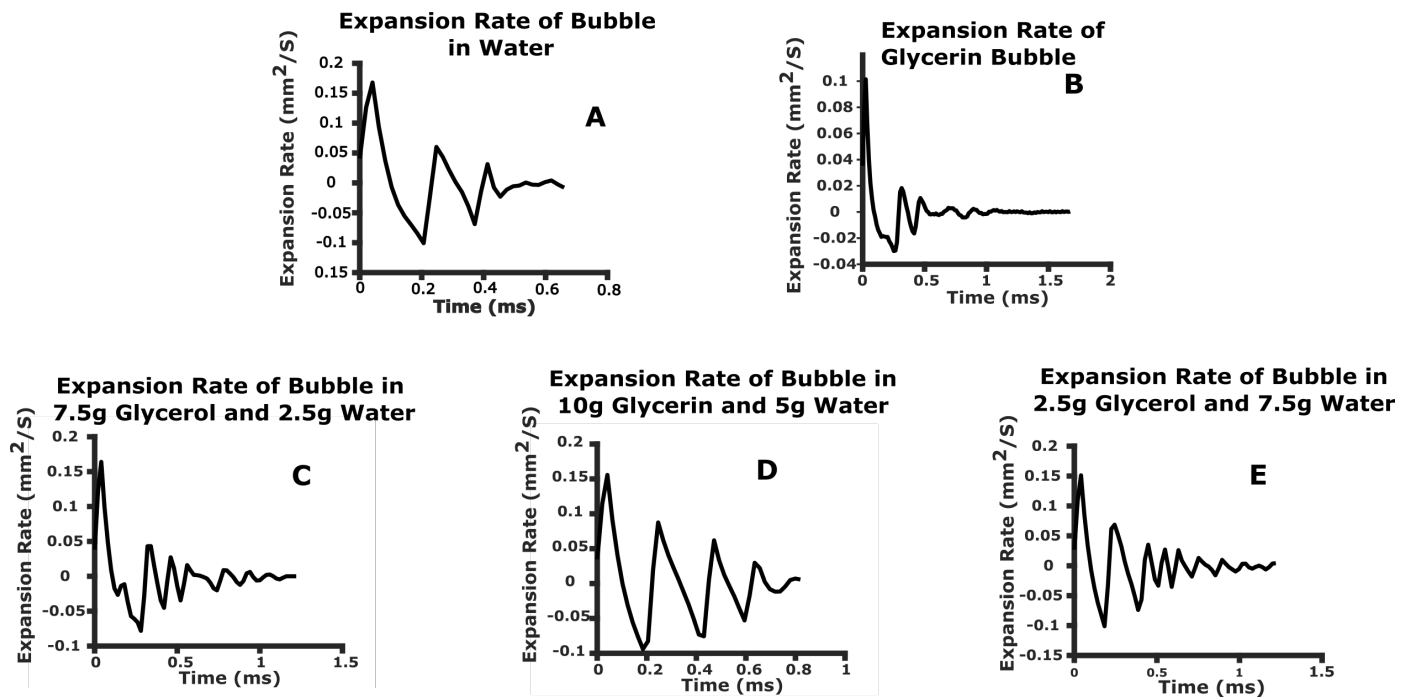


Figure 8. Plots of the rate of growth of each bubble with respect to time. (A) Rate of growth of the bubble in water. (B) Rate of growth of the bubble in 95% anhydrous glycerol. (C) Rate of growth of the bubble in a mixture of 7.5 g glycerol and 2.5 g water. (D) Rate of growth of the bubble in a mixture of 10 g glycerol and 5 g of water. (E) Rate of growth of the bubble in 7.5 g water and 2.5 g glycerol.

3.5. Vibration Spectrum

During the breakdown processes, an audible “pop” can be heard as the bulk movement of the material contributes to a vibration or mechanical response of the material and enclosing vessel. Figure 9 shows the measured vibration spectrum of the water, glycerol, and ambient system measurements on the left axis. The right axis shows the acoustic attenuation of glycerol estimated based on Stoke’s Law of Sound Attenuation [16]. The plot shows only the intensity of the vibrations at several frequencies. Using the setup shown in Figure 4, it is not feasible to measure the direction of motion using a homodyne Michelson interferometer. Both the water and glycerol measurements show peaks at 2.8 kHz, 3.3 kHz, and a cluster of peaks over the range of 4.5–7 kHz. The magnitude of each peak is higher with water than with glycerol. There are also three peaks at 7.6 kHz, 8.5 kHz, and 9.3 kHz present in the water spectrum but not the glycerol spectrum. The peak present at 8.5 kHz is close to the deduced wall vibration frequency discussed in Fu et al. [10]. The observations above make sense given the negligible acoustic attenuation of water and significant attenuation of glycerol. It is worth noting that the characteristic frequencies, assigned in Table 2, fit within the band of frequencies of 2–4 kHz shown in Figure 9.

Table 2. Qualitative descriptions of the oscillations.

Mass Concentration of Glycerol	Characteristic Frequency (kHz)	Envelope Decay Constant (1/s)
0	3.03	4924
0.25	2.73	2555
0.667	2.32	2010
0.75	2.49	3464
1	2.68	2860

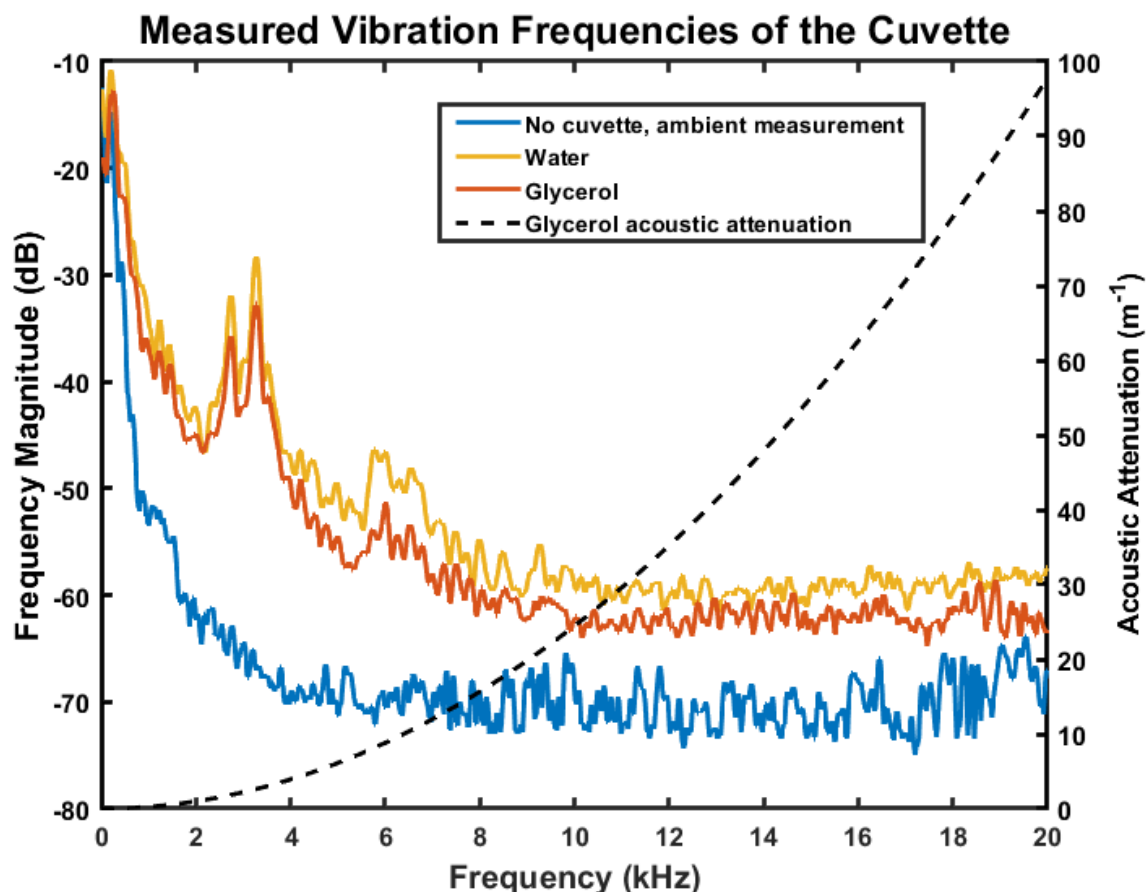


Figure 9. Measured vibration spectrum of the cuvette when water or glycerol is irradiated, with acoustic attenuation of glycerol plotted as well.

4. Discussion

The observed bubbles broadly follow the patterns predicted by the Rayleigh–Plesset Equation and Xiu-Mei et al. [5]. Variation and deviation from the Rayleigh–Plesset equation and the modified versions presented in Zhong and Eshraghi et al. are believed to be due to feedback from the walls of the cuvette and the limited volume of the sample chamber interacting with the fluid viscosity [17]. Table 2 shows qualitative descriptions of the behavior of the bubble. The decay constant is determined by fitting an exponential decay to the oscillation peaks. Noting the superficial resemblance of the area oscillations shown in Figure 7 to a full-wave rectified sine wave, a characteristic frequency is calculated from the time between the first and third peaks. The response of the bubbles does not appear to be linearly related to the concentration of glycerol or, subsequently, the density. That said, several observations can be made: the maximum bubble diameter decreases with the addition of glycerol; the time to the first “collapse” is generally longer with the addition of glycerol; with mass fractions of glycerol greater than 2/3, there is a significant damping effect on the subsequent expansions’ and the characteristic frequency assigned matches the band of frequencies from 2 to 4 kHz visible in Figure 9. The correlation between measured vibration frequencies and bubble characteristic frequencies implies a connection between the sample environment’s acoustic feedback and the bubble lifetime. This implication corroborates the work of Fu et al. and their conclusion that rarefaction waves from the bubble–wall system influence laser-induced bubble dynamics [10].

5. Conclusions

Presented in this work is an experimental investigation into the formation of bubbles under laser radiation in viscous and non-viscous fluids with hard wall boundary conditions. Bubble area is directly measured using a high-speed microscope. The potential interference in the bubble formation due to acoustic feedback from the sample volume walls is investigated by measuring the vibration spectrum of the vessel. To date, this is one of the few investigations to explicitly investigate sample volume oscillations and the only one to investigate them using non-contact methods. The presented work contributes to the development of further knowledge regarding plasma medicine and online process monitoring using laser-induced breakdown spectroscopy. Future work will benefit from multiple modalities of observation as well as variation in the size of the sample vessel. Sample vibrations should be measured with a heterodyne vibrometer and hydrophone, while the bubble area should be measured at higher spatial and temporal resolutions.

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Data Availability Statement: Due to the large volume of data and the size when compressed, the data are not hosted publicly but are available upon request.

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Conflicts of Interest: The authors declare no conflicts of interest.

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