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Peculiarities of Efficient Plasma Generation in Air and Water by Short Duration Laser Pulses

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Abstract

We have conducted experiments to demonstrate an efficient generation of plasma discharges by focused nanosecond pulsed laser beams in air and provided recommendations on the design of optical systems to implement such plasma generation. We have also demonstrated generation of the secondary plasma discharge using the unused energy from the primary one. Focused nanosecond pulsed laser beams have also been utilized to generate plasma in water where we observed self-focusing and filamentation. Furthermore, we applied the laser generated plasma to the decomposition of methylene blue dye diluted in water.

Introduction

Generation of plasma in the air, gasses liquids, and solids by laser beams and associated phenomena have been studied intensively using both CW and pulsed lasers (Refs. 1 to 11). Currently the technology has found applications in such diverse areas of technology as high energy physics (Refs. 12 and 13), plasma-assisted spattering and coating (Refs. 14 to 16), laser-induced breakdown spectroscopy (Refs. 17 and 18), and waste decomposition and cleaning (Refs. 19 to 21).

In aeronautics and space applications, laser generated plasma has been reported in areas of flow and noise control (Refs. 22 to 24) as well as plasma thrusters and ablation-driven propulsion (Refs. 25 to 28). Recently a demonstration of a supersonic shock wave mitigation by the laser-based energy deposition has also been reported (Refs. 29 and 30). However, practical implementations of the laser based energy deposition on a flying vehicle are severely limited by weight, size, and power requirements for the lasers. While focused laser beams are often mentioned in the studies, no attempts have been made to optimize the configuration of the laser beam focusing system. Such studies coupled with efficient generation of plasma discharges is especially needed where weight and size of the hardware and power consumption are important.

In many cases optical focusing systems are used to concentrate the incident laser light to such small volumes that energy density in these volumes is sufficient for breaking down the ambient gas or air. The effects of the optics on plasma formed as a result of the breakdown has also been a topic of study (Refs. 4, 31, and 32). The design of optical focusing systems in view of their efficiency to generate plasma and ability to manipulate it has become a significant issue.

An efficient plasma generating system, in addition to being sufficiently small in weight and size to be used in airborne applications, would permit to control the location of the plasma discharge in aerodynamic flows.

The development of efficient optical systems to generate laser driven plasma discharges should also benefit water purification processes. Currently the water purification processes employ plasma generation by electrical discharges (Refs. 33 to 35) and a short duration pulsed voltage technique has been especially attractive because the resultant non-thermal plasma does not lead to heating up the liquid media (Refs. 36 to 38). However, the high voltage nanosecond pulses generate a very strong pulsing electromagnetic field that may disrupt operations of electronic devices and instrumentation as well as interrupt cable and wireless communications.

To mitigate the effects of pulsed electromagnetic fields the plasma generating apparatus is confined to Faraday-like cages to keep the fields contained in enclosures. Such mitigation techniques lead to a significant increase in weight and mass as well in complexity of the entire plasma generating system. Moreover, the use of high voltage require special safety precautions as well as training and certification of operators. Thus, the use of focused pulsed laser beams may be an alternative to high voltage pulsing systems in water purification applications.

Plasma Generation in the Air

Among pulsed lasers used to generate plasma those with nanosecond pulse duration and operating at the 2nd harmonic of the Nd:YAG fundamental wavelength of 1064 nm (Refs. 39 to 41) are especially attractive since they are in general lighter, smaller, and provide satisfactory power and energy requirements for plasma generation.

To demonstrate the impact of the efficient design of the optical system, an experimental setup employing a pulsed Nd:YAG laser operating at the 2nd harmonic (λ = 532 nm) was constructed. The setup consists of the laser and an optical system. The laser in the setup has a pulse duration of about 5 nsec, repetition rate of about 15 Hz, and a maximum energy per pulse of about 120 mJ. The maximum consumption of electrical power during the laser operation is 1.15 W. The optical system focuses a nearly collimated laser beam that exits the laser aperture into a small volume with the energy density sufficient to generate plasma. It was also constructed to achieve the plasma generation with the minimum energy per pulse required.

While perhaps many optical system could be constructed to focus laser beams into a small spot, the optical system in our setup has two parts, a collimator and a focusing lens. The collimator is a Galileo telescope, the first lens facing the incoming laser beam being negative.

The plasma generating setup is shown schematically in Figure 1. Lenses 1 and 2 together form a Galileo telescopic system in which the magnification is determined by a ratio of focal lengths of Lens 2 to Lens 1. The selection of the Galileo telescopic system prevents formation of a plasma spark between Lenses 1 and 2. The distance d_{12} between Lenses 1 and 2 is an algebraic sum of focal lengths f_1 and f_2. Thus, the geometrical distance $d_{12} = f_2 - f_1$. Lens 3 focuses the beam into its focal point F_3 where the plasma is generated.

During the experiments the energy and power of laser pulses were measured at three locations along the direction of laser propagation, at the laser exit aperture, before the focal point F_3 where the plasma was generated, and after that point. The three positions of the power meters are shown in Figure 1 and marked as “Power Meter Position I”, “Power Meter Position II”, and “Power Meter Position III, correspondingly. Moreover, positions of the power meter before and after the focal point F_3 were nearly equidistant from that point.

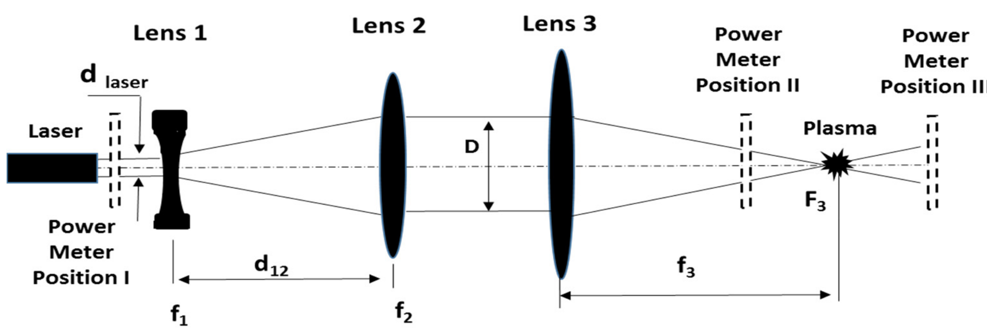


Figure 1.—Schematics of the experimental optical setup to generate plasma.

TABLE 1.—DEPENDENCY OF PLASMA GENERATION ON THE OPTICAL SYSTEM PARAMETERS

Lens L_1 f_1	Lens L_2 f_2	Lens L_3 f_3	Minimum energy	Minimum power
mm	mm	mm	mJ	mW
–50	150	85	18 to 23	300 to 360
–50	150	100	26 to 29	400 to 430
–50	150	150	59 to 61	900 to 910
–50	150	200	No plasma	No plasma
–50	200	85	~10	155 to 170
–50	200	100	16 to 17	250 to 265
–50	200	150	20 to 22	290 to 310
–50	200	200	68 to 70	900 to 1050

The first set of measurements was intended to demonstrate the dependency of the plasma generation on the constituent parameters of the optical system involved. In the process we changed lenses L_2 and L_3 and the distance d_{12} accordingly to maintain the Galilean telescopic configuration. The laser pulse energy and power were measured by the power meter in Position III and while the plasma was still generated the initial pulse energy emitted by the laser itself was decreased until the plasma disappeared. The minimum values of the pulse energy and power at which the plasma disappeared were recorded. It is seen from Table 1 that the most efficient generation of plasma in our setup is associated with a larger D and a shorter f_3 where a larger D is associated with a longer focus length f_2 and a shorter focal length f_3. Since in a telescopic optical system the ration of absolute values f_2/f_1 is a linear magnification of the system, an increase in f_2 also leads to a corresponding increase in the beam diameter D, $D/d_{laser} = f_2 /f_1$. Thus, the results show that a larger D and a shorter f_3 lead to a more efficient generation of plasma. That could be explained from the fact that a larger D and a shorter f_3 concentrate more energy into a very small volume around the focal point and trigger a formation of volumetric plasma at lower initial pulse energy levels. Thus, since the changes in the constituent parameters lead to a change in the energy density in a small volume along focal point F_3, not all combinations of those parameters necessarily lead to generation of plasma.

The constituent parameters of the optical system, namely the focal lengths and mutual positions of lenses, determine the location of the plasma discharge. For instance, since a change in the position of Lens 1 leads to a violation of the Galileo configuration of the telescopic system, the plasma discharge will appear at a point different from the original point F_3. Specifically, if the distance d_{12} was increased the position of the plasma discharge would move closer to Lens 3.

Figure 2 shows schematically positions of lenses in near-Galileo configurations with focusing lenses as well as hypothetical locations of plasma discharges. The schematic represents three cases with Figure 2(a) being the Galilean telescopic configuration with the distance d between lenses L_1 and L_2, using optical sign convention, $d = f_1' + f_2'$, were f_1' and f_2' are focal lengths of lenses L_1 and L_2 respectively. Figures 2(b) and (c) show schematically the positions of the plasma discharges when $d > f_1' + f_2'$ and $d < f_1' + f_2'$ respectively. The variations in the position of the discharge can be achieved by moving lens L_1 with a negative focal lens while keeping relative positions of lenses L_2 and L_3 unchanged.

As is seen from Table 1, the efficient generation of plasma requires a large diameter D and short focal length f_3. That means that Lens 3 in Figure 1 has a large spherical aberration and as a result the plasma discharge at F_3 has a spheroid elongated along the beam propagation.

In the second set of measurements we used the system configuration with f_1 = –50 mm, f_2 = 200 mm, and f_3 = 100 mm. The initial pulse energy was sufficient to generate plasma at the focal point F_3 and the pulse energy was measured at three locations as shown in Figure 1. The measurements were conducted twice at two different values of the initial pulse energy measured at the Power Meter Position I. Results of the measurements at corresponding positions of the power meter are presented in Table 2. The table shows that only about 20 percent of the energy contained in a single pulse was consumed in the experiment during the plasma generating process. The unused pulse energy could then be collected and focused to generate a secondary plasma discharge by an optical attachment as shown in Figure 3.

The optical attachment to generate the secondary plasma discharge has, in general, two optical components. The first one, Lens 4, is placed after the plasma discharge that occurs near focal point F_3 of Lens 3. It acts as an optical collector and collects the pulse energy contained in the laser beam after passing through that focal point. The second one, Lens 5 acts as a focusing lens and forms another plasma discharge after Lens 5 as shown in Figure 3.

Generation of plasma is accompanied by emission of audible sound (Refs. 42 and 44) and high frequency electromagnetic waves (Refs. 45 and 46) as well as visible and ultraviolet light (Refs. 47 and 49).

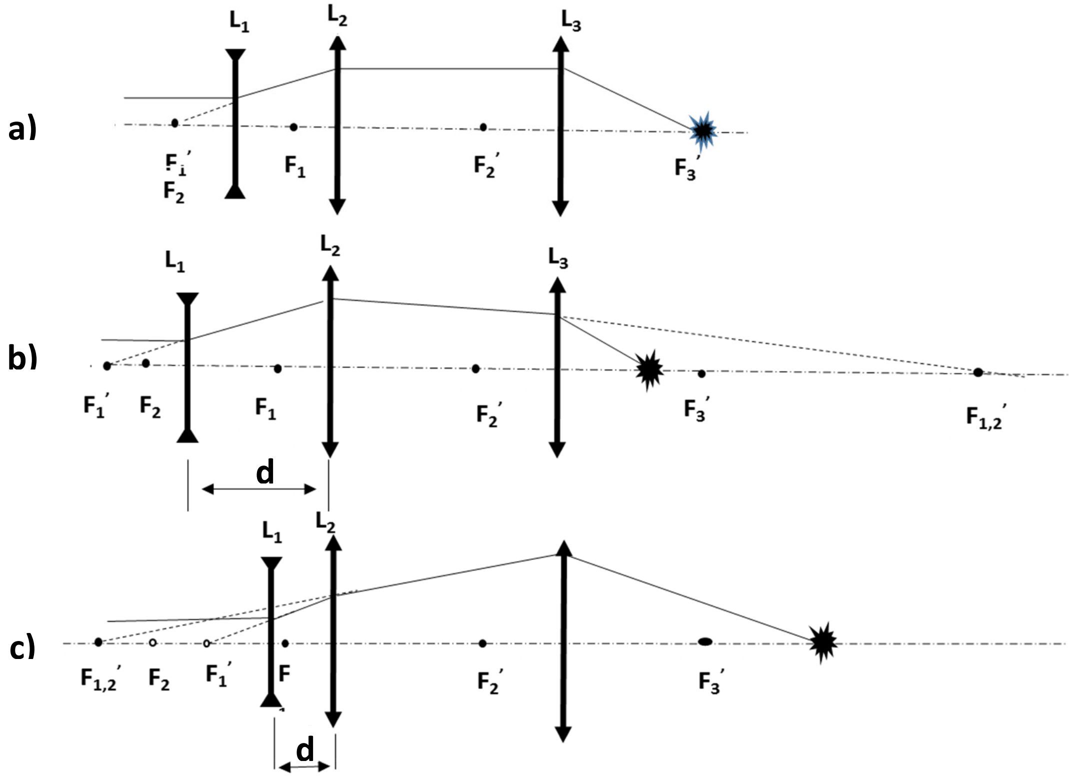


Figure 2.—Examples of plasma discharge generating optical systems. (a) Galilean telescopic system, $d = f_1' + f_2'$; (b) Optical system, $d = f_1' > f_2'$; (c) Optical system, $d = f_1' < f_2'$.

TABLE 2.—EXAMPLES OF THE PULSE ENERGY BEFORE AND AFTER FORMING PLASMA

Power meter position II	Power meter position III
70 mJ	55 to 60 mJ
27.5 to 29.5 mJ	22 to 24 mJ

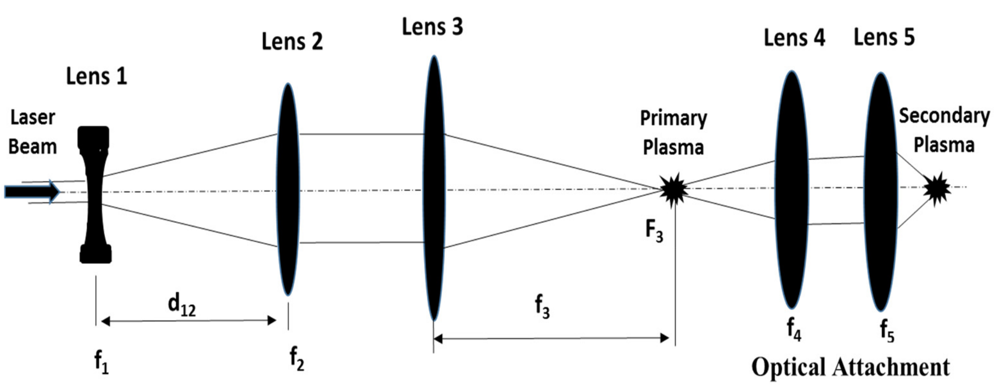
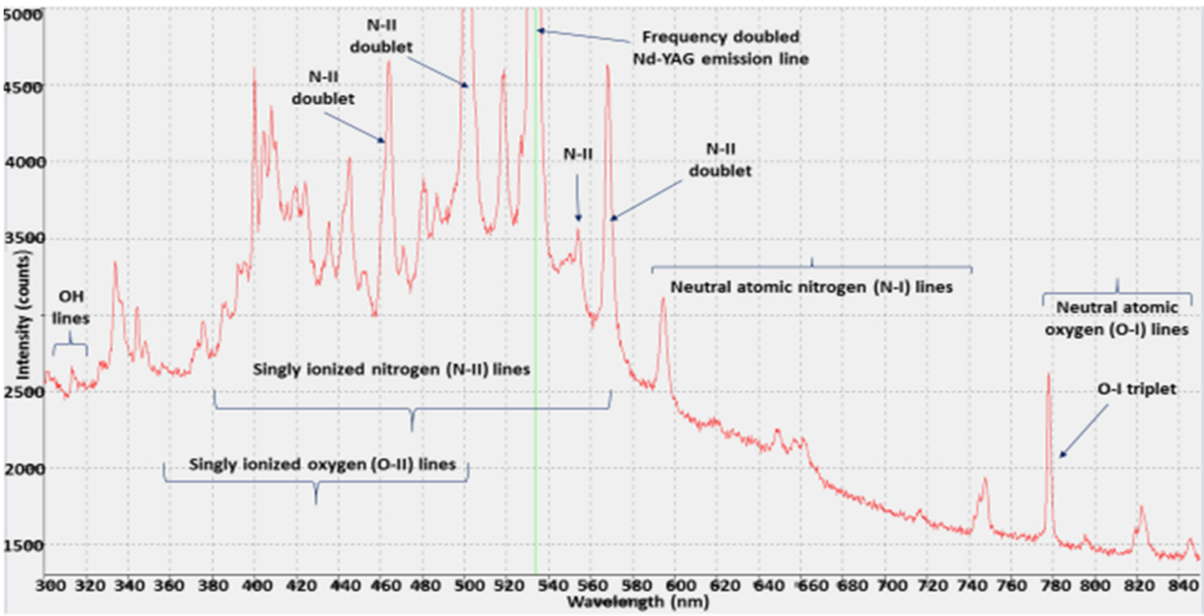


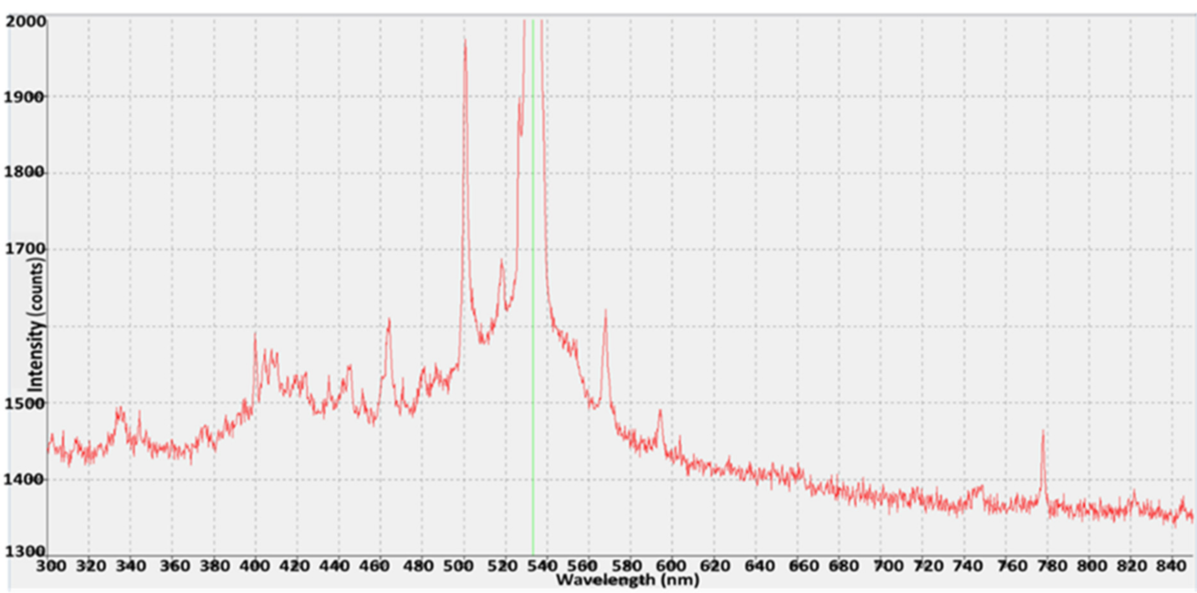
Figure 3.—Schematics of an optical system to generate a secondary plasma discharge.

The spectra of the plasma discharges at both, the primary and secondary positions were analyzed by a spectrometer. The spectrometer had a GRIN light collector at the end of a fiber optic cable that fed the collected light to the spectrometer for processing. The light collector was placed close to the plasma discharges in the direction normal to the direction of the incident beam propagation.

Examples of the observed spectra of the primary and secondary plasma discharges are presented in Figure 4. Both spectra show a presence, in addition to the frequency spectrum of the incident laser pulse, spectrum bands of both neutral atomic and singly ionized N and O lines at locations similar to those that have already been reported (Refs. 50 and 51) as well as OH lines that appear at 307 and 309 nm (Ref. 52). Among the neutral atomic oxygen lines the strongest is located around 775 nm and is associated with a neutral atomic oxygen (O-I) triplet that occurs at the 777 nm wavelength. Strong lines associated with nearly doublets N-II lines are also clearly visible.



(a)



(b)

Figure 4.—Spectra of plasma discharge. (a) primary discharge. (b) secondary discharge.

Both spectra have similar features and show a presence of spectra lines associated with the dissociation of the main components of air, molecules of O_2 and N_2.

The presence of a strong spectral component at the fundamental frequency of the incident laser beam in the spectrum in Figure 3 and data presented in Table 2 also suggest that the focused laser beam, in addition to generating the plasma discharge, is also a source of radiation at the fundamental incident laser wavelength.

The optical system shown in Figure 3 was not optimized for minimum losses the secondary plasma discharge was barely visible to naked eye on the bright background. The presence of the discharge was made visible upon reflection of the laser beam formed by Lens 4 and Lens 5 from a screen as depicted in Figure 5.

The analyses of light spectra emitted by the plasma discharge at the point F_3 can be enhanced by placing a light condenser between the discharge and the fiber optic light collector of the spectrometer as is shown in Figure 6.

The condenser has to have a wide acceptance angle to collect as much light from the plasma discharge as possible and at the same time be corrected for chromatic aberration over a very broad range of wavelengths. Thus, to achieve the maximum performance for certain ranges of wavelengths, the distance between the condenser and the light collector has to vary. In Figure 6 the light condenser has the focal length of 85 mm and diameter of about 52 mm. The distance between the light condenser and fiber optic light collector was varied to maximize the amount of light focused at the location of the fiber optic light collector for various regions of the light spectrum emitted by the plasma.

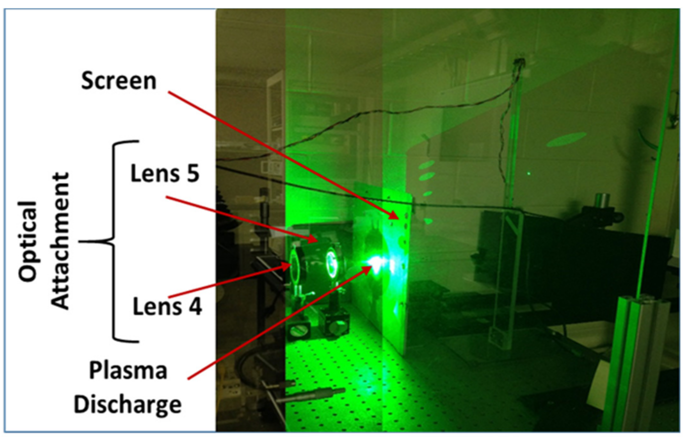


Figure 5.—Formation of a secondary laser discharge.

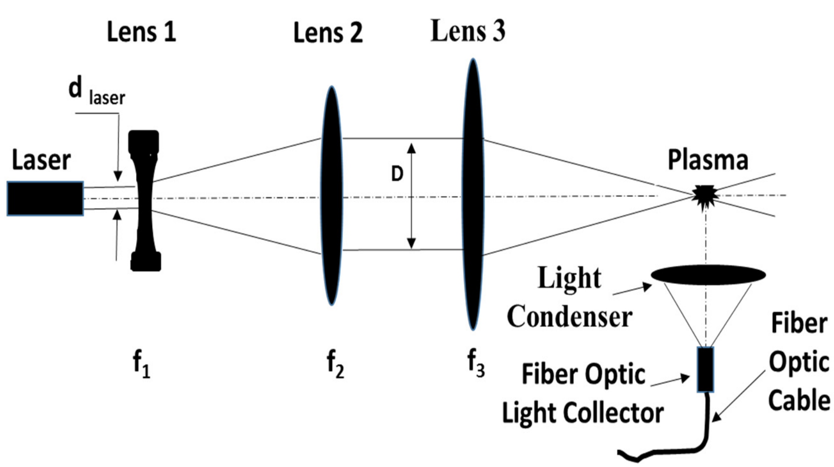


Figure 6.—Example of optical arrangement to collect light from plasma discharge.

Plasma Formation in Water

General properties of plasma generated in water have been described in the literature (Refs. 53 to 55) including of plasma generated by nanosecond laser pulses (Refs. 56 to 58). The process of plasma generation in aqueous media by laser pulses is also accompanied by formation of bubbles, shock waves acoustic, and ultrasound waves, as well as emission of UV and visible light (Refs. 59 to 62).

A comparison of plasma discharges observed in air in an empty cylindrical jar and the same jar filled with distilled water is shown in Figure 7.

The setup to generate plasma shown in the Figure 7 employs an optical arrangement similar to that described in Figure 1 and provides an efficient plasma generation. During the experiment an empty glass container is placed in the path of the beam in such way that the plasma discharge appears approximately in the middle of the container. To compensate for losses introduced by the glass walls of the container, the laser pulse energy is slightly increased. Figure 7(a) shows a photograph of the plasma discharge inside the empty glass container.

The water in the jar introduces more loses and in order to trigger plasma discharges the initial laser pulse energy is further increased to achieve plasma generation without changes in the laser focusing optics. Formation of a bright channel of light and multiple plasma discharges in the water is clearly observed and depicted in Figure 2(b). The light channel and discharges are formed along the longitudinal axis of the incident laser beam and the discharges are concentrated in the vicinity of the middle of the container. The location of the concentration of discharges changes with the position of the focusing Lens 3 (See Fig. 1).

The appearance of a light channel and multiple discharges are associated with non-linear processes, such as self-focusing and filamentation, triggered by high intensity laser pulses (Refs. 63 to 66).

To demonstrate the operation of system in polluted water we have used an aqueous solution of methylene blue, a non-hazardous dye commonly used for these purposes (Refs. 67 to 69). A commercially available 1 percent solution of methylene blue dye in water has been further diluted to 0.00025 percent or about 2.5 ppm and 100 mL of the resultant solution were placed in a square jar and exposed to the laser radiation focused inside the jar. The solution was exposed to laser radiation in 90 min intervals for a total of 6 hr. At the end of each interval the jar with solution was photographed and then placed back into the laser beam. Also, during the 90 min intervals the amount of the pulse energy that propagated through the solution was recorded using a high energy power meter connected to a laptop. An identical sample of polluted water was also exposed to continuing illumination of the focused laser beam over 6 hr without interruptions. The results were identical to a naked eye.

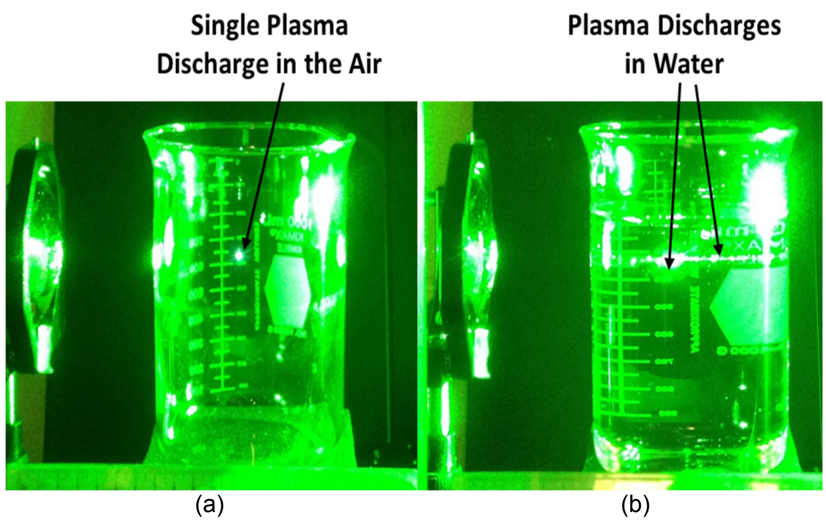


Figure 7.—Formation of plasma discharges. (a) in the air. (b) in water.

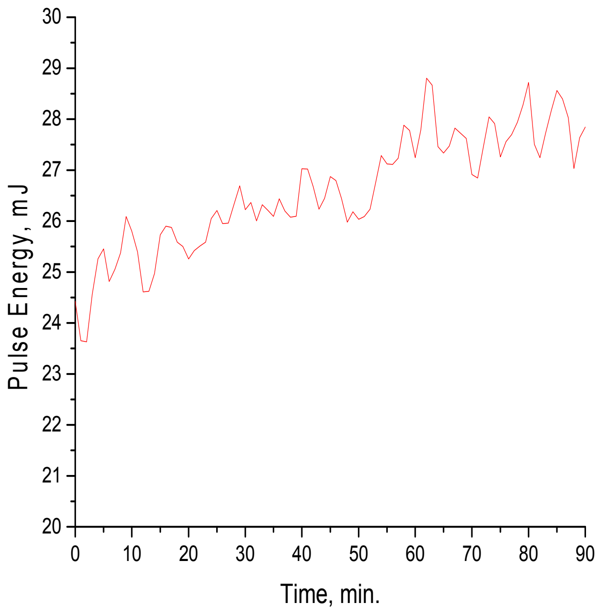


Figure 8.—Pulse energy readings over 90 min exposure to laser plasma.

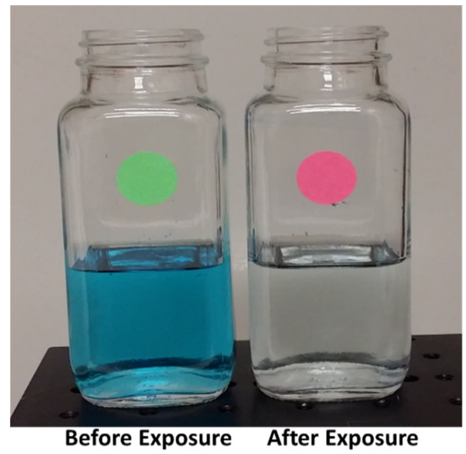


Figure 9.—Contaminated water before and after exposure to laser radiation.

The trend in the power meter readings over one of the 90 min periods and pictures of contaminated water before and after 6 hr long exposure to the laser radiation are shown in Figures 8 and 9 respectively. As expected, Figure 8 shows an increase in energy readings during the period of recording due to clearing of the water in the jar. Figure 9 shows photographs of the solution in the jar before and after 6 hr long exposure. In Figure 9 the jar to the left with a green dot contains a control sample that represents the contaminated water solution before the exposure and the jar to the right with the red dot has the solution after the exposure. The laser pulse energy measured before the beam entered the jar was between 66 and 68 mJ.

The plasma generated in the water by a laser beam was centered approximately at the center of the jar and the plasma discharges formed around the beam decomposed methylene dye in the vicinity of the beam. To speed up the process we introduced bubbles in the water and did see improvements. An increase in the pulse repetition rate would also speed up the plasma triggered decomposition of the dye in water. However, the repetition rate should be maintained at a slow enough rate to provide a sufficient time interval between pulses to maintain the plasma in a transient state.

Conclusion

We have demonstrated some peculiar properties of plasma generated by nanosecond pulsed laser beams in air and water. First, in the air we have demonstrated an efficient way of generating plasma by using properly arranged focusing optics and confirmed findings by others. Moreover, we have demonstrated that the efficient focusing optic can generate secondary discharges by collecting the energy that is not used during the first discharge and focusing it again in a different location. The secondary discharges can be generated by optical attachments that are similar to those used in this paper. They also may be different and involve prisms, reflectors, and other optical components to change the location of the secondary plasma discharges.

It is clear from the selected configuration of the optical focusing system that the location of the plasma discharge depends on the focal length of Lens 3. A shorter focal length with the same beam diameter will bring the plasma discharge closer to the surface of the lens. Since a large ratio of the beam diameter to the focal length violates the assumption of the paraxial principle and leads to large spherical aberrations, the resultant plasma discharge becomes elongated along the longitudinal beam axis. In cases when such phenomenon is beneficial further enhancement of spherical aberration could be achieved with a conical lens in place of Lens 3.

The location of the discharge can also be changed by changing relative position of Lens 1 and Lens 2. That permits to locate the discharge, for instance, in aerodynamic flow at a certain distance from Lens 3 which could be permanently installed in the body of a moving object or flying vehicle.

We have also demonstrated that focused nanosecond duration laser pulses produces plasma discharges in water and observed phenomena that had been usually seen with femtosecond lasers, such as self-focusing and filamentation. We believe that these phenomena had assisted with the distribution of the plasma discharges and production of the ionized byproducts. Utilization of optical configurations similar to those described in the paper make the generation of the plasma even more efficient.

Since we demonstrated that the unused portion of the incident laser beam can be used to form secondary plasma discharges, that feature could further improve the formation of oxidizers in water and assist with decomposing water pollutants. To achieve that, special optical attachments have to be designed to collect the unused laser energy, redirect and refocus it in the area of interest.

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