



Successive laser-induced breakdowns in atmospheric pressure air and premixed ethane–air mixtures



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ARTICLE INFO

Article history:

Received 27 September 2013

Received in revised form 3 December 2013

Accepted 31 December 2013

Available online 22 January 2014

Keywords:

Combustion stabilization

Repetitive laser ignition

ABSTRACT

Two successive focused laser pulses are employed to experimentally simulate laser-induced breakdown plasmas at high repetition rates. We find that energy absorption of the second laser pulse by the plasma produced by the first laser pulse is enhanced slightly when the time interval between the pulses is shorter than several tens of nanoseconds but falls to almost zero when the time interval is between a few hundreds of nanoseconds and several tens of microseconds. This behavior is attributed to gas heating by the first breakdown event. In premixed ethane–air mixtures, we identify another strong reduction in the second laser pulse absorption when this pulse coincides with the heat released by combustion, typically milliseconds after the first laser pulse. The fuel–air equivalence ratio (ϕ) and base flow speed are also varied in this study. The results show that the window of reduced absorption coinciding with heat release due to combustion is narrowed when the base flow speed is increased, and also under fuel lean and fuel rich conditions. These results suggest that the use of pulsed high frequency laser breakdowns for premixed combustion stabilization is optimized when laser pulse repetition rates below a certain frequency (e.g., 500 Hz at the conditions that ϕ is 1 and the base flow speed is 4.9 m/s) to maximize laser energy coupling and for improved anchoring of the flame base.

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1. Introduction

Pulsed plasmas have been shown to stabilize premixed flames at fuel-lean conditions [1–5] and diffusion flames at blow-off conditions [6]. Most pulsed plasmas used in these studies [1–4,6] have been produced between the electrodes with high applied potentials (several kVs), short pulse durations (tens of nanoseconds), and high repetition frequencies (several tens of kHz). One of the shortcomings associated with the use of electrode-driven pulsed discharges, particularly at high pressure, is that the discharges have to be placed in regions of the flow where the plasma that is generated has the greatest stabilization efficacy. Electrodes are also relatively immobile and invasive. Their presence can influence the flame dynamics, and in hot combusting environments the electrodes can experience considerable erosion and degradation. These drawbacks can be eliminated by replacing these discharges with a focused laser beam. The laser breakdown plasma can be placed, by design of appropriate optics, in any position within the flow. While the use of laser-induced sparks has been studied as possible replacements for conventional spark plugs [7,8], their use in flameholding has not been extensively studied. Flameholding requires a continuous ignition source to anchor the combustion under blow

off conditions. A recent study reported on the use of 1 kHz repetition rate femtosecond laser-induced plasmas to stabilize lean premixed methane–air flames [5]. Theoretical studies have also been carried out on flame stabilization using continuous ignition sources [9]. Plasmas produced by laser repetition rates higher than tens of kilohertz and their interaction with flames have not been investigated in detail, despite conventional wisdom, that would suggest that such higher repetition rates may have a higher efficacy for flame anchoring.

Besides its potential application in combustion, laser-induced breakdown has been studied and now developed as a tool for element composition analysis of both solids and gases [10,11]. In so-called laser-induced breakdown spectroscopy (LIBS), multiple laser pulses with short times between them have been exploited to enhance the ensuing plasma emission intensity [12]. In the study of air [13], this enhancement in emission was found to occur only when the time between successive laser pulses was less than tens of nanoseconds. Increased time between pulses resulted in a near transparency of the air to the second laser pulse. Schlieren images taken of the post-plasma associated with laser breakdown of air at 2 kHz [14] reveal that the gas density is significantly distorted by the breakdown events. Clearly, the effects of previous breakdown events carry over to affect the breakdown ability of subsequent laser pulses. This is expected to be important in the use of high repetition rate laser breakdown in flame stabilization.

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In this paper, two successive 532 nm wavelength laser pulses of 8 ns temporal width have been employed to experimentally simulate the efficacy of a high frequency laser-induced plasma in combustion stabilization. We vary the time interval between these laser pulses, from tens of nanoseconds to several milliseconds (corresponding to an effective laser repetition rate between tens of megahertz and hundreds of hertz), and examine the coupling of pulse energy to the plasma generated in air, and in premixed ethane–air mixtures. In the later case, the fuel–air equivalence ratio and base flow speed are varied to understand the effects that successive breakdown laser pulses have on the plasma energy coupling and combustion stabilization. The flame dynamics (e.g., CH^* chemiluminescence) initiated by successive laser pulses is recorded by high speed photography. Experiments were also performed in methane–air and propane–air mixtures but are not discussed here as the results are qualitatively (and quantitatively) similar to those for ethane–air cases.

2. Experimental setup

A schematic of experimental system used to study laser-induced breakdown plasmas produced by two successive laser pulses in atmospheric pressure air is shown in Fig. 1a. The same setup is used for breakdown studies in premixed flames except that the air supply system is replaced with a nozzle that supplies an ethane/air mixture. A detailed diagram of the laser-induced ignition and flame stabilization experiment with relevant

dimensions and a superimposed photograph of the flame are shown in Fig. 1b. A frequency-doubled Gemini PIV-200 Nd:Yag laser (ESI, New Wave Research) operating at 2 Hz provides two successive 532 nm wavelength laser pulses each having a temporal width and diameters of 8 ns and 8 mm, respectively. The time interval between the two successive laser pulses can be varied from tens of nanoseconds to several milliseconds. The laser output is focused to produce a breakdown plasma using a 2.54 cm diameter coated Plano-convex spherical lens (Thorlabs N-BK7, -A). Lenses of different focal lengths, ranging between 2.54 cm and 17.5 cm, are used in these studies. The laser power is measured at locations before and after the focusing lens and plasma by a laser power meter (Ophir Optronics, Inc.) set to time-average over thirty seconds (approximately 60 laser pulse pairs). The amount of laser energy absorbed by the plasma is determined by subtracting the measured transmitted power from the incident power and dividing it by the laser repetition rate. Experiments are first carried out with just a single laser pulse, and then compared to that obtained for a pulse pair to determine the contribution to absorption by the second time-delayed pulse. For experiments in premixed ethane–air mixtures, the time-resolved CH^* flame chemiluminescence is recorded using a PI-MAX intensified charge-coupled device (ICCD) camera synchronized to the laser by a delay generator (SRS, DG 535). The flame emission is recorded with a 10 nm-bandpass filter centered at 430 nm. The image field of view is 9.1 mm in width and 25.1 mm in height and the breakdown kernel forms 3.3 mm above from the bottom of the image field. The temporal gate width is set to be 100 μs when the equivalent ratio (ϕ is between 0.75 and 1.5 and 200 μs when ϕ is either 0.5 or 2.0. The captured images are averaged over twenty-five laser pair events. For breakdown experiments in air, the base flow speed is 1.4 cm/s (upward, as depicted in Fig. 1a). The air is filtered by a high efficiency particulate air (HEPA) filter (26.67 cm by 25.4 cm) to prevent dust from affecting the laser breakdown process. For the breakdown experiments in premixed ethane–air, the nozzle diameter is 9.8 mm, the base flow speed is varied between 3.3 m/s and 6.5 m/s, and ϕ ranges between 0.5 and 2.0.

3. Experimental results

3.1. Laser-induced plasma generated by a single laser pulse in air

The minimum (threshold) energy needed to produce breakdown by single laser pulse in air and its dependency on the lens focal length is shown in Fig. 2a. In general the threshold energy decreases with decreasing focal length, as expected for a Gaussian beam in which the laser beam waist is inversely proportional to the square of the focal length [15]. However, the threshold energy determined with a 2.54 cm focal length is found to be slightly greater than that determined for a 5 cm focal length because of the anticipated increased distortion in the laser pulse wavefront due to the spherical aberration associated with the increased lens curvature [15]. For our experimental conditions, we find that a 5 cm focal length lens results in the lowest breakdown threshold energy (approximately 2.5 mJ).

The laser energy that is absorbed by the plasma for a single laser pulse and its variation with incident pulse energy is depicted in Fig. 2b. The figure includes the results for focal lengths of 5 cm, 10 cm, and 15 cm. The results are not corrected for energy loss resulting from the scattering from the front and rear surfaces of the lenses. For the range of incident pulse energy (between 7.5 mJ and 60 mJ) studied, the absorption is seen to increase, although with diminishing rates, as the incident laser energy increases for all of the tested focal lengths. The absorption is seen to be slightly lower for longer focal lengths at lower incident laser

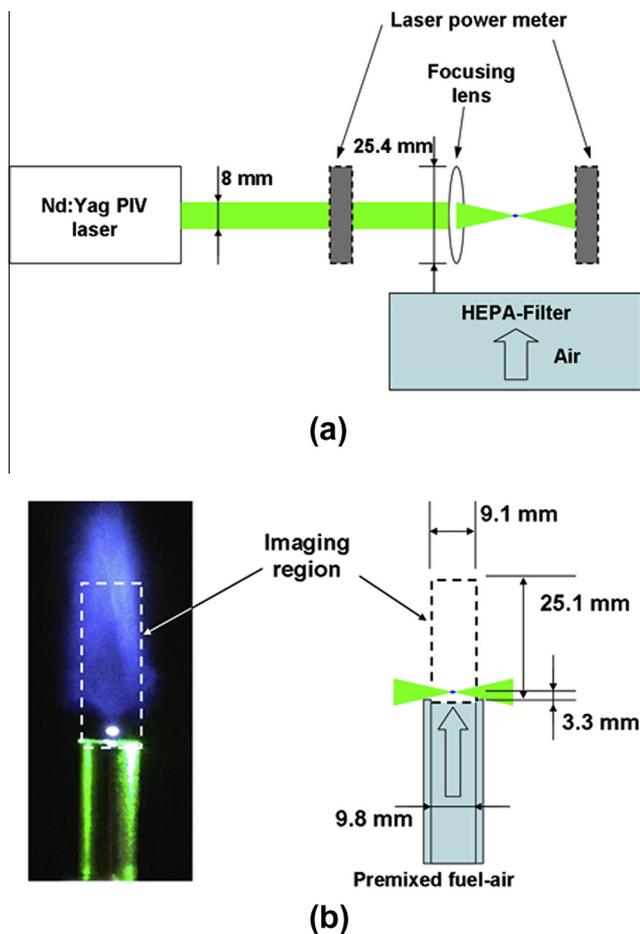


Fig. 1. Schematic of the experimental set-up to study laser-induced breakdown plasmas using single or double laser pulses (a) in atmospheric pressure air and (b) in a premixed ethane–air mixture.

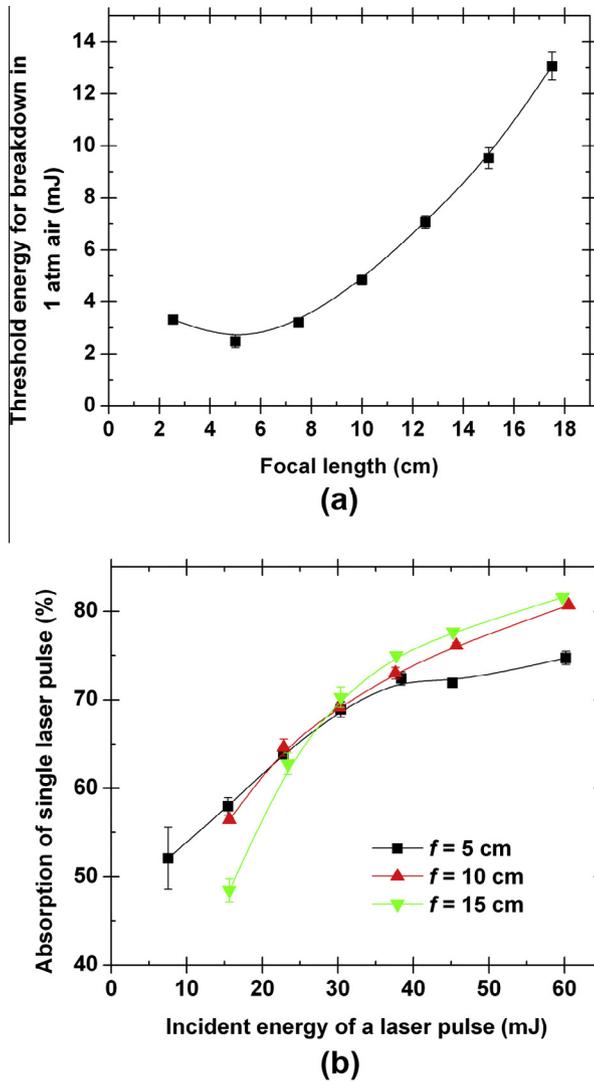


Fig. 2. (a) Focal length dependency of the threshold energy required to produce a breakdown plasma, and (b) incident laser energy dependence of the absorption for focal lengths of 5 cm, 10 cm, and 15 cm.

pulse energy (less than 30 mJ) and slightly higher for the longer focal lengths when the laser pulse energy is greater than 30 mJ.

3.2. Laser-induced plasma generated by two successive laser pulses in air

The laser power of two successive focused laser pulses is measured to determine the coupling of the second laser pulse to the breakdown plasma that is produced by the first pulse. The amount of laser energy absorbed by the second pulse for incident laser energies of 15 mJ, 22.5 mJ, and 30 mJ and as a function of the time interval between pulses (between 10 ns and 1 ms) is shown in Fig. 3a. Here, the focal length of the focusing lens is 5 cm. We see that when the time between pulses is within a several tens of nanoseconds, the absorption of the second pulse is enhanced slightly. This is expected since the plasma generated by the first pulse has not yet recombined before the arrival of the second pulse and provides a medium for increased energy absorption from the entire second pulse (through inverse Bremsstrahlung energy coupling to the plasma electrons). However, when the time between the first and second pulse is between 150 ns and 30 μ s, the coupling of laser energy to the medium diminishes considerably and

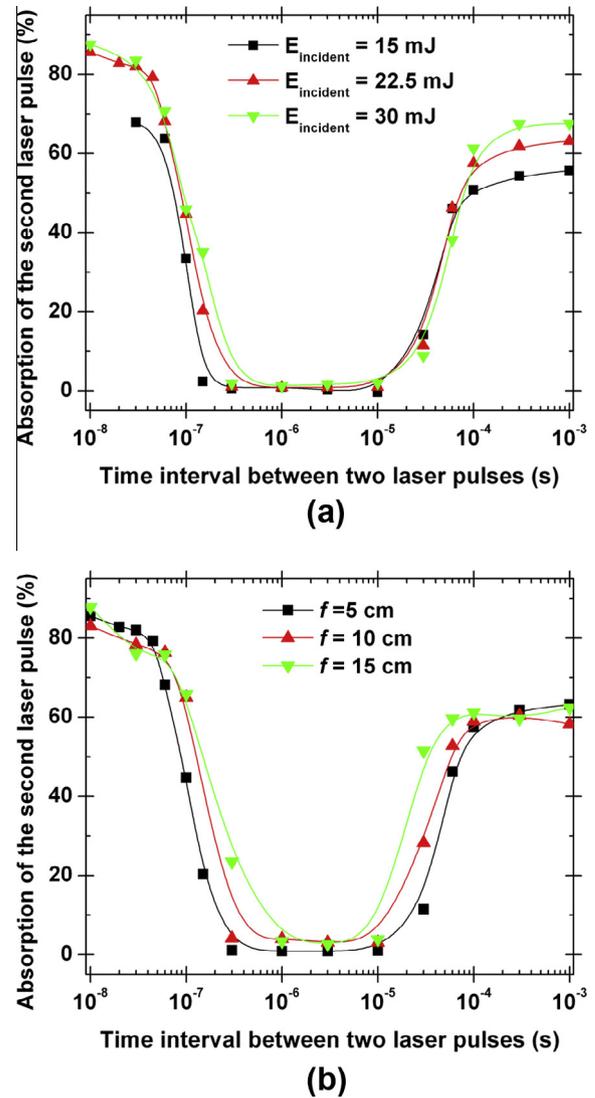


Fig. 3. The dependency of the absorption of the second laser pulse on time interval between laser pulses (a) for incident energies of 15 mJ, 22.5 mJ, and 30 mJ (with 5 cm focal length lens), and (b) for different lens focal lengths of 5 cm, 10 cm, and 15 cm (with 22.5 mJ laser incident energy).

the absorption is near zero. We attribute this to the gas heating and expansion that takes place on time scales associated with the speed of sound, long after the plasma has recombined. If the temperature of the post-plasma region is about 3000 K, the plasma will expand at a rate of about 1 mm/ μ s, resulting in a significant drop in total density. Others have reported that a strong pressure wave is formed at the breakdown kernel [14,16] and that the threshold energy to produce the breakdown in air increases as the gas density decreases [17], consistent with our findings. We see that when the time interval between the pulses is increased to levels beyond 30 μ s, the absorption of the second pulse recovers its single pulse value. This increased time interval is enough for the gas to cool and to return to its undisturbed state, or to be displaced by upstream air as a result of the modest imposed flow. Some (small) difference is seen in the recovery for different incident laser energies. These findings suggest that there exists an upper limit for laser repetition rates resulting in efficient laser coupling into air. Under the conditions of our experiments, the interval between pulses should be greater than 100 μ s (corresponding to a 10 kHz repetition rate) to avoid this decreased absorption. We note that this result is based on the study of absorption by two successive

laser pulses. Repetitive laser pulses will require a slightly increased pulse interval compared to that of two successive laser pulses as the repetitive breakdowns release more heat and lower the quasi-steady state gas density.

Experiments with successive laser pulses were carried out with different focal length lenses while keeping the incident laser energy at 22.5 mJ. The laser energy absorbed by the second laser pulse and its variation with the time interval between the pulses for lens focal lengths of 5 cm, 10 cm, and 15 cm is shown in Fig. 3b. We see similar results to that of Fig. 3a, with a near zero absorption of the second pulse for time intervals between 1 μ s and 10 μ s. The decrease and recovery of the second pulse absorption is found to occur later and earlier in time when the focal length is increased. We believe that this is due to the increased beam waist (and hence increased generated plasma volume), resulting in an overall cooler plasma for constant total deposited energy. We expect the volume of the produced breakdown plasma to be proportional to the fourth power of the focal length (for Gaussian optics) [15]. When using a 15 cm focal length lens, the absorption of the second laser pulse is seen to recover to 80% of its single pulse value at 30 μ s. In general, we find that the laser pulse repetition rate can be increased slightly while still maintaining a high subsequent pulse absorption with the use of a longer focal length lens.

3.3. Laser-induced plasma generated with a single laser pulse in premixed ethane–air

The time evolution (propagation) of a premixed ethane–air flame at $\phi = 1$ that is ignited by the breakdown plasma produced by a single laser pulse is shown in Fig. 4. The base flow speed is 4.9 m/s. In the experiment, the lens focal length is 10 cm and the

incident laser energy is 22.5 mJ. Images are taken at times ranging from 0.05 ms to 5.8 ms after the breakdown laser pulse. Following the initial breakdown process, the flame kernel is seen to expand. The downstream flame front propagates upwards at speed of about 8 m/s. In the absence of a successive pulse, the flame lifts away from the discharge kernel due to blow-off and the flame base is then seen to drift upwards at speed about 3 m/s. Relative to the base flow, this flame base therefore appears to propagate at a speed of about 1.9 m/s. It is noteworthy that this is higher than the laminar flame speed and we attribute this to the thermal expansion as a result of the heat release due to combustion, and the curvature of the flame front.

The time-resolved images of a premixed ethane–air flame ignited by a single laser breakdown pulse for varying the fuel–air equivalence ratios at a fixed base speed of 4.9 m/s are shown in Fig. 5. All cases shown generate a flame kernel, however, the flame kernel is found to be quenched when ϕ is either less than 0.5 or greater than 2.0. These limits are very close to the known fuel lean and fuel rich flammability limits. When ϕ is around 1, the combustion initiated by the breakdown is self-sustained and has a characteristic expanding flame front. As expected, the extent of the expansion is greatest when $\phi = 1$.

3.4. Laser-induced plasma with two successive laser pulses in premixed ethane–air

The energy that is absorbed by the second laser pulse following the production of plasma by the first pulse in an ethane–air mixture at $\phi = 1$ is compared to that for pure air in Fig. 6. For the comparison to the single pulse results, the lens focal length, the incident laser energy, and the base flow speed are the same as those used in the single pulse experiment, i.e., 10 cm, 22.5 mJ,

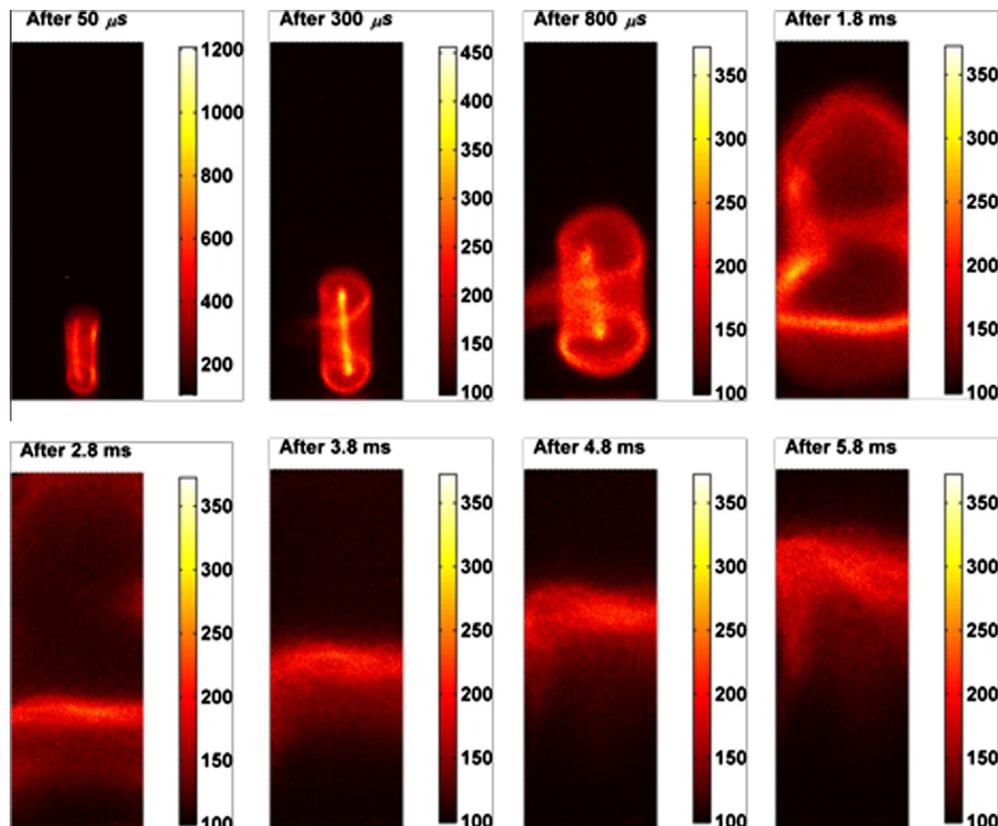


Fig. 4. Time evolution of the premixed ethane–air flame at $\phi = 1$ ignited by the breakdown plasma produced by a single laser pulse. Images are taken at times ranging from 0.05 ms to 5.8 ms after the breakdown laser pulse.

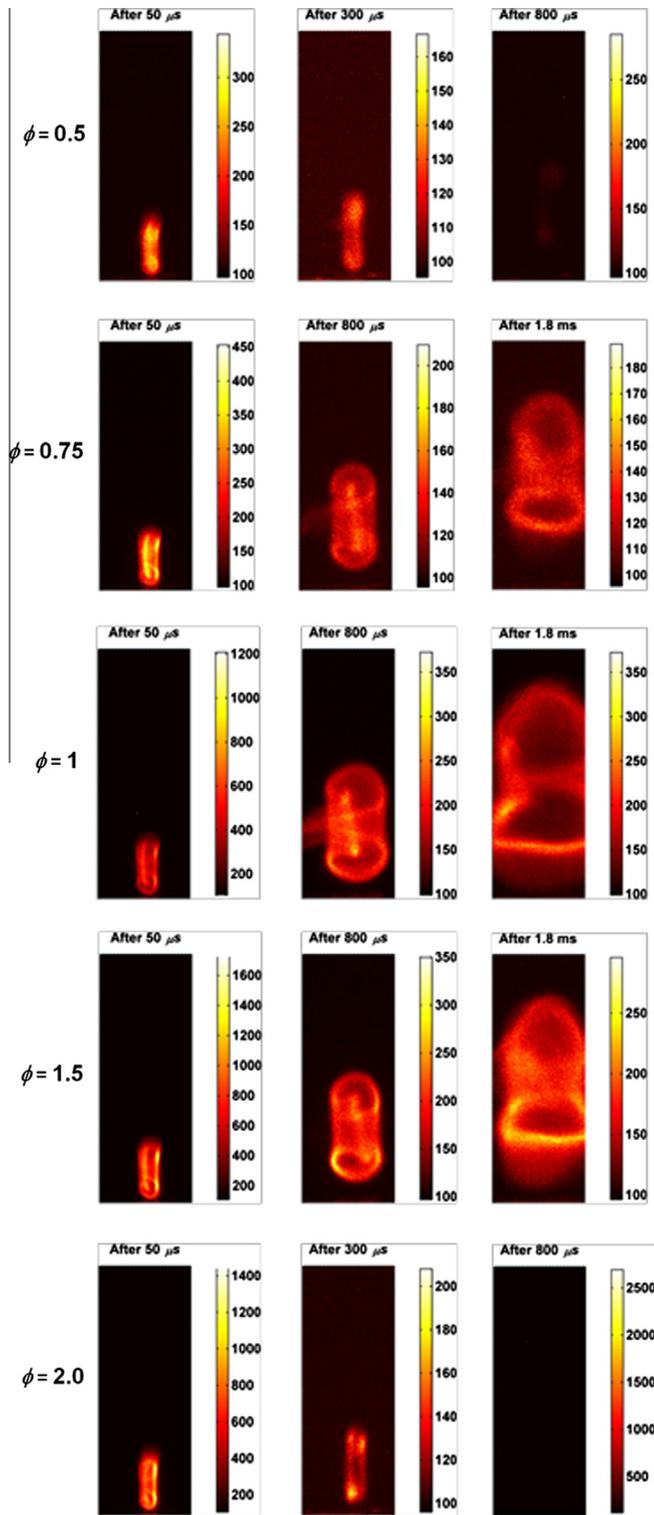


Fig. 5. Time evolution of the premixed ethane-air flame ignited by a single laser pulse breakdown for varying ϕ between 0.5 to 2.0. Images are taken at times ranging from 0.05 ms to 1.8 ms after the breakdown laser pulse.

and 4.9 m/s, respectively. For time intervals between pulses less than 100 μ s, the absorption characteristics are similar. It seems that the presence of the fuel in the mixture plays a minor role in the absorption of the second pulse for these short time intervals. However, at later times, i.e., between 100 μ s and 2 ms, we see a second region in which the absorption of the second pulse

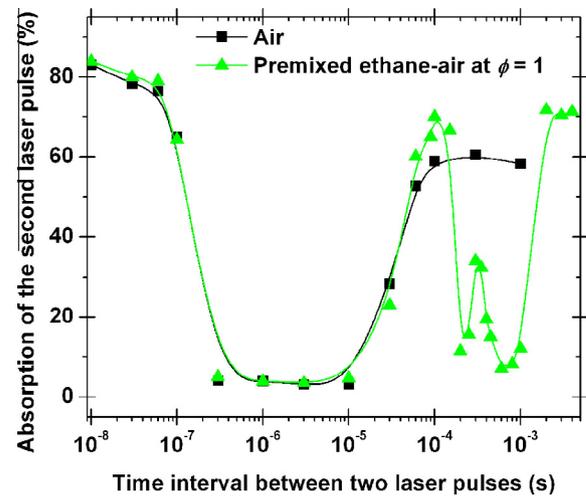


Fig. 6. Absorption of the second laser pulse energy into the plasma produced by the first pulse in atmospheric pressure air (flow at speed of 1.4 cm/s) and in a premixed ethane-air mixture at $\phi = 1$ (base flow at speed of 4.9 m/s). The lens focal length is 10 cm and the incident laser energy is 22.5 mJ.

experiences a dramatic decrease and subsequent recovery that is not seen in the case of pure air. This second region of diminished absorption is due to the combustion process. During the combustion of the fuel, the heat released once again raises the gas temperature and lowers the gas density, hence reducing the breakdown probability. As a result, these findings suggest that optimum coupling of laser energy to a combustible mixture for flame stabilization requires a lower laser repetition rate than that for pure air. For our tested conditions, the laser frequency should be less than 500 Hz.

The second laser pulse absorption is then measured while varying the equivalence ratio and the base flow speed. The results for varying the equivalence ratio from $\phi = 0.5$ to 2.0 while the base flow speed is fixed at 4.9 m/s are shown in Fig. 7. The recovery in the second laser pulse absorption occurs earlier in time as the equivalence ratio becomes either fuel-lean or fuel-rich. This is because the gas at the breakdown location is refreshed by the cool unburnt mixture. It can be seen from Fig. 5 that the flame drifts away from the nozzle rim at a slower rate when the ratio is close to 1. The results for varying the base flow speed between 3.3 m/s

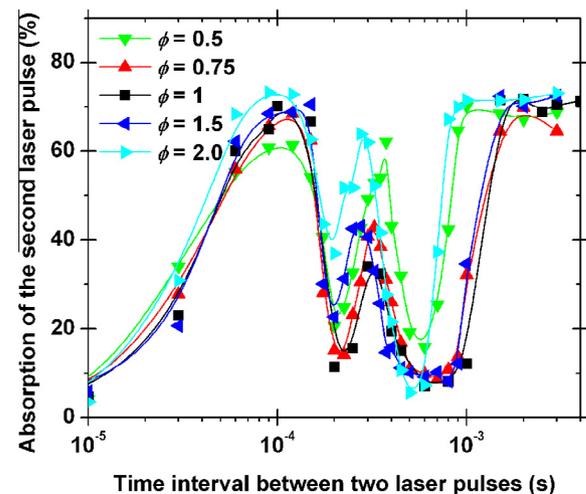


Fig. 7. Absorption of the second laser pulse by the plasma produced from the first pulse in premixed ethane-air mixture. The equivalence ratio is varied between $\phi = 0.5$ and 2.0. The base flow speed is fixed at 4.9 m/s.

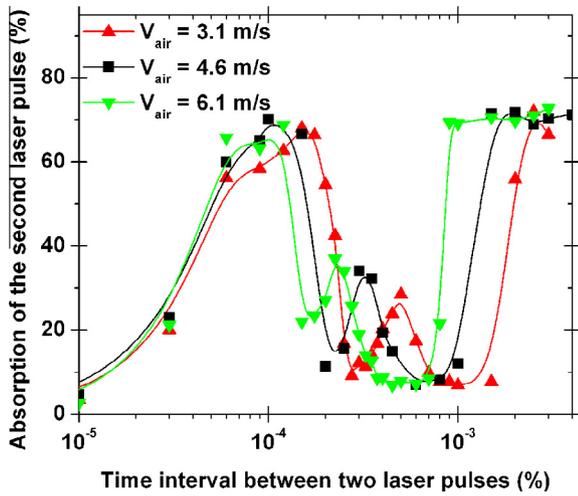


Fig. 8. Absorption of the second laser pulse by the plasma produced from the first pulse in premixed ethane–air mixtures ($\phi = 1$) while varying the base flow speed from 3.3 m/s to 6.5 m/s.

to 6.5 m/s at $\phi = 1$ are shown in Fig. 8. As expected, the recovery in the second laser pulse absorption occurs sooner as the base flow speed increases. This finding indicates that in repetitively ignited conditions, the laser repetition rate for optimum energy coupling

can be increased slightly under fuel-lean or -rich conditions and higher base flow speeds.

The time evolution of the premixed ethane–air flame at $\phi = 1$ ignited and stabilized by the breakdown plasma produced by the two successive laser pulses are shown in Fig. 9. The two different time intervals between laser pulses are tested. The first is 1 ms, which would correspond to a laser repetition rate of 1 kHz (see Fig. 9a), and the other is 3 ms, corresponding to a laser repetition rate of 333 Hz (see Fig. 9b). The time marker denoted in each image is the delay of the image following the firing of the second laser pulse. We see that for 1 ms time interval between the pulses (Fig. 9a), the second pulse does not produce a concentrated flame kernel and the images look very much like the sequence in Fig. 4, which are generated by a single laser pulse (1 ms must be added to the time marker in Fig. 9a to compare directly to images in Fig. 4, where the time delays are relative to the first laser pulse). The structures seen in Fig. 9a are due entirely to the first laser pulse as the low gas density generated by the expansion of the combusted gas from the first laser pulse precludes any absorption of the second pulse. We see from the third frame in Fig. 9a that it takes at least 2 ms for the flame to be swept some distance away from the region closed to the initial breakdown kernel and to replenish the region with a cooler, higher density unburnt charge. Under these conditions then a repetitively pulsed laser at 1 kHz would result in only a half of breakdown events and hence inefficient energy coupling. When the time interval between laser

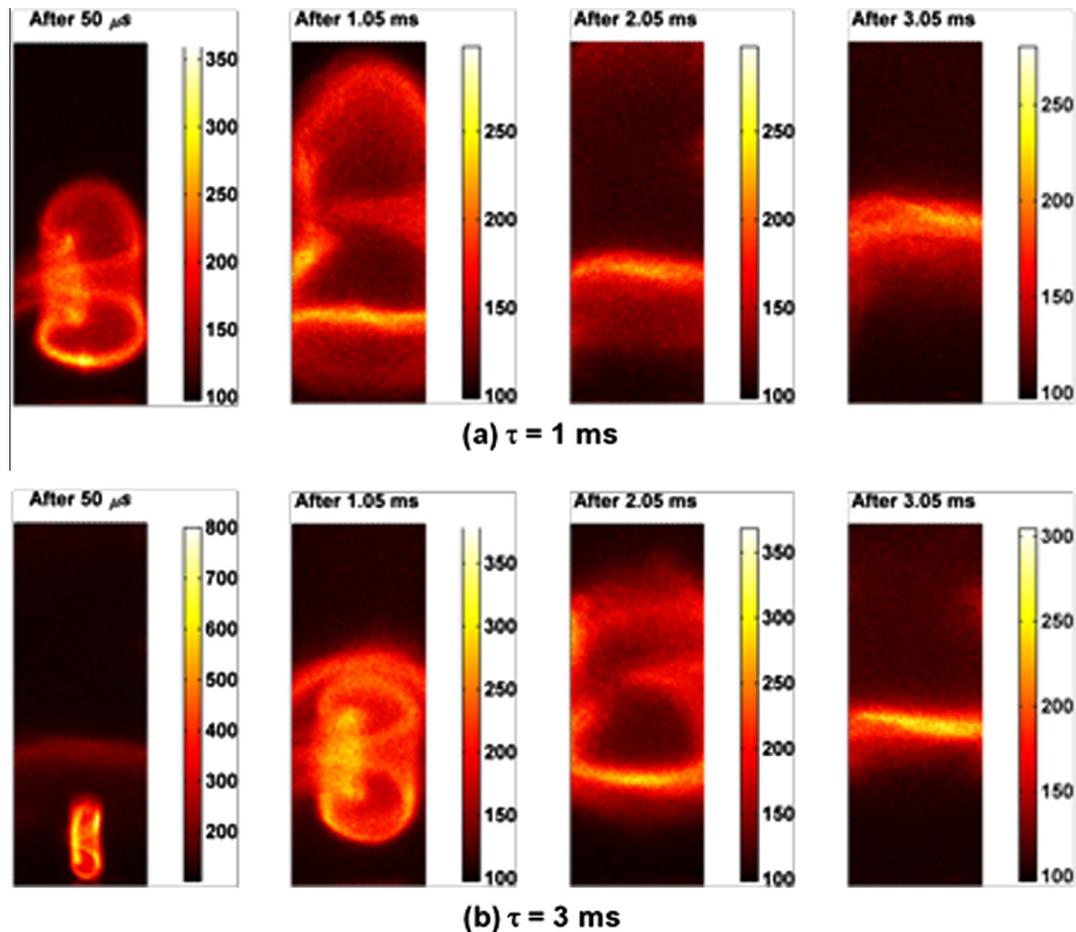


Fig. 9. Time evolution of the premixed ethane–air flame ($\phi = 1$) ignited by the breakdown plasma by two successive laser pulses separated by (a) 1 ms, and (b) 3 ms. The base flow speed is 4.9 m/s, and images are taken at 0.05, 1.05, 2.05, and 3.05 ms after the second laser pulse.

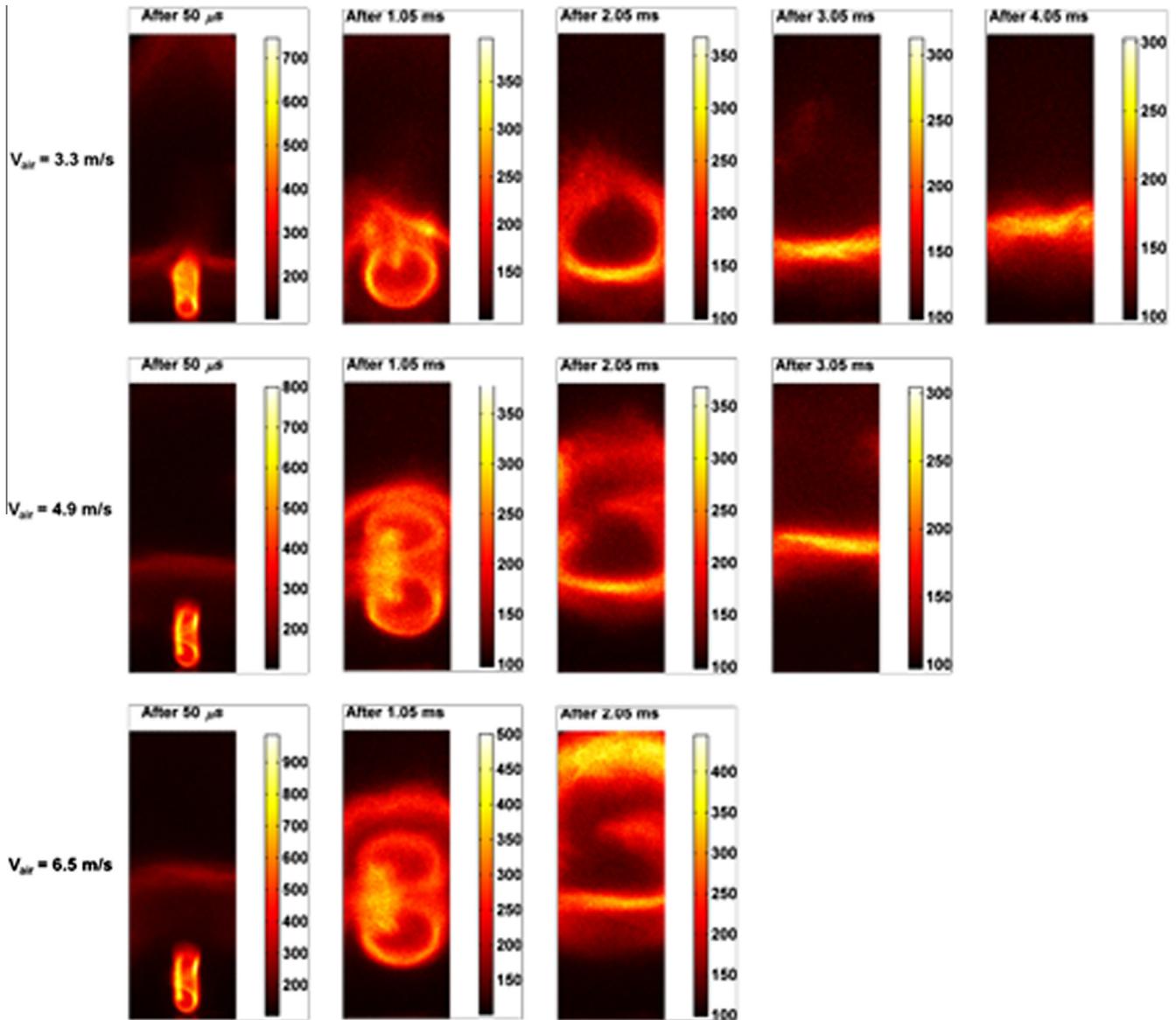


Fig. 10. Time evolution of the premixed ethane–air flames ($\phi = 1$) ignited by two successive laser pulse breakdowns separated by 3 ms while the base flow speed is varied from 3.3 m/s to 6.5 m/s. Images are taken at times ranging from 0.05 ms to 4.05 ms after the second laser pulse.

pulses is therefore increased to 3 ms (Fig. 9b), the second laser pulse now produces an intense flame kernel (first frame of Fig. 9b) that expands and propagates toward the remnant base of flame generated by the first pulse (second and third frames of Fig. 9b). These two flames appear to merge at about 3.05 ms, consuming the unburnt fuel/air between them. We expect that the use of a laser (of sufficient pulse energy for breakdown) with a repetition rate of 333 Hz – 500 Hz would lead to a quasi-continuous anchoring of the flame to the laser generated plasma.

The time-evolution of the premixed ethane–air flames ignited by the successive laser pulse breakdowns are recorded while varying the base flow speed from 3.3 m/s to 6.5 m/s at a fixed pulse interval of 3 ms. These results are shown in Fig. 10. For all of the base flow speeds, the flame produced by the second laser pulse catches up to the flame base that is produced by the first laser pulse since the downward flame front propagates faster than the flame base. The location where the flames merge shifts downstream for the higher base flow speeds. As these flame fronts merge, combustion of the fuel–air mixture between kernels becomes more complete. This happens even at laser repetition rates

slightly lower than optimum values, however, at the cost of increased flame base fluctuations.

4. Conclusion

Two successive focused laser pulses were employed to simulate experimentally, laser-induced breakdown plasmas at high laser repetition frequency in air and in premixed ethane–air mixtures. In both air and premixed ethane–air, a slight enhancement in absorption was achieved when the time interval between the pulses is within tens of nanoseconds whereas almost no absorption of the second pulse is seen when the time interval is between hundreds of nanoseconds and tens of microseconds (corresponding to pulse repetition rates of 100 kHz). The reason for this transparency is the low gas density as a result of plasma expansion that persists for several tens of microseconds after the initial laser pulse. In the case of premixed ethane–air mixtures, a second temporal window appears where there is little or no absorption of the second laser pulse. This window of transparency is a consequence of flame

ignition, and the resulting low gas density that persists for milliseconds. The window is in the 0.1–2 ms range, depending on the equivalence ratio and the base flow speed. As a result, for conditions where the equivalence ratio is 1 and the base flow speed is 4.9 m/s, the use of a 1 kHz laser for flame stabilization would result in a situation in which only half of its laser pulses produce the breakdown events leading to absorption. It appears that for the conditions studied here, the laser repetition rate should be less than about 500 Hz to produce conditions that lead to a nearly quasi-continuous anchoring of the flame base.

Acknowledgments

This research is supported by a Grant from the National Science Foundation. M.S. Bak also acknowledges support from Stanford University through a Stanford Graduate Fellowship.

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