

# Successive laser ablation ignition of premixed methane/air mixtures

Moon Soo Bak<sup>1,2,\*</sup> and Mark A Cappelli<sup>2</sup>

<sup>1</sup>*School of Mechanical Engineering, Sungkyunkwan University, 2066 Seobu-ro, Jangan-gu, Suwon, Gyunggi-do 440-746 South Korea*

<sup>2</sup>*Mechanical Engineering Department, Stanford University, 452 Escondido Mall, Stanford, California 94305-3032 USA*

\*[moonsoo@skku.edu](mailto:moonsoo@skku.edu)

**Abstract:** Laser ablation has been used to study successive ignition in premixed methane/air mixtures under conditions in which the flow speed leads to flame blow-out. A range of laser pulse frequencies is experimentally mimicked by varying the time interval between two closely spaced laser pulses. Emission intensities from the laser ablation kernels are measured to qualitatively estimate laser energy coupling, and flame CH\* chemiluminescence is recorded in a time-resolved manner to capture the flame evolution and propagation. A comparison of the measurements is made between the two successive breakdown ignition events. It is found that the formation of the subsequent ablation kernel is almost independent of the previous one, however, for the successive breakdowns, the first breakdown and its ensuing combustion created temporal regions of no energy coupling as they heat the gas and lower the density. Flame imaging shows that the second ablation event successfully produces another flame kernel and is capable of holding the flame-base even at pulse intervals where the second laser pulse cannot form a breakdown. This study demonstrates that successive ablation ignition can allow for the use of higher laser frequencies and enhanced flame stabilization than successive breakdown ignition.

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**OCIS codes:** (140.3440) Laser-induced breakdown; (350.5400) Plasmas; (280.2470) Flames.

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## 1. Introduction

Non-equilibrium plasmas have been found to be effective in stabilizing combustion at fuel-lean and blow-out conditions [1–5]. Among these non-equilibrium plasmas, nanosecond pulsed discharges have been used because of their lower required power budget in forming a high density plasma [6]. Previous studies demonstrated that repetitive pulsed discharges can reduce the ignition delay time [1–3] and hold the flame at its lean flammability limit [5]. Experiments have been carried out over a range of pressures and flow conditions, and the most likely reaction pathway has been identified to be the dissociative quenching of excited state species that results in a significant production of reactive atomic oxygen (O) and hydroxyl (OH) radicals [3,5,7,8]. In despite of the success of these repetitive discharges in stabilizing combustion, the intrusion of electrodes and connecting wires has limited their placement within the flame, as they can perturb the flow field and ensuing combustion. Recent studies [9,10] have been carried out of combustion stabilization using a laser-induced breakdown at high laser pulse repetition frequencies. Yu et al. [9] showed that the repetitively produced laser breakdown enhances a flame speed over a range of fuel/air equivalence ratios, and the mechanism for stabilization is believed to be similar to that associated with the electrode-driven discharges.

Laser-induced plasmas have been used extensively in spectroscopic analyses [11,12]. When generated in conjunction with surface ablation, they are often used for sample elemental analysis. In application to combustion, the type of laser plasmas has been mostly breakdown plasmas and their use is for flame ignition rather than flame holding [12–16]. A few studies [16, 17] have used laser ablation plasmas as ignition sources, but to our knowledge, no study has been reported on the use of flame holding with repetitive laser ablation plasmas. Morsy et al. [16] used laser ablation for studies of ignition in a static cell, mainly to develop a method to produce multiple ignition kernels using a single pulse and to reduce the total combustion time. Li et al. [17] discovered that ablation generates plasma kernels at much lower incident laser pulse energy, i.e., lowers the threshold energy for breakdown. Therefore, although the intrusion of an ablating surface constrains the system, repetitive ablation for flame stabilization can be achieved at lower incident laser energies.

In this study, two successive laser pulses are used to produce successive ablation and ignition for potential applications related to flame holding. The base flow speed is set to be greater than the flame speed for flame blow-out, and the pulse interval is varied between a few nanoseconds and a few milliseconds to mimic conditions associated with a Q-switched laser of a broad range of laser frequencies. The coupling of the laser pulse energy into the plasma is estimated using plasma emission intensities, and the flame development following the ablation ignition is characterized using CH\* chemiluminescence. In previous work [10], we conducted a similar study on successive laser breakdowns in premixed ethane-air mixtures

(in the absence of an ablating surface). The study found that laser frequencies should be higher than a certain limit to avoid a decreased energy coupling of the laser pulses. Here, we reproduced these past studies and compare them to the new results of successive ablation ignition under the same laser and combustion conditions.

## 2. Experimental setup

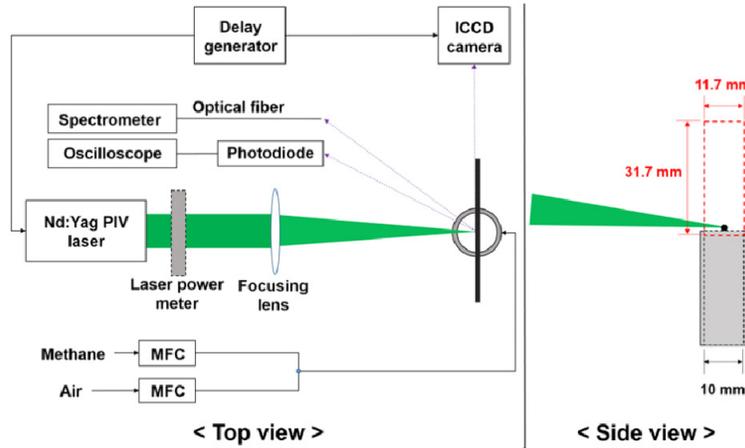


Fig. 1. Experimental setup for successive laser ablation ignition and breakdown ignition.

A schematic of the experimental setup for successive ignition by laser ablation or laser-induced breakdown in premixed methane/air mixtures is shown in Fig. 1. A Nd:Yag laser (New Wave Research, Gemini 200) that operates at 2 Hz generates a single or two successive laser pulses of frequency-doubled 532 nm. The laser pulse width is 8 ns, and the beam diameter is 8 mm. These laser pulses are focused either onto a surface to produce laser ablation or into a flow to induce breakdown using a plano-convex lens. The material for ablation is chosen to be a 2% thoriated tungsten carbide rod with a diameter of 1 mm because of its high thermal resistivity and small flow blockage area. The lens focal length is between 5 cm and 17.5 cm for single laser pulse experiments, and the pulse interval is varied from a few nanoseconds to a few milliseconds for successive laser pulse experiments.

In this study, a Bunsen-type burner is used for premixed combustion. Methane is premixed with air at an equivalence ratio of  $\phi = 1$  and this mixture exits a stainless steel nozzle with an inner diameter of 10 mm. Methane and air flow rates are controlled using mass flow controllers (Brooks 5850E series for methane and Unit Instruments UFC-3020A for air), and the total flow rate is fixed at  $360 \text{ cm}^3/\text{s}$ . The resulting exit flow speed of the nozzle is 4.6 m/s, and this speed is deliberately set to be greater than the laminar flame speed to cause flame blow-out. For the laser ablation ignition, the ablation (tungsten) rod is placed on top of the nozzle on flow centerline. The incoming laser beam is slightly angled at about  $10^\circ$  to prevent the nozzle from blocking any portion of the beam. During the experiment, the rod is moved back and forth at a speed of 0.25 cm/s using a translational stage equipped with a stepping motor (Superior Electric SLO-SYN® Motor) to expose a different portion of the surface following each laser pulse. Without this, a significant degradation in plasma strength is observed as a result of accumulative ablation in one place. For the breakdown ignition, the breakdown is formed 3.0 mm above from the exit of the nozzle; otherwise the laser pulse ablated inner wall of the nozzle before it induces breakdown. The incident energies of the laser pulses are measured and adjusted with the aid of a power meter (Ophir Optronics).

Two different measurements are carried out in this study. The first is a characterization of the energy coupling and the other is flame imaging by  $\text{CH}^*$  chemiluminescence. For the energy coupling measurement, the emission intensities of the plasmas are measured by a fast

time response photodiode (Thorlabs, DET210) whose signal is recorded with a high resolution oscilloscope (Tektronix TDS7104 1GHz). Since strong reflection of laser light can damage the photodiode cell, a short band-pass filter with a cutoff wavelength of 500 nm (Thorlabs, FES0500) is used in the optical train. The emission spectra of these plasmas are also characterized using a compact spectrometer (Ocean optics S2000 with a resolution of approximately 1 nm) for wavelengths between 200 nm and 1050 nm. For flame imaging, an intensified charge-coupled device (ICCD) camera capable of a relatively short gate width (Princeton PI-MAX) is used to record the time evolution of the flame ignited by single or successive laser plasmas. Predominantly CH\* chemiluminescence is detected as a result of the use of a 10 nm windowed band-pass filter that is centered at 430 nm (Edmunds Optics). The imaging field of view is 31.7 mm in height and 11.7 mm in width, and its lower boundary is located 1.85 mm below the nozzle exit. The camera trigger is synchronized to the laser pulse using a pulse delay generator (Stanford Research Systems, DG535), and the camera gate width is set to either 100  $\mu$ s or 300  $\mu$ s. A 100  $\mu$ s gate width is used when the time delay is less than 500  $\mu$ s after the plasma formation. One hundred images of each condition are acquired, and the images presented in this paper are averaged over this data set.

### 3. Results

#### 3.1 Threshold energies for laser ablation, breakdown, and flame ignition

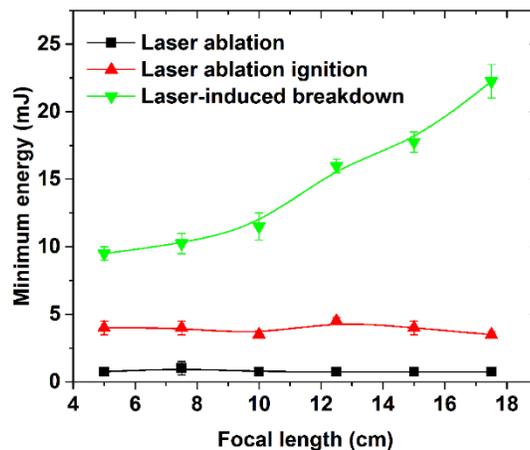


Fig. 2. Minimum energies to produce laser ablation, breakdown, and flame ignition in premixed methane/air at  $\phi = 1$  with a single laser pulse.

The minimum energies needed to produce laser ablation, breakdown, and flame ignition are measured in premixed methane/air with  $\phi = 1$  are shown in Fig. 2. A single laser pulse is used for these measurements, and the lens focal length is varied between 5 cm and 17.5 cm. We see that the threshold energy for the production of a laser ablation plasma is found to be less than 1 mJ, and that for the flame ignition by the ablation is about 4 mJ. It is noteworthy that despite the relatively low energy required for the formation of an ablation plasma, an energy below approximately 4 mJ is not enough to ignite the flame. The threshold energy required for laser ablation is independent of the lens focal length over the range studied. In contrast, the energy required for breakdown (in the absence of an ablation surface) is found to be much higher than that for ablation. This threshold energy is strongly dependent on the lens focal length, and the lowest required energy of 9.5 mJ is measured using the shortest focal length of 5 cm. These results are similar to those of previous studies [17] although the minimum threshold is different from theirs as the laser energy absorption into the plasma is sensitive to optical alignment as well as laser beam divergence. Laser breakdown always resulted in flame

ignition because of its high energy threshold. It is noteworthy that the minimum ignition energy of 4 mJ that is found for laser ablation is still higher than the value (0.4 mJ) obtained from standardized tests of electrical spark ignition [18]. This finding suggests that a significant portion of the energy is channeled into the ablation process itself, and the heating of the surface with a lesser amount transferred to the background gas for the initiation of combustion.

### 3.2 Energy coupling of successive laser pulses into ablation and breakdown

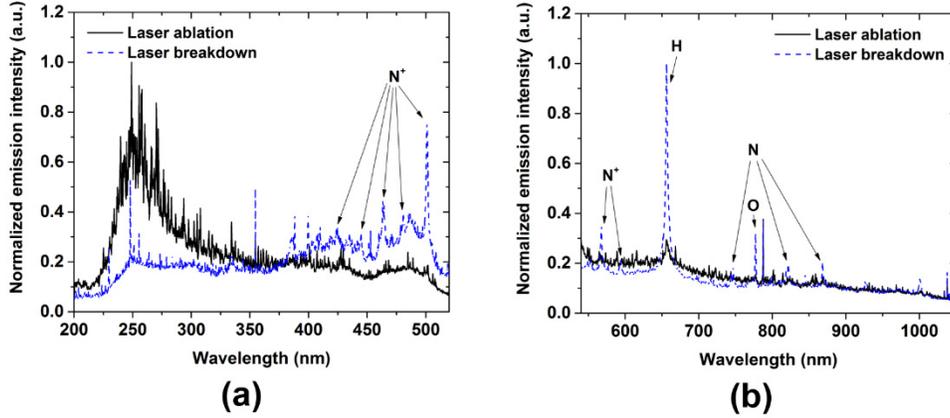


Fig. 3. (a) Optical emission spectra of laser ablation and breakdown in premixed methane/air at  $\phi = 1$  for the wavelength range from 200 nm to 520 nm and (b) the range from 540 nm to 1050 nm.

Two successive laser pulses are used here for the successive formation of a plasma and ignition. The incident laser energies per pulse are set to be  $7 (\pm 0.25)$  mJ and  $20.5 (\pm 0.25)$  mJ for the laser ablation and breakdown, respectively, while the lens focal length is 10 cm. In this measurement, the plasma emission intensity that the photodiode detects is used as a qualitative indicator of how much laser energy is coupled into the plasma. The photodiode collects the emission over a wide spectral region, so, for reference, the emission spectra (time-

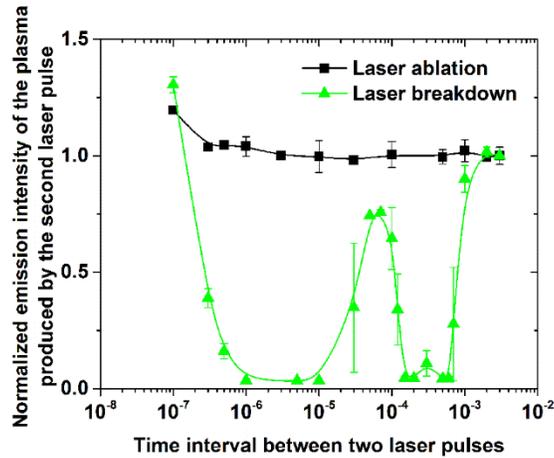


Fig. 4. Emission intensities of the second laser ablation and breakdown that are normalized to the intensities of the first laser ablation and breakdown in premixed methane/air at  $\phi = 1$ .

integrated for 500 ms) are also recorded. These emission spectra are shown in Fig. 3. Spectral lines of atomic N, O, and H are found to be strong for breakdown plasmas. In contrast, the spectrum for ablation plasmas is more like the broadband continuum emission. This may indicate that for ablation plasma, the laser energy couples mostly to the ablation surface rather than exciting the gas in front of the surface.

Measurements of the photodiode-recorded plasma emission with the successive laser pulses are shown in Fig. 4. The symbol/lines represent the emission intensities that the second laser pulse induces normalized by that produced from the first laser pulse. The results indicate that the second laser pulse produces ablation for all pulse intervals and the level of energy coupling is not degraded when compared to that of the first ablation pulse. However, for the case of laser breakdown, the second laser pulse becomes transparent to the gas when the pulse interval is between 300 ns and 30  $\mu$ s or between 120  $\mu$ s and 700  $\mu$ s. Our previous study [10] revealed that this is related to the decreased gas density that requires more energy for the induction of breakdown. The first temporal region of transparency is attributed to the gas heating induced by the first breakdown, and the second region of transparency is a consequence of gas heating following combustion. Comparing the ablation and breakdown results, the unaffected energy coupling of the second ablation suggests that the laser energy is first coupled to the ablation surface, and this coupling is independent of the presence of the prior pulse. The energy is subsequently transferred to the gas during the ablation process. It is also noteworthy that the emission intensities induced by the second laser pulse are slightly enhanced when the time interval is less than 100 ns.

### 3.3 CH\* chemiluminescence imaging of flames ignited by a single laser pulse

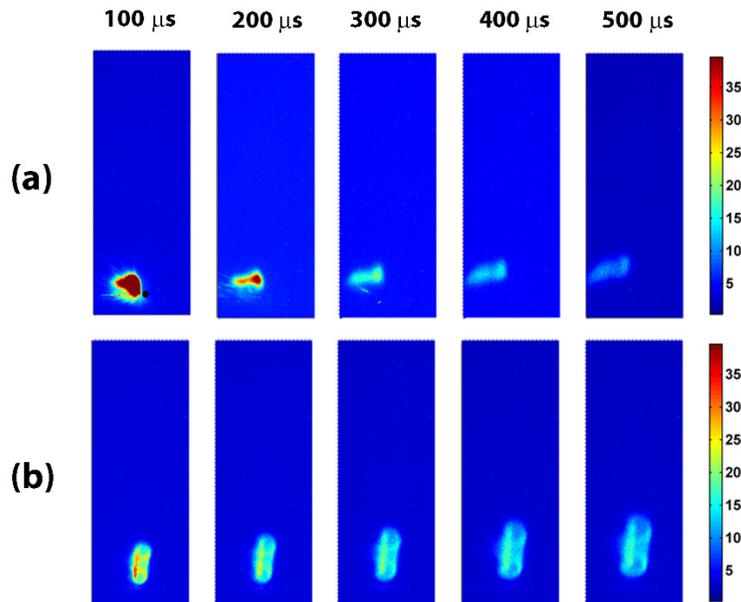


Fig. 5. CH\* chemiluminescence of flames ignited (a) by a single pulse laser ablation and (b) by a single pulse laser breakdown.

CH\* chemiluminescence imaging of the flame is carried out to understand how the resulting flame develops following laser ablation or laser breakdown. Images of the flames ignited by the single laser ablation and breakdown are shown in Fig. 5(a) and 5(b), respectively for the times between 100  $\mu$ s and 500  $\mu$ s following the laser pulse arrival. Since the variation in chemiluminescence intensity is severe, the intensity (arbitrary units) greater than 40 is let to

be saturated for the comparison among the flames measured at different times. Within  $100\ \mu\text{s}$  following the laser ablation (Fig. 5(a)) or laser breakdown (Fig. 5(b)), both plasmas lead to the formation of a flame kernel, albeit of different shapes. The flame kernel produced by the laser ablation propagates against the direction of the incoming laser beam in part due to the presence of the surface. In contrast, the flame kernel produced by the breakdown undergoes an almost a uniform expansion while its center slowly drifts downstream. The flame-base is found to be at similar (vertical) locations between the two cases, but the directed flame propagation of the ablation ignition leaves the plasma region replaced with the unburnt mixture much earlier in time. This difference in flame evolution turns out to be responsible for the decreased energy coupling of the subsequent laser pulse especially when the pulse interval is not long enough (e.g., less than 1 ms under these conditions). At these times, the downward flame speeds are measured to be about 5.1 m/s.

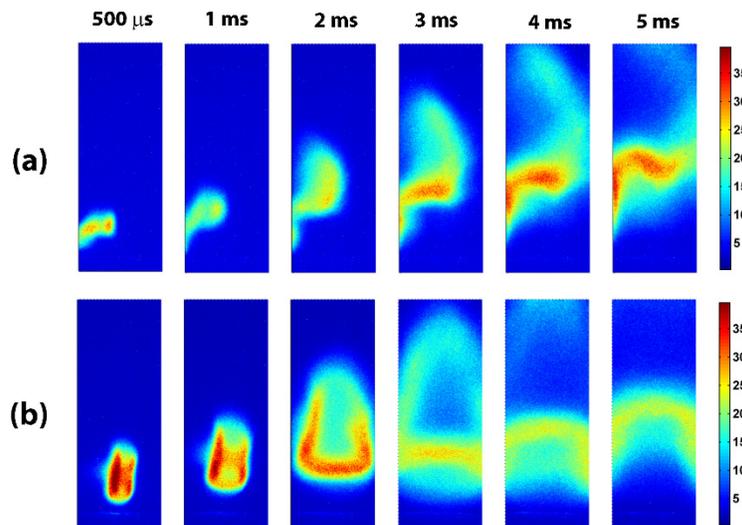


Fig. 6.  $\text{CH}^*$  chemiluminescence of flame propagation ignited (a) by a single pulse laser ablation and (b) by a single pulse laser breakdown.

The flame structures that propagate long after the ablation and breakdown ignitions are shown for times between 1 ms and 5 ms in Fig. 6. For the ablation ignition (Fig. 6(a)), the flame propagates away from the direction of the initial flame front (e.g., to the right in these images) while the flame-base is lifted by the base flow. The flame propagation of the breakdown ignition is also similar to that of the ablation ignition except that the flame-front is smoother and the flame-base locates slightly further downstream. The differences are mostly a result of the intrusion of the ablation rod, as the rod creates a small region of flow recirculation in its wake. The flame speeds are estimated to be around 2.91 m/s and 1.77 m/s based on the flame-base location and the base flow speed. This range of flame speed is higher than the laminar flame speed and this is attributed to the flame curvature.

### 3.4 $\text{CH}^*$ chemiluminescence imaging of flames produced by successive laser pulses

We find from Fig. 4 that the laser ablation exhibits almost no degradation in energy coupling between the first and subsequent laser pulse, unlike the behavior seen for the breakdown plasma. However, it remains to be confirmed that the deposited energy by the second laser pulse eventually also results in flame ignition. In this section, flame images are recorded at a time of  $100\ \mu\text{s}$  after the successive laser pulses while varying the interval between pulses from  $500\ \mu\text{s}$  to 3 ms. The images for the cases of ablation ignition and breakdown ignition are compared in Fig. 7(a) and 7(b), respectively. In Fig. 7(a), we see that the second ablation

successively produces a second flame kernel for any of the tested pulse intervals. The only difference between images is that the spatial separation between the flame-fronts produced by the two laser ablations is larger for the longer time intervals. However, for the case of laser breakdown (see Fig. 7(b)), the second laser pulse does not produce a second flame kernel when the pulse interval is less than 1 ms (as also expected from Fig. 4). The flame kernel from the first breakdown event appears to overlap the region of the subsequent laser pulse focus. As the pulse interval increases, the flame base drifts downstream by a sufficient amount and a fresh unburnt (and presumably colder) mixture replaces the hotter region generated by the first laser pulse. Although the 1 ms pulse interval allows to produce a second relatively weak breakdown and flame kernel, a complete separation between the upper and lower flame-fronts is achieved when the pulse interval is greater than 2 ms under the tested conditions. As a result, for these flow conditions, a maximum laser frequency (about 500 Hz) exists for optimum energy coupling when repetitive breakdown ignition is used. It is noteworthy that laser ablation ignition does not have that constraint.

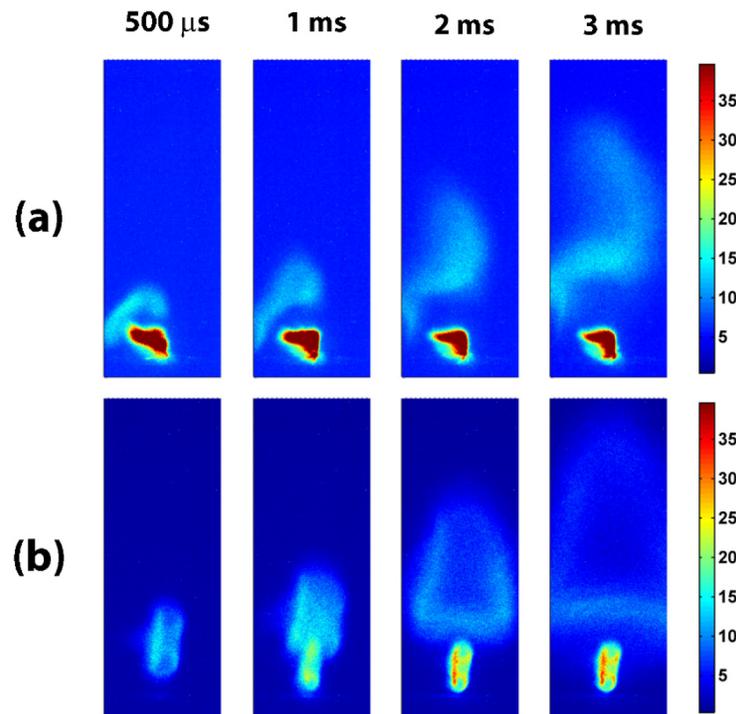


Fig. 7. CH\* chemiluminescence of flames stabilized (a) by successive laser ablations and (b) by successive laser breakdowns.

A merging of the upper and lower flame-fronts produced by two successive laser pulses is studied for a pulse interval of 3 ms. The results of successive laser ablation ignition and breakdown ignition are shown in Fig. 8(a) and 8(b), respectively for times of 1 ms to 5 ms after the second laser pulse (e.g., the first images labeled 1 ms correspond to the images labeled 4 ms in Fig. 6). At a delay of 1 ms, the upper flame-fronts seem to be at a location slight further downstream as a result of the presence of the second flame kernels when compared to the locations of these flame fronts for single laser pulse ignitions. At this time delay, the flame fronts are still spatially separated. By 2 ms, these flame fronts merge and the flame chemiluminescence becomes more intense as the unburnt mixture (between the flames)

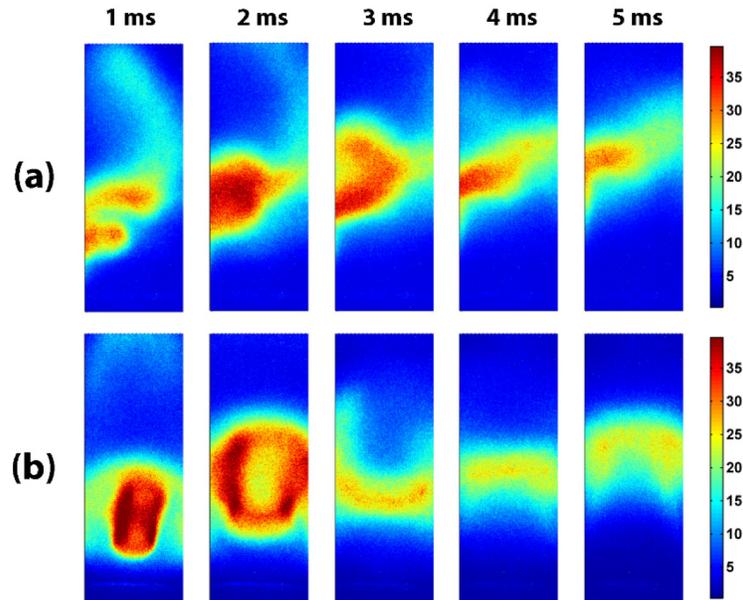


Fig. 8. CH\* chemiluminescence of flames ignited (a) by successive laser ablations and (a) by successive laser breakdowns.

also burns. The merging process appears to be complete after 3 ms, and the flame-bases locate close to where those of the single laser pulse ignitions were at these times. While this study has used only two successive laser pulses to study repetitive flame ignition dynamics, it is demonstrated that the flame-base can be held to a position very near that of where ignition occurs even at flow speeds higher than the flame speeds using a laser repetition frequency of 333 Hz (corresponding to 3 ms pulse interval). Furthermore, based on the findings that the laser ablation exploits most of the laser energy and has negligible dependence on the previous ablation, it is expected that the repetitive laser ablation will allow the use of higher laser frequencies for enhanced flame stabilization.

#### 4. Conclusion

Successive laser pulses were employed to produce flame ignition by laser ablation and breakdown in a premixed methane/air mixture. Single pulse experiments confirmed that the ablation requires a lower threshold for plasma formation and ignition compared to that of breakdown in the absence of an ablation surface. With the use of two successive laser pulses, the energy coupling for ablation is almost independent of the presence of a previous ablation pulse whereas with laser breakdown plasmas, the first pulse and its ensuing combustion prevents the second laser pulse from inducing a breakdown. Flame imaging of CH\* chemiluminescence indicates that the energy coupled to the second laser ablation event successfully produces a second flame kernel that enables flame holding even at pulse intervals where the second laser pulse cannot form a breakdown in the absence of an ablation surface. The flame-fronts produced by the successive ignitions were also shown to merge at downstream locations not too far from the ignition kernels. The study suggests that successive laser ablation can allow the use of higher laser frequencies and enhanced flame stabilization than successive laser breakdown at comparable laser pulse energy.