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Short Communication

Unfocused laser ignition of high-pressure He–H_2-O_2 combustible mixtures

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ABSTRACT

We report consistent ignition of high-pressure (p_{fill} > 20–30 bar) hydrogen-oxygen mixtures diluted with helium in two different combustion vessels, using an unfocused Nd:YAG laser. This corresponds to laser irradiances several orders of magnitude below the minimum ignition energies reported in the literature. This unusual phenomena has led us to try to measure the amount of laser radiation absorbed by the gas medium. By placing a mirror inside a cylindrical vessel and filling it up to 100 bar with He-H_2 or He-O_2 non-combustible mixtures, we obtain the pressure-dependent absorptivity of the combustible He-H_2-O_2 mixture. We find no measurable absorption of the laser signal by the medium, for the overall pressure range, to the experimental apparatus sensitivity (about 1% of the laser irradiance). The exact mechanism for ignition remains henceforth unknown. One possibility could the creation of seed electrons created by autofocusing ionization of dust/impurities in the gas, but this has yet to be experimentally confirmed.

1. Introduction

Laser-induced spark ignition has experienced increasing interest in recent years due to its advantages over conventional spark-plug ignition. It is specially effective in high pressure regimes where conventional spark plugs have low life time [1,2]. Ignition of a flammable mixture occurs once a critical amount of energy in the form of heat and/or radicals is deposited in a specific location. If a subcritical amount of energy is deposited, the initial flame kernel quickly decays as a result of transport processes that diffuse heat and radicals at a higher rate than the volumetric production rate from the kernel flame. Conversely, if a supercritical amount of energy is deposed, called the minimum ignition energy, the local temperature decays to the adiabatic flame temperature in an amount of time sufficient to ensure that temperature gradients in the kernel are shallow enough that energy diffusion processes may be lower than the source terms due to chemical reactions [3]. This means that a minimum amount of energy needs to be deposed over a given volume to ensure reliable ignition of mixtures, a concept that has been evidenced in numerical simulations carried out by Maas [4]. This energy gradient, balancing chemical production terms and loss terms due to diffusion, is critical to the ignition of such mixtures, with typical values for a stoichiometric H_2-air mixture around 0.02 mJ.

We report the successful ignition of He-H_2-O_2 mixtures resorting to an Nd:YAG laser without the usual focusing lens setup, which corresponds to irradiances about three orders of magnitude below the accepted minimum, if the gas filling pressure exceeds a given threshold (p_{fill} > 20−−30 bar). This phenomena was observed in two combustion vessels of different sizes (a 3 L prototype, and the final 50 L combustion chamber). In this communication, we present a sample of the shots produced in both vessels (by means of a piezoelectric pressure gauge), and a further investigation on the transmittance of the unfocused laser signal in such high-pressure gases where we examined two combination of inert gases (He-H_2 and He-O_2) to reproduce the conditions of the flammable gas, in an attempt to infer whether any measurable laser absorption process can be experimentally identified.

2. Fundamentals of laser ignition processes

In a typical setup a laser source outside of a closed vessel will be focused in a specific point inside through a focusing optics system. The average irradiance I of a laser with pulse energy E_{pulse}, pulse duration Δt_{pulse}, defined as the $FWHM$ (Full Width at Half Maximum of a

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Gaussian pulse), and beam diameter $2w$, is given by:

$$I = \frac{E_{pulse}}{\Delta t_{pulse}} \frac{1}{\pi w^2} . \quad (1)$$

Achieving ignition through a laser source implies an efficient energy transfer to the combustible mixture, starting the chemical reactions leading to a full scale combustion. From a fundamental stand point, this may be achieved through four different processes, which are: laser thermal ignition; laser-induced photochemical ignition; laser-induced resonant breakdown ignition; and laser-induced spark ignition.

It is also common to relate the amount of transmitted power through an optic medium through the Beer–Lambert law, Eq. (2). The ratio of transmitted power P_t to the incident power P_0 is the transmittance $\mathcal{T}$,

$$P_t/P_0 = \mathcal{T} = \exp(-\alpha_{abs} d). \quad (2)$$

For a medium whose optic length d and absorption coefficient α_{abs} are constant, the transmittance is also constant. If a gas medium becomes ionized then α_{abs} increases and the laser's power is easily absorbed.

2.1. Laser-induced spark ignition

Laser-induced spark ignition is probably the most useful method for practical applications since no particular laser wavelength is required. The process begins with multiphoton ionization (MPI) of a few gas molecules which release electrons that readily absorb the laser energy via the inverse *bremsstrahlung* process, increasing their kinetic energy. These accelerated electrons collide with other molecules and atoms, ionizing them and releasing more electrons, which then accelerate and repeat the process. This creates an electron cascade leading to a plasma formation which then starts chain-branching reactions and ignites the mixture.

Multiphoton ionization processes are in some cases essential for the initial stage of breakdown because photon energy is in the infrared band, while the ionization energy is in the ultraviolet band. The presence of impurities, as aerosol, dust or soot, for example, can significantly facilitate the generation of these initial electrons as they more easily absorb infrared radiation. When using ultrashort pulses (picosecond order) the multiphoton process must provide electrical breakdown all by itself, since there is insufficient time for electron–molecule collisions to occur and form the electron cascade process. While with nanosecond pulses there is time enough for electron–molecule collisions generate more electrons, through the electron cascade mechanism [5]. Loss processes such as electron diffusion, radiation, collisional quenching of excited states, among others, are present and may increase the effective ignition energy. A spark originated by these means is a source of highly reactive chemical intermediates at very high pressures and temperatures. This spark emits light, heat and a shock wave to the surrounding medium, resulting in an ignition kernel. A sufficiently strong kernel permits a transition into full scale combustion [5].

Syage et al. in [6] studied ignition of hydrogen-air and hydrogen-air-CO_2 mixtures using different pulses. The results show that the pulse duration, if below 15 ns, and the laser wavelength do not affect the ignition process nor the minimum ignition energy. Spiglanin et al. [7] investigated the shape and the structure of developing flame kernels as function of time and mixture composition in laser-induced spark ignition in hydrogen-air. As stated by the authors, gas motion dominates early flame kernel growth. However, the ignition success depends on the chemistry of the reactions, which determines if the gas transits from hot plasma to propagating flame or not. Phuoc and White in [8] studied the ignition probability dependence on the ignition location for a diffusion jet flame. An important conclusion that they state is that as long as the pulse energy is enough to guarantee the gas breakdown any higher energies will not affect the observed distribution of the ignition probability. Thus, the distribution of ignition probability is mostly likely attributed to local variations of air:fuel equivalence ratio within the jet. Turbulence intensity and velocity gradient at the ignition location may also influence ignition probability.

Table 1
Minimum breakdown or ignition irradiance reported in the literature.

I (W/cm^2)	Reference	Conditions/Model
1.93×10^{11}	Bradley, [15]	Air at 1 MPa
1×10^{10}	Phuoc, [5]	Theoretical
1×10^{12}	Phuoc, [12]	Air, CH_4, O_2, N_2, H_2 at 150 and 3040 Torr
1×10^{14}	Srivastava, [9]	Drude model
1×10^{12}	Weinrotter, [1]	H_2-air 2.8 MPa air:fuel ratio 3
1×10^{10}	Lee, [13]	Propane-air at 1 atm

2.2. Minimum laser intensity

For an electric breakdown to occur in a gaseous mixture, initial electrons are required to start an electron cascade. As aforementioned, these electrons may come from MPI, where a molecule absorbs laser photons and become ionized. A significant wavelength dependence is expected as this is a quantum effect. Srivastava in [9] states that for the first electron to be produced by MPI the irradiance should be of the order of 10^{14} W/cm^2. This high value is expected because the ionization energy is much larger than the energy of a single photon. Ionization energy of O_2 molecules is 12.07 eV, of whereas a typical Nd:YAG laser at 1064 nm has a photon energy of 1.16 eV. Thus, 10 or more photons are need to produce one free electron.

Experiments and studies done by many different authors report different values for breakdown intensity, yet always with orders of magnitude below 10^{14} W/cm^2. For example, Srivastava [9] reports the intensity in the focus to be of 10^{12} W/cm^2, while Phuoc in [5] even states 10^{11} W/cm^2 to be sufficient. Values reported by various authors and conditions for the minimum irradiance needed to achieve gas breakdown/ignition are summarized in Table 1.

Several authors, [1,2,9–11], defend that this seed electrons do not come from MPI but from impurities in gas mixture (e.g. dust, aerosol or soot particles). This hypothesis is supported by a non-dependence of the minimum pulse energy (MPE) with the laser wavelength, and, a strong pressure dependence reported in [2,8,11–13], whereas MPI predicts a very weak dependence. Ronney in [14] estimated the minimum ignition energy for developing a flame kernel, which is proportional to the gas density, meaning proportional to the gas pressure. On the other hand its thermal conductivity and specific heat limit this increase, leading to a very weak pressure dependence. The experimental data disagrees with this weak pressure dependence, with Kopecek et al. in [11] reporting a dependence of the form $I \sim p^{-1/m}$, $m \approx 1$.

2.3. Gas breakdown

As previously discussed, a laser with $I > 10^{11}$ W/cm^2 interacting with a gas may ionize it. Whenever ionization occurs, a light flash and sharp acoustic sound is observed, similar to that of an electric spark-plug discharge.[1] Seed electrons, generated by either MPI or ionization of impurities, soot, aerosol particles, organic vapors, dust, e.g., gain energy via inverse *bremsstrahlung* and ionize more molecules. The gas electrical breakdown occurs when a significantly fraction of the gas, 10^{-3}, is ionized, or when we have an electron density of 10^{16} electrons/cm^3 [16]. The cascade ionization process is more intense at higher pressures and for longer laser pulses, since electron–atom or electron–ion collisions have a characteristic time of μs. According to Morgan [16], the condition for gas breakdown is the product of gas pressure p and laser time pulse duration Δt_{pulse}, $p\Delta t_{pulse} > 10^{-7}$ torr s.

The defining parameter that ensures ignition is the laser's irradiance I, previously defined in Eq. (1). A high enough irradiance will cause an electric breakdown in the gas, that will led to a successful ignition [8].

[1] Laser-induced sparks are smaller, shorter in duration and produce larger temperature and density gradients.

Table 2
"Bombe" shots parameters for Fig. 2.

Shot nr.	p_{fill} (bar)	p_{peak} (bar)	ϕ	X_{He} (%)	Focus
B126	50	486	1.08	73	Yes
B127	50	536	1.23	72	Yes
B128	50	305	0.78	73	No
B135	50	310	0.69	71	No
B136	50	330	0.98	73	No

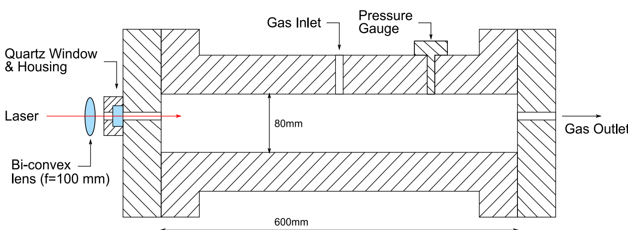

Fig. 1. Schematic cutaway of the "bombe" and laser setup.

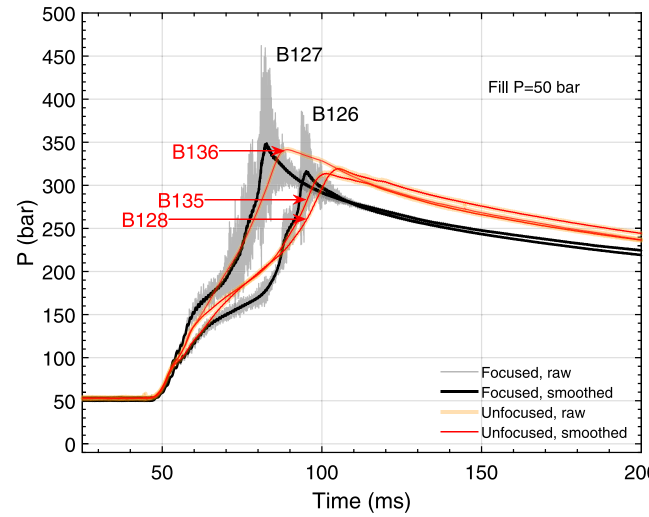

Fig. 2. Pressure rise *vs.* time of the small-scale combustion chamber ("bombe") ignited with both focused and non-focused laser beam.

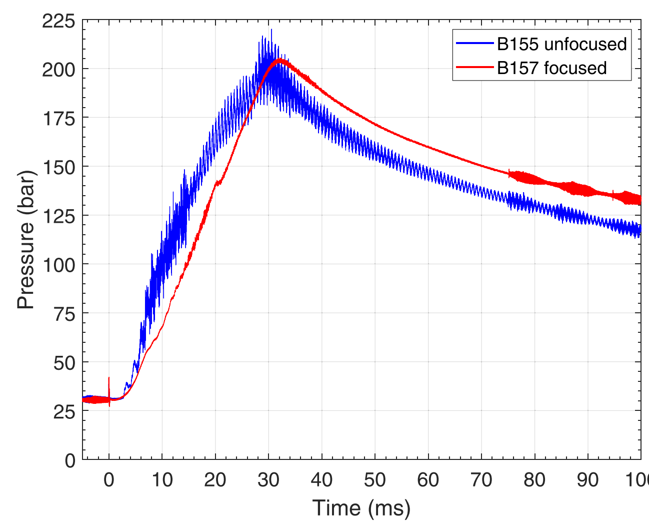

Fig. 3. Pressure signal comparison of B157 and B155, p_{fill} = 30 bar, ϕ = 1.18, X_{He} = 73%.

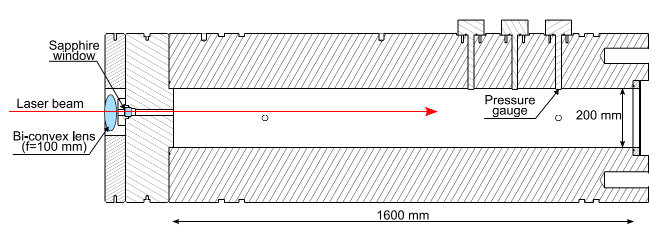

Fig. 4. Schematic cutaway of the ESTHER driver combustion chamber, the laser ignition system is the same as the "bombe".

3. Prior observations

The European Shock Tube for High Enthalpy Research (ESTHER) [17,18] is a facility capable of reaching shock-speeds in excess of 10 km/s. The facility comprises a 50 L combustion chamber driver where an hydrogen-oxygen mixture diluted in helium with filling pressures up to 100 bar is ignited to a post-combustion pressure up to 600 bar. A test-scale model (3 L) "bombe" was firstly built to demonstrate the full driver and to ensure the safety and validation of all subsystems. The initial configuration deployed a hotwire ignition system which was later advantageously replaced by a laser ignition system. The resulting setup is depicted in Fig. 1. During the test campaign it was found that for filling pressures above 20–30 bar the laser could ignite the gas mixture without the use of the lens, reducing the irradiance to around 10^8 W/cm^2. This value is two orders of magnitude below the reported minimums in the literature, which always consider a focusing lens in the setup.

Fig. 2 presents the recorded pressure signal in both focused and unfocused ignition mode. A series of shots (B126, B127, B128, B135, B136) were performed and measured with a filing pressure of 50 bar. The helium dilution is around 70 % in terms of molar fractions, and the mixtures fuel:air equivalence ratio (ϕ) range from rich to lean. Shots B126 and B127 were carried with a focusing lens, the others with the laser remaining unfocused. The properties for this series of shots are summarized in Table 2.

Fig. 3 compares the pressure signal of shots B157 and B155, whose only difference is the use or not of the focusing lens. The signal of B157 (focused beam) has lower acoustic oscillations than the B155 where the oscillations start at the beginning of the pressure rise.

This small-scale combustion chamber was de-comissioned and replace with the full-scale shock tube driver shown in Fig. 4. This is a 50 L cylindrical combustion chamber with 1600 mm of inner length and 200 mm of inner diameter. The optical window is now made of sapphire and the laser ignition system is the same as the one used in the "bombe". Because there is only one gas filling system, only the full-scale combustion chamber is operational. The non-focused ignition phenomena was again observed now in the large vessel, with Fig. 5 comparing the pressure signal of two sets of shots at two different filling pressures in the shock tube driver. The initial conditions are presented in Table 3.

Table 3
Driver shots parameters for Fig. 5.

Shot nr.	p_{fill} (bar)	p_{peak} (bar)	ϕ	X_{He} (%)	Focus
S028	50	334	0.85	72	Yes
S030	50	367	0.85	72	No
S046	80	756	0.72	70	No
S047	80	563	0.72	70	Yes

4. Optical transmissivity measurements

The experimental setup comprises two main elements: a high-pressure combustion chamber ("bombe") and a high-power pulsed

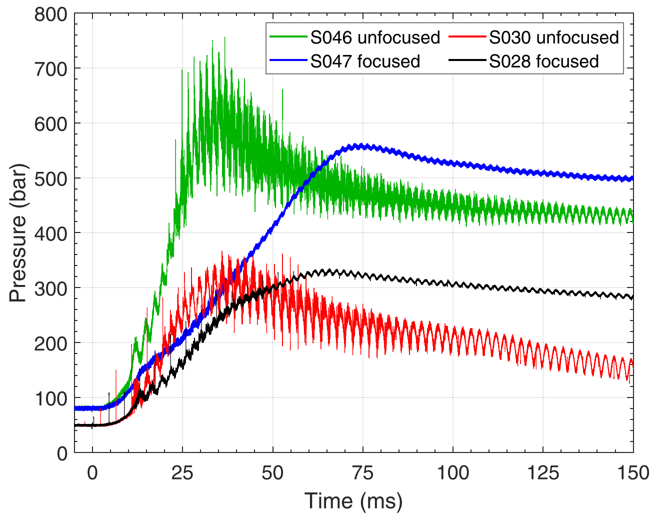

Fig. 5. Pressure signal comparison of S028 *vs.* S030 and, S046 *vs.* S047. Parameters shown in Table 3.

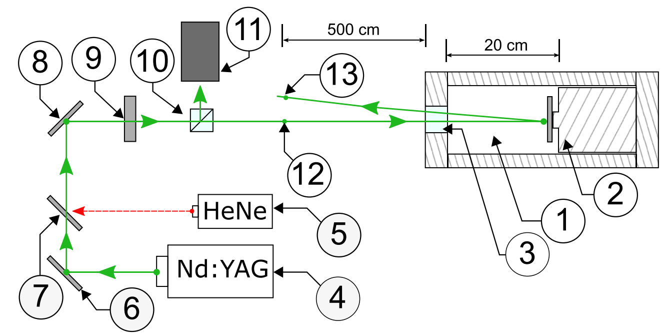

Fig. 6. Schematic for the Laser absorption experiment, not to scale. 1, 2, 3 - elements in the "bombe"; 4–13 - elements on the optical breadboard. 1 - Combustion chamber; 2 - chamber reduction cylinder and 0° mirror; 3 - Fused silica window 4 - Nd:YAG laser (λ_0 = 1064 nm); 5 - HeNe laser; 6 - 45° mirror; 7 - Beam combiner; 8 - 45° mirror; 9 - Half-wave plate; 10 - Beam splitter cube; 11 - Beam dumper; 12 and 13 - Laser power reading location before and after crossing the "bombe".

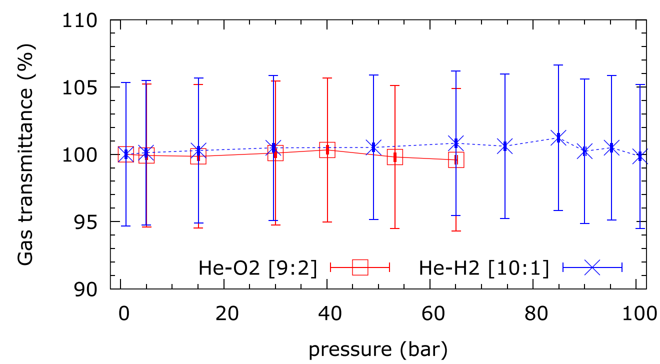

Fig. 7. Test gas transmittance as a function of filling pressure, for mixtures of He-O_2 [10:1] and He-H_2 [9:2]. Error bars above one hundred have no physical meaning.

laser. The combustion chamber has an associated gas filling system and the laser has an associated beam conditioning system. A cylinder is positioned inside the combustion chamber, at the opposite side of the window to support a 0° mirror used to reflect the high power beam that enters the window. The window itself is a fused silica cylinder of 50.8 mm diameter and 10 mm thickness.

The gas filling system description may be found in [19]. The high-power laser and the beam conditioning elements are positioned on an optical breadboard, in front of the optical port of the "bombe". The high-power laser is a Quantel Brilliant laser, a 10 Hz Q-switched Nd:YAG laser emitting 200 mJ pulses at 1064 nm, its fundamental wavelength. The laser is placed facing the opposite of the line of sight towards the "bombe" window, with two 45° mirrors, allowing height and azimutal deviation, deflecting the laser beam towards the "bombe". To help the alignment, a continuous-wave He-Ne laser beam with 3 mW at 632.8 nm is combined with the Nd:YAG one. In such way, and for security reasons, the Nd:YAG laser is kept switched off whenever possible. The half-wave waveplate and the beam-splitter are used to regulate the beam power, a common procedure with linearly polarized laser beams, as is the case. The unwanted power is diverted from the beam splitter cube to the beam dump. The mirrors, beam splitter, beam combiner and half-wave beamplate are from CVI optics. The only two optical elements not installed in the optical breadboard are the silica window in the optical port of the "bombe" and the 0° mirror in the end of the combustion chamber. This last mirror is reflecting at an angle slightly off-axis, to separate the entering beam from the exiting one, allowing the measurement of the laser beam power in both situations on the breadboard by a powermeter, a Coherent Molectron Powermax 500 A. The schematics of the setup are presented in Fig. 6.

4.1. Experimental procedure

The laser is run at 10 Hz, the beam entering the combustion chamber with pulses averaging 200 mJ after conditioning. No focusing of the laser occurs throughout its optical path. The chamber is filled with an He-O_2 [10:1] and then an He-H_2 [9:2] mixture up to a pressure of 100 bar. This mimics a nominal [8:2:1] He-H_2-O_2 combustible mixture without the associated reactivity. Repeating the experiment twice with those two mixtures allows pinpointing the influence of chemical composition from any differences in absorption that may arise between these two sets of experiments. Then, the absorptivity of the He-H_2-O_2 mixture may be correlated from both partial mixtures absorptions.

The initial measurement is carried out at maximum laser irradiance, with subsequent measurements being carried out at lower irradiance, adjusted using the half-wave plate. After about 5 to 10 different input powers, the pressure in the chamber is reduced using the venting valve, and the procedure is repeated. Once the first mixture (He-O_2 [10:1]) is concluded, a second run is carried out in a similar fashion, considering a (He-H_2 [9:2]) mixture.

To make a final cross-check of the experiment, allowing us to evaluate any spurious power losses through air and optical components, another run of the experiment is made with an open chamber and without the silica window, and a final run measuring the power loss between points 12 (P_{in}) and 13 (P_{out}). No measurable loss to the sensitivity of our powermeter could be identified. A measurement of input/output power is carried out with the window and the mirror placed inside the combustion chamber in room air, yielding an "effective" transmittance of about 55%

5. Results and discussion

The transmittance $\mathcal{T}$ of the setup was calculated using Beer–Lambert's law, see Eq. (2). The values are then corrected by the silica window transmittance, to have the gas transmittance values, presented in Fig. 7. Gas transmittance is constant and around 1 for the pressure range between 1 and 100 bar, and for both mixtures. This means the gas is optically transparent to the 1064 nm laser radiation, even though at 100 bar there is 100 times more matter inside the chamber.

The experiments show that the laser energy is not absorbed directly by the gas mixture. The result is somewhat expected as there are no electronic transitions for helium, oxygen or hydrogen near the 1064 nm region to absorb the laser photons. The gas transparency to the laser implies that seed electrons are not generated by MPI. In multiphoton ionization an atom/molecule absorbs a number of photons so that it releases an electron as it ionizes. This free electron would rapidly absorb the laser's energy via inverse bremsstrahlung that would create a plasma that in turn decreases the gas transmittance. As stated by Srivastava [9], the laser's irradiance should be around 10^{14} W/cm^2 to create the first electrons through the MPI process. It is our opinion that the seed electrons do come from impurities in gas mixture, as defended by [1,2,5,9–11] because an unfocused laser beam is incapable of reaching the irradiance levels in the order of 10^{14} W/cm^2. Nonetheless ignition/breakdown is achieved at lower irradiance levels as shown in Table 1. The referred authors argue that seed electrons coming from the ionization of microparticles explains the strong pressure dependence of MIE [2,8,11–13], which we can also observed as reported in Section 3. This pressure dependence for the MPE observed by [2,8,11–13] is also evident in this work, where ignition could be achieved with a non-focused laser, but only if sufficiently high gas filling pressures are used, with ignition being achieved with multiple laser pulses above $p_{fill} = 20$ bar, and with a single laser pulse above $p_{fill} = 30$ bar.

The laser has a pulse energy of 193 ± 2 mJ, a time width of 5 ns and a measured area of 23.15 ± 0.43 mm^2, which yields an irradiance of $(1.336 \pm 0.115)\times 10^8$ W/cm^2. This irradiance is not sufficient to ensure a visible electrical breakdown of room air, yet it is sufficient to ignite our high pressure combustible mixture. Phuoc in [5] asserts that once electrical breakdown is achieved, gas ignition will necessarily follow up. Nevertheless, it seems to be the case that ignition may be achieved without a macroscopic electrical breakdown of the gas in the usual fashion, with a bright flash and the emission of noise. It cannot be ruled out that an ignition kernel may develop as the result of gas breakdown owing to electron release by dust/impurities and this hypothesis remains speculative pending more studies.

6. Conclusion

Laser-induced spark ignition is a process highly influenced by the gas filling pressure. The beam irradiance is key in determining the ignition success. We report that for pressures above 20–30 bar one can ignite a He-H_2-O_2 mixture with a non-focused laser. This translates to an irradiance around 10^8 W/cm^2, two to six orders of magnitude below the reported literature in Table 1. Despite being incapable of creating a macroscopic spark and visible electric breakdown in atmospheric air, the laser setup can still excite and ignite the mixture. The electrical breakdown may be initiated by a microscopic spark formed by impurities excited by the laser.

A laser absorption experiment was done to measure the energy absorption of a unfocused Nd:YAG laser at 1064 nm for two gas mixtures with pressures ranging between 10 and 100 bar. Less than 1% (5% with error bars) of the energy is absorbed by the gas[2] No significant differences in absorption were observed between the He-O_2 and He-H_2 mixtures, nor within the 10 to 100 bar of filling pressure range. One explanation for this non-focused laser ignition at high pressure may be related to the unintended presence of solid microparticles, like dust, in the chamber. This hypothesis was already proposed by other authors [1,5,8,12], where impurities absorb the laser energy leading to high temperature spots, which produce free electrons. These will start the avalanche process previously described, by exciting the gas and the chemical reactions. The initiation effect is expected to have no wavelength dependence, which is in agreement with the absence of electronic excitation transitions from ground state for He, O_2 or H_2 near 1064 nm.

Further research is needed to more extensively understand why and in what conditions these events can take place. More ignition experiments could be performed with a careful tuning of the relevant parameters such as filling pressure and laser irradiance, coupled to a careful statistical analysis of the results. Another possibility pertaining to the impurities autofocusing hypothesis is to test ignition using ultra-high purity gases. The opposite option would be to seed the combustible mixture with dust/aerosols. Then the threshold pressures for unfocused ignition could be compared. Ultimately, one should try as much as possible to optically access the interior of the chamber, in the attempt to visually observe the corresponding ignition kernels.

[2] A more detailed error propagation analysis may be found in [20], which means a very small portion of the laser pulse energy is sufficient for triggering ignition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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