Laser Ignition of a High-pressure H_2/He/O_2 combustible mixture

Ricardo João Grosso Marques Ferreira
ricardojoaogmferreira@tecnico.ulisboa.pt
Instituto Superior Técnico, Lisboa, Portugal
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Laser ignition is an efficient method to ignite high pressure combustible mixtures. It consists on focusing a nano second laser pulse into a small spot size creating an electrical spark and igniting the mixture. The performed tests at the ESTHER shock-tube facility reveled that unfocused laser beam could ignite mixture with filling pressures of 30 up to 100 bar. The irradiance in this condition were about 10^8 W/cm^2, three orders of magnitude lower than the average irradiance reported in the literature. We set to study how the gas absorbs laser at this high pressure conditions, which revealed to be $< 1\%$ of total laser energy and supports the seed electron theory providing breakdown in laser-induced spark ignition. Ignition is provided by the rapid ionization of volatile dust particles present in the mixture, exciting H_2 molecules, forming highly reactive radicals and starting the chain reactions. Another experiment was done in CCRC at KAUST measuring the minimum pulse energy for H_2-air mixture at atmospheric pressure, showing a lower pulse energy for stoichiometric mixture and difficulties in igniting mixture close to the lean flammability limit.

Keywords: Laser, Ignition, ESTHER, High Pressure, Hydrogen.

I. INTRODUCTION

A new shock-tube for the support of Planetary exploration is currently being developed at Instituto Superior Técnico, under funding from the European Space Agency. This pioneering experimental facility Portugal, starts a new age for the portuguese (and european) space research thanks to a state-of-the-art design developed inhouse. In order to create the shockwave a rapid expanding gas is needed, which is achieved by the combustion of H_2-O_2-He mixtures. Several experiments were conducted in the combustion chamber to qualify all the different systems of ESTHER facility. Initially, a laser ignition system using a focusing lens was used to increase the irradiance and ignite the gas mixture, [1]. The interesting question arose from successful ignition from filling pressure equal or higher than 20 bar with and without focusing lens, which was not reported in the literature, for this range of gas filling pressure. This result was coherently observed throughout the experimental campaign, proving it was not an aberration of the system, but a physical phenomena. This phenomena was not understood and motivate us to research and try to comprehend the reason and mechanism behind this.

Laser ignition is a new and promising method to ignite combustible mixtures. Nowadays the commercial source of ignition in gas turbines and reciprocated (commonly known as piston) engines are manly electric spark-plugs. Laser ignition provides important advantages as technology looks for more efficient and less pollutant combustion devices. Moreover, higher efficiencies can be achieved using higher pressure and leaner fuel:air equivalence ratio (ϕ) mixtures, as [2, 3] report. Some of the problems associated with spark-plugs include secondary high voltage breakdown, poor timing sensor reliability and electrode degradation. It is in this conditions that laser looks as a promising choice of ignition, whose main advantages are, for example: choice of arbitrary position and timing of ignition; absence of quenching effects by the spark plug electrodes; no erosion effect on the electrodes; Easier multipoint ignition and shorter ignition delay and shorter combustion time.

Fundamentally laser ignition can be achieved through four different methods or phenomena [4]. The method of excitation can be divided and into four categories: a) **Laser Thermal ignition** - Thermal ignition utilizes the energy of the laser beam to increase the kinetic energy of the target molecules, in either translational, rotational, or vibrational form. This will break chemical bonds of molecules starting the chain branching reactions. This kind of ignition occurs along the beam's path and not point-like. To avoid the formation of shockwaves there is a need to use continuous wave laser beams with wavelength (λ_0) absorbed by the gas mixture. b) **Laser-induced photochemical ignition-** In this mechanism, laser photons dissociate the target molecules into highly reactive radicals. These radicals will then start the chemical chain-branching reactions, ignition and full-scale combustion. A clear disadvantage for practical application is that the laser wavelength and target molecule's absorption wavelength require a close match. c) **Laser-induced resonant breakdown ignition** - This process involves two steps, first a non-resonant multiphoton photodissociation of some molecules in the gas mixture, followed by the resonant photoionization of one, or more, of the generated atoms. This two processes lead to the formation of seed electrons, which absorb more energy leading to a formation of a plasma and then to ignition. Once again, the is a need for tuning the laser wavelength to resonate with the gas mixture. The final part of this mechanism is similar to spark ignition, which will be described next. d) **Laser-induced spark ignition** - Laser spark ignition is probably the most useful method for practical applications due to not being necessary a tuning of the laser's wavelength. The process begins with multiphoton ionization (MPI) of few gas molecules which release electrons that readily absorb the laser's energy via the inverse bremsstrahlung process, increasing their kinetic energy. These accelerated electrons collide with other molecules and atoms ionizing them, releasing more electrons, which then accelerate and repeat the process. This creates a electron avalanche leading to a plasma formation which then starts chain-branching reactions and igniting the mixture. Multiphoton ionization processes are in some cases essential for the initial stage of breakdown because photon energy is less than ionization energy. When using ultrashort pulses (picosecond order) the MPI process must provide electrical breakdown all by itself, since there is insufficient time for electron-molecule collisions to occur, the electron avalanche refereed before. While with nanosecond pulses there is time for electron-molecule collision to generate more electrons, forming a plasma through this electron avalanche. A spark originated by this mean is a source of highly reactive chemical intermediates at very high pressure and temperature. This spark emits light, heat and shock wave to the surrounding medium, resulting in a ignition kernel. A sufficiently strong kernel allows a transition into full scale combustion.

Loss processes such as electron diffusion, radiation, collisional quenching of excited states, among others, are present an can increase the effective ignition energy. A spark originated by this mean is a source of highly reactive chemical intermediates at very high pressure and temperature. This spark emits light, heat and shock wave to the surrounding medium, resulting in an ignition kernel. A sufficiently strong kernel permits a transition into full scale combustion. Lee et al. [5] used glass fibers coated with different materials to study the plasma composition influence on minimum ignition energy. They conclude that minimum ignition energy was independent of the constitution for short pulses, while for longer (1 ms) the presence of inhibitors could extinguish the flame.

Syage, Fournuier [6] studied ignition of hydrogen/air and hydrogen/air/CO_2 mixtures using different pulses. The results show pulses duration below 15 ns and laser's wavelength do not affect the process and the minimum ignition energy. Spiglanin, MCilroy et al. [7] investigate the shape and the structure of developing flame kernels as function of time and mixture composition in laser-induced spark ignition in hydrogen/air. Gas motion dominates early flame kernel growth but, the ignition's 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 [8] studied the ignition probability as function of the ignition location for a diffusion jet flame. It was observed that as long as the pulse energy is high enough to create 100% gas breakdown, the ignition probability is not impacted by the higher values of pulse energy. Thus, the distribution of ignition probability is influenced by local variations of air:fuel equivalence ratio within the jet, as well as turbulence intensity and velocity gradient at the ignition location are also factors that influence ignition probability.

The common factor in all is that the laser's energy stimulates the gases molecules increasing their energy up to the ignition energy of the mixture.

Minimum laser intensity For an electric breakdown to occur in a gaseous mixture initial electrons are required to start an electron cascade. As stated before, these electrons may come from a process called multiphoton ionization, where a molecule absorbs laser photons and become ionized. A significant wavelength dependence is expected as this is a quantum effect. Srivastava [9] state that for the first electron to be produce by multiphoton ionization the irradiance, I, should be of the order $10^{14}\mathrm{W/cm^2}$. 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 wheres a typical Nd:YAG laser with a λ_0=1064 nm has a photon energy of 1.25 eV. Thus, 10 ou more photons are needed to produce one free electron.

Experiments and studies done by many different authors report different values for breakdown intensity, yet always orders of magnitude below $10^{14}\mathrm{W/cm^2}$. For example, Srivastava reports the intensity in the focus to be of $10^{12}\mathrm{W/cm^2}$, while Phuoc [4] even stats 10^{11} $\mathrm{W/cm^2}$ to be sufficient. Several authors, [9–13] defend that this seed electron do not come from multiphoton ionization but from impurities in gas mixture (e.g. dust, aerosol or soot particles). This hypothesis is supported by a non dependence of minimum pulse energy (MPE) on the laser's wavelength and strong pressure dependence, reported by [8, 12–15].

The parameter which determines the ignition success is the laser's irradiance. It is defined as the power per unit area, as follows. For high enough irradiance a spark is possible to be created, which ignites the mixture. The average irradiance I is given by,

$$I = \frac{E_p}{\Delta t_{pulse}} \frac{1}{A} \quad = \frac{P_p}{A}, \tag{1}$$

where Δt_{pulse} is the pulse time duration, E_p is the pulse's energy, P_p is the pulse's power and A the pulse's area.

Laser absorption When a laser beam passes through a medium it gets partially absorbed. The fraction of absorbed laser depends on the medium composition itself and on its optical length. The laser power absorption follows the Beer-Lambert law, which in integral form is given by:

$$P = P_0 \exp(-\alpha \cdot l) = P_0 \cdot \mathcal{T} \quad , \tag{2}$$

where P_0 is the initial power of the laser, α the absorption coefficient, l the optical path length and t the transmittance coefficient. If the total (initial) power, P_0, is given by the sum of transmitted power and absorbed power,

$P_0 = P_{abs} + P_{trans}$, therefore from Eq. (2), P_{abs} is given by:

$$P_{abs} = (1 - \exp(-\alpha \cdot l))P_0 \quad . \tag{3}$$

It is clear from Eq. (3) that the deposited energy in the mixture only depends on the absorption coefficient, since the laser power and optical length are constants for a fixed setup.

A combustion phenomena is characterized by several different factors, its pressure p, temperature T, duration, mixture, etc. One of the most important parameters is the equivalence ratio, ϕ, this ratio relates the amount of fuel and oxidizer (normally air) in a particular mixture to its stoichiometric conditions, as follows:

$$2H_2 + O_2 + He \rightarrow 2H_2O + He$$

$$\phi = \frac{n_{fuel}}{n_{air}} \bigg/ \left(\frac{n_{fuel}}{n_{air}}\right)_{stoic} \quad . \tag{4}$$

In stoichiometric conditions (ϕ=1) there is no excess of neither air or fuel in the gas mixture, in rich condition (ϕ¿1) the fuel is in excess and in the lean condition (ϕ¡1) there is an excess of air in the mixture. Depending on the values of ϕ the mixture will successfully ignite or not. The limits of the ignition are known as flammability limits.

As reported in [1] ignition was achieved without a focusing lens, meaning that the laser's irradiance is up to 4 orders of magnitude less to what the literature reports. The reasoning to this relates to the filling pressure, as ignition for low pressure could not be achieved without lens. To achieve ignition, a flame kernel must be developed with a minimum energy density, as presented in [16] and the energy must be provided by the laser. A non resonant and non focused laser cannot form a plasma through MPI process, meaning that the gas mixture must absorb energy throughout other processes. To investigate the laser absorption at high pressures, an experimental setup was designed and assembled utilizing ESTHER facilities in the Hypersonic Plasmas Laboratory. The focus of this experiment is to understand how transmittance varies for different pressures and power fluxes.

A second experiment was designed and assembled to study how minimum pulse energy and ignition probability varies as function of the air:fuel equivalence ratio for H_2-air pre-mixtures flows. This was done in the Clean Combustion Research Center at King Abdullah University of Science and Technology (KAUST), under the supervision of Professor Deanna Lacoste.

II. EXPERIMENTAL SETUP

A. Laser absorption at high pressures in ESTHER

This first experimental setup was composed of two parts, the combustion chamber and the laser and respective optical components. The alignment was made using a diode red laser as guide, so when the red and Nd:YAG lasers are aligned the latter is turned off as a safety measure. The combustion chamber is composed of a cylindrical metal vessel with 80 mm inner diameter and 600 mm of length, with a bolted top cap having a fused silica window as laser's entry point to the chamber as depicted in FIG. 1. The optical setup is shown in FIG. 2. A cylinder was placed inside the chamber to support a 0° mirror designed to reflect the Nd:YAG laser beam. The Nd:YAG laser (Quantel Brilliant, 1064 nm, 360 mJ, 5 ns) is reflected by two 45° mirrors (High power CVI mirrors) for beam height and azimuthal deviation, passes through a half-wave plate and a beam splitter cube, then proceeds entering the chamber. It is possible to adjust the laser power sent to the chamber using the the half-wave plate and the polariser cube. The half-wave plate linearly polarizes the light and the beam splitter cube divides it as function of the polarization, part into a beam dumper and the other in direction of the combustion vessel. Inside the chamber the beam is reflected so a measurement of its power can be performed after crossing the combustion chamber. The experiment follows these steps:

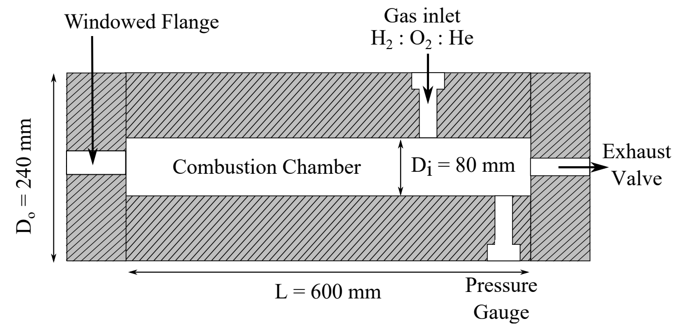

FIG. 1: Schematic representation of the ESTHER's combustion chamber prototype.

A powermeter (Coherent Molectron Powermax 500A) was placed right before the silica window to measure the input laser power, and then placed in a point of the beam's optic path to measure the transmitted power.

Filling of the chamber with an He-O_2 [10:1] or He-H_2 [9:2] mixture up to a filling pressure of 100 bar. This is done to mimic a [8:2:1] He-H2-O2 combustible mixture [1], but without the reactivity (since firing the laser in such a mixture would initiate ignition and would destroy the mirror inside the vessel). Any differences in absorption from the He-O_2 and He-H_2 experiments can be attributed to the different chemical compositions, allowing the determination of the absorptivity of the He-H_2-O_2 combustible mixture through a correlation of both partial mixtures absorptions. A round of experiments with the aforementioned He-O_2 mixture was firstly done by

[1] numbers represent each species molar fractions

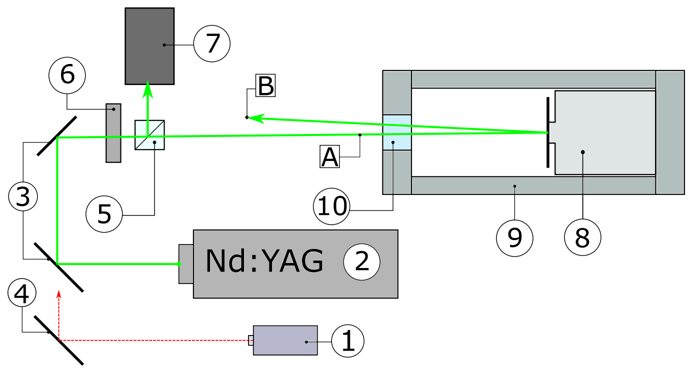

FIG. 2: Schematic for the Laser absorption experiment.

1 - Alignment red diode laser; 2 - Nd:YAG laser (λ_0= 1064 nm); 3 - 45° mirrors; 4 - 45° mirror; 5 - Beam splitter cube; 6 - Half-wave plate; 7 - Beam dumper; 8-0° mirror and support cylinder; 9 - Combustion Chamber; 10 - Fused Silica window; A - Entry power measuring point; B - Exit power measuring point.

measuring the laser input power at point A (FIG. 2), followed by measuring the laser transmitted power at point B (FIG. 2).

The powermeter is then placed, again, at point A. The power is again adjusted using the half-wave plate and we repeat the measurements. After 5 to 10 different input powers, the pressure in the chamber is reduced using the venting valve. A second run of the experiment was then made using He-H_2 [10:1] with the same steps described above.

Finally, to evaluate any power losses through air and optical components, another run of the experiment was made with an open chamber and without the silica window, and a final run measuring the power loss between points A (P_{in}) and B (P_{out}). The values of P_{in} and P_{out} are the average power over time for a 10 Hz pulse repetition, therefore 2000 mW equates to an average E_{pulse} of 200 mJ.

It is important to notice a limitation of our experiment as the chamber only has one window. Thanks to that a 0° mirror must be placed inside to reflect the laser beam and, as referenced before, the combustible mixture cannot be used. For the final combustion chamber, there is the possibility to perform a multi-pass laser absorption experiment since two pairs of facing optical ports exist in the design. Obviously such experiments require better windows, with a transmittivity higher than 99% in the characteristic wavelength of the laser radiation.

B. Minimum pulse energy to ignite H_2-air mixtures

In order to measure the influence of fuel:air equivalence ratio, pulse repetition and pulse energy in the ignition probability of H_2-air flows at atmospheric pressure, the following setup was assemble. Identically to the ESTHER experiments, this setup is also divided in two parts, an optical system and the combustion/burner as depicted in FIG. 3. When comparing it with the ESTHER experimental setup two major differences are presented, the gas is flowing rather than being stationary and its pressure is not up to 100 bar but atmospheric. This implies the need to focus the laser, otherwise ignition cannot be achieved due to its low pressure. The optical system is composed of a Nd:YAG Laser (Litron Lasers Nano S 130-10, 1064 nm, 140 mJ, 6-8 ns, 2 mm radii, 10 Hz), two 45° mirrors (High power CVI Mirrors), one $f = -100$ mm, 1" diameter, bi-concave lens (Thorlabs), one $f = 700$ mm, 2" diameter, plano-convex lens (CVI) and one $f = 100$ mm, 2" diameter, bi-convex lens (Thorlabs). The laser beam is reflected in the two mirrors and then expanded through the bi-concave and plano-convex lenses, a Galilean beam expander. At last, the laser is focused using the bi-convex lens to a spot size. Using this lens setup a spark in air is formed centered with the burner about 1 cm above the burner, which will ignite the combustible mixture. The hydrogen flow is provided by pressurized bottle (99% purity), while the air from an external compressor, both regulated and controlled with a set of mass flow controllers (MKS Instruments) and a four-channel readout (MKS Type 247D). The gases are mixed in a mixing box before passing through a flash arrestor, preventing the flame from burning backwards in

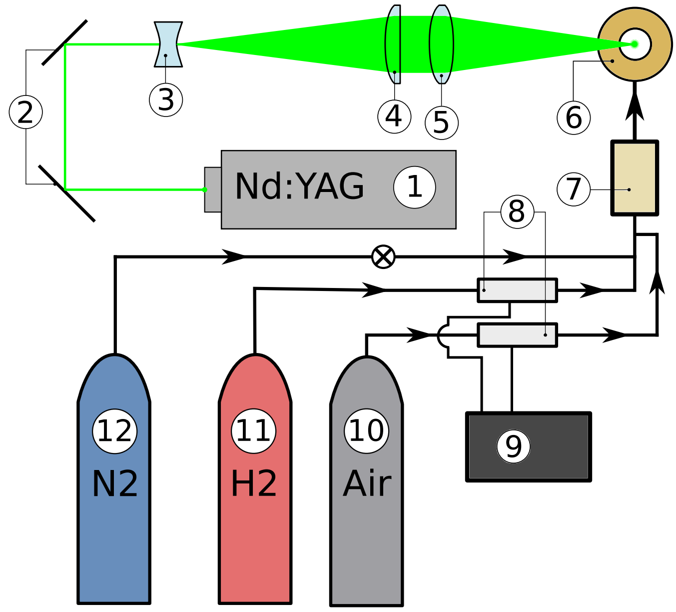

FIG. 3: Schematic for H_2-air ignition experiment.

1 - Nd:YAG laser; 2 - 45° mirrors; 3 - bi-concave lens (f=-100 mm); 4 - plano-convex lens (f = 700 mm); 5 - bi-convex lens (f = 100 mm); 6 - Burner (10 mm diameter); 7 - Mixing box; 8 - Massflow controllers; 9 - Four-channel readout; 10-12 - Compressed gas bottles.

accidental case. The gas flows at constant rate through the burner, and were tunned to be three to five times the burning velocity, S_L, of H_2-air mixtures at 1 atm. This prevent the flame from burning towards inside the burner and causing damage, also, it makes an ease and prompt to operate experiment. Finally, a N_2 gas line is connected to the mixing box which is open in order to extinguish the burning flame. To calculate the different flows, $\dot{Q}$, the following set of equations were used:

$$\begin{aligned} \dot{Q}_{tot} &= \dot{Q}_{H_2} + \dot{Q}_{air} \quad , \\ \dot{Q}_{tot} &= A_{burn} v_{burn} \quad , \\ \phi &= 2.381 \times \frac{\dot{Q}_{H_2}}{\dot{Q}_{air}} \quad , \end{aligned} \tag{5}$$

where ϕ is the fuel:air equivalence ratio, A_{burn} is the cross-section of the burner and v_{burn} the burning velocity of mixture. One must notice that in pure H_2-O_2 burning the stoichiometric coefficients is 1/2, but air is formed of a mixture of 1 part of O_2 to 3.762 parts of N_2. Burning velocities for different ϕ were taken from [17]. The burner has 10 mm of diameter, thus, A_{burn}=78.54 mm^2.

Before the start, the mass flow controllers were turned on and warmed up during 1 hour. The air and H_2 flows were regulated so the total flow velocity equals four-times the burning velocity for a fixed value of ϕ. The laser was then warmed up for 15 minutes and set to fire in burst mode, meaning it will fire N pulses and then stop. The air flow was open, followed by the H_2 and wait 1 minute to homogenize the mixture, the laser was then fired and checked if ignition was or not achieved. This was done 10 times, and the number of successful ignitions registered. Next, the laser's energy output was altered by varying the Q-Switch delay time, between 150 and 500 μs, and the experiment ran again. Finally, the flow values were regulated to a different fuel:air equivalence ratio and the described procedure repeated.

Note that the laser energy for each Q-Switch delay was measured and calibrated. The laser's maximum energy output was 140.8±0.2 mJ. The results obtained with a powermeter (Coherent Fieldmax II) are presented in Tab. I.

First and foremost is necessary to address that experimental uncertainty is to large in the latter measurements. For $P_{in} \leq 800$ mW the used powermeter is no longer accurate measuring P_{out} due to begin in the lower region of the scale, thus, data is inconsistent and must be filtered out. This is observable in Tab. II, whose two last rows present experimental uncertainties relative to the measurement of 7 and 14 %, respectively. Not only this, but the powermeter scale was set to a maximum of 3000 mW, meaning, this measurements were taken from the lower 10% of the scale, where most instruments are not calibrated. All the data was gathered and saved in similar manner to the one here presented.

Errors and uncertainties propagation was treated using a linear approach, since no set of statistical data was collected, therefore, only systematic errors/uncertainties and respective propagation were considered. The focus of this experiment is to understand how transmittance varies for different pressures and power fluxes.

TABLE I: Nd:YAG Laser pulse energy as function of the delay time of the Q-switch

Q-Switch delay (μs)	Energy fraction (%)	Pulse energy (mJ)
150	100	140.8±0.2
175	96.3 ± 1.1	135.7±1.7
200	88.8 ± 0.8	125.0±1.3
225	79.9 ± 0.5	112.5±0.8
250	73.1 ± 0.4	103.0±0.8
275	64.7 ± 0.4	91.0±0.7
300	53.4 ± 0.3	75.2±0.6
325	39.9 ± 0.3	56.2±0.4
350	30.5 ± 0.2	43.0±0.3
375	21.9 ± 0.2	30.8±0.3
400	13.7 ± 0.1	19.2±0.2

TABLE II: Input and output laser power measurements for He-O_2 mixture for 65.09 filling pressure.

P_{in} (mW)	P_{out} (mW)
1950 ± 25	875 ± 25
1585 ± 25	700 ± 25
1200 ± 25	550 ± 25
810 ± 25	350 ± 25
400 ± 25	175 ± 25

III. RESULTS AND DISCUSSION

A. Laser absorption at high pressure in ESTHER

Figures 4 and 5 show the overall transmittance of the optical system measured for He-O_2 and He-H_2 with partial composition of [10:1] and [9:2], respectively. The transmittance remains constant, about 0.45, for all the gas mixture filling pressures and laser input power between 1200 and 1950 mW. No observable difference was detected when changing the gas mixture composition.

Recalling Eq. (2), Beer-Lambert's law, the ratio $P(x)/P_0$ for a fixed position x is only function of the parameter α. This parameter is itself function of the medium on which the laser interacts and not of the laser itself, therefore is independent of P_0. In order to change α, the medium has to change, either becoming ionized by the laser or becoming saturated due to highly absorbed laser energy. Since neither of this nor others scenarios were observed, one can conclude that $d\alpha/dP_0 = 0$ for the current laser and gas parameters. In order to understand how the transmittance (and absorption) are function of the filling pressure, we took the average value for transmittance for fixed pressure, depicted in FIG. 6. One can

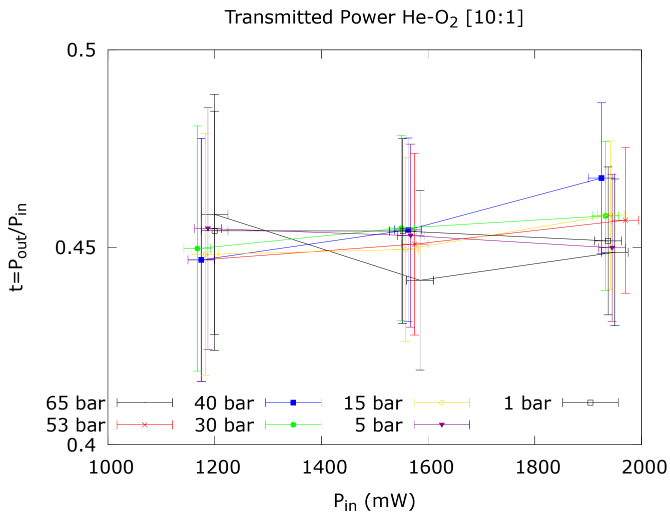

FIG. 4: Fraction of transmitted laser power, transmittance, as function of the input power P_{in} and filling pressure for a mixture of He-O_2 [10:1]

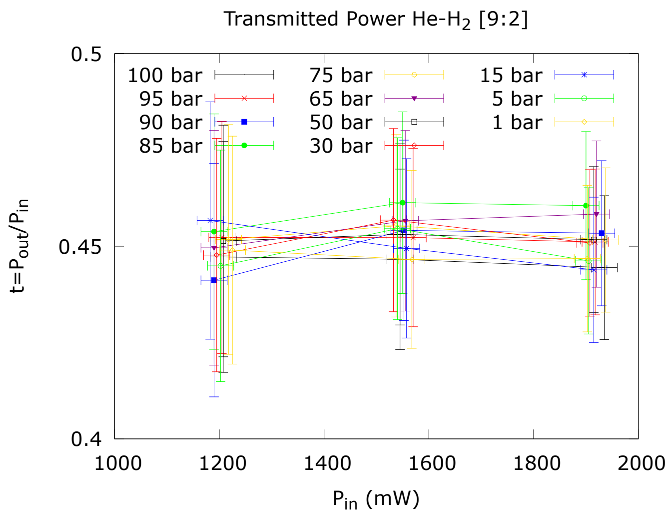

FIG. 5: Fraction of transmitted laser power, transmittance, as function of the input power P_{in} and filling pressure for a mixture of He-H_2 [9:2]

notice from Fig. 6 that the average transmittance over P_{in} remains nearly constant with the pressure. This observation implies that despite the density being 100 times higher in the beginning of the experiment comparing to the end, the fraction of energy absorbed by the gas, since all remaining mediums (silica window, mirror and optical distances) are set constant, is about the same.

The total transmittance $\mathcal{T}$ can be written as follows,

$$\mathcal{T}(p) = \mathcal{T}^2_{gas}(p) \times \mathcal{T}^2_{air} \times \mathcal{T}^2_{window} \times \mathcal{T}_{mirror} = \mathcal{T}_{opt}\mathcal{T}^2_{gas}(p) \quad , \tag{6}$$

where $\mathcal{T}_{gas}$, $\mathcal{T}_{air}$, $\mathcal{T}_{window}$ and $\mathcal{T}_{mirror}$ represent the par-

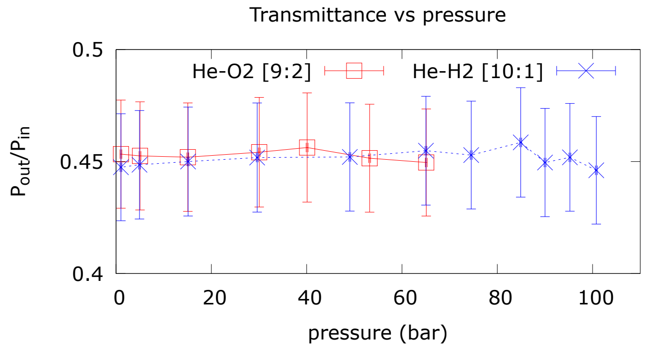

FIG. 6: Average transmittance for both mixtures function of filling pressure

tial transmittances of the chamber's gas mixture, air between powermeter and laser output, silica window and 0° mirror, respectively. The optical system transmittance is coupled into the $\mathcal{T}_{opt}$ term of Eq. (6). A squared term means two passes through, like the silica window, which is crossed twice by the laser beam. The gas filling system does not allow sub-atmospheric pressures inside the chamber, so the minimal achievable pressure was the atmospheric pressure p_0. In order to be possible to compute the gas transmittance $\mathcal{T}_{gas}$, a normalization was performed to the transmittance $\mathcal{T}(p_0)$. Separating the gas transmittance, $\mathcal{T}^2_{gas}(p) = \mathcal{T}^2_{gas}(\Delta p) \times \mathcal{T}^2_{gas}(p_0)$, where Δp is the excess pressure in relation to the atmospheric and coupling $\mathcal{T}_{gas}(p_0)$ with the optics part, $\mathcal{T}_{opt}$, we get:

$$\mathcal{T}(p) = \mathcal{T}_{opt} \times \mathcal{T}^2_{gas}(p_0) \times \mathcal{T}^2_{gas}(\Delta p) = \mathcal{T}_{p_0+opt} \times \mathcal{T}^2_{gas}(\Delta p) \quad . \tag{7}$$

Taking the data from FIG. 6, solving Eq. (7) in order to $\mathcal{T}_{gas}(\Delta p)$ and plot it, we get the gas transmittance as shown in FIG. 7. The transmittance of the gas mix-

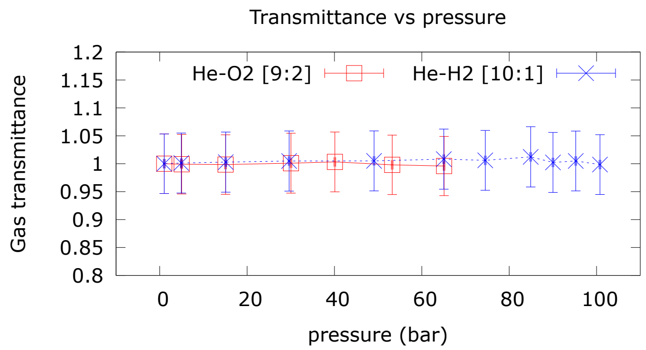

FIG. 7: Gas transmittance for both mixtures as function of filling pressure[a].

[a]Note that the error propagation analysis leads to error bars with transmittance above 1. This obviously has no physical bearing

tures is constant and about 1, meaning the laser energy absorption is nearly 0 for this laser's wavelength. This observation was expected as the laser's wavelength was not tuned for the absorption wavelength of the mixture composing gases. Another evidence to this conclusion is that both mixtures He-H_2 and He-O_2 transmission coefficients are, up to our precision, equal. As expected, FIG. 7 presents a nearly constant value of transmittance about 1. This results means that the gas mixture is transparent to the laser light. As also observed in FIG. 6, no significant differences exist between the two gas mixtures and, for both of them, the gas transmittance is ≈1. This supports the idea that the laser's energy is absorbed initially by dust and other micro-particles, not the gas molecules, forming the seed electrons.

We have already reported that laser ignition was achieved in the ESTHER combustion chamber without focusing lens. This is a scientific breakthrough due to the irradiance being orders of magnitude lower than the ones presented in the literature, $\geq 10^{11}$ W/cm^2. The laser setup parameters used as ignition system in ESTHER combustion chamber are presented in Tab. III, both laser spot area and optical transmittance for the silica window were measure, A_{beam}=23.15±0.43 mm^2 and $\mathcal{T}_{window}$=0.801, meaning I=1.336×10^8 W/cm^2.

TABLE III: ESTHER laser ignition system parameters

E_{pulse} (mJ)	Δt_{pulse} (ns)	A_{beam} (mm^2)	$\mathcal{T}_{window}$	I (10^8 W/cm^2)
193 ± 2	5	23.15 ± 0.43	0.801	1.336 ± 0.039

Weinrotter et al. in [10] made ignition experiments up to 4 MPa (about 40 bar) of filling pressure and reported a irradiance of 10^{11} W/cm^2, three orders of magnitude greater than our results. On this same reference the authors already hinted that first electrons are not generated through MPI, but by ionizing particles within the mixture. Our observation on the gas transmittance being pressure independent validates that dust and other particles are the ones which initially absorb the laser's energy. Whenever electrical breakdown occurs the laser's energy is absorbed by the plasma through inverse bremsstrahlung and transferred through pressure and heat to the unburned gas. The energy absorbed by the particles is low enough to not be detected by our setup, but can create a localized plasma hot enough to excite H_2 molecules around it and start the chemical chain reactions, leading to full scale ignition. Another observation was made, the initial silica window had several small points of damage on the inner face after 4 months and several combustion shots of use, hinting to being created as these micro particles are projected towards it when rapidly ionized by the laser.

B. Minimum pulse energy for ignition of H_2-air flows

A first run of the experiment was performed with 10 pulse burst and ϕ between 0.3 and 5. The optical system has an optical transmittivity of 92.68% (see the optical equations presented in Chapter 2) the irradiance is equal to 9.846 $\times 10^{14} E_{pulse}$ W/cm^2, with E_{pulse} in joules. Electrical breakdown was observed for E_{pulse} of 125 mJ or higher, meaning 1.233×10^{14} W/cm^2, which is in agreement with the literature. For lower energies and different optical arrangements sparks could occur but not at every pulse. This supports the idea of seed electrons being created by ionization of dust and/or micro particles. The ignition probability for a 10-burst pulse attempt is depicted in Fig. 8 as function of pulse energy and fuel:air equivalence ratio. In FIG. 8 we observe that for E_{pulse}=103 mJ

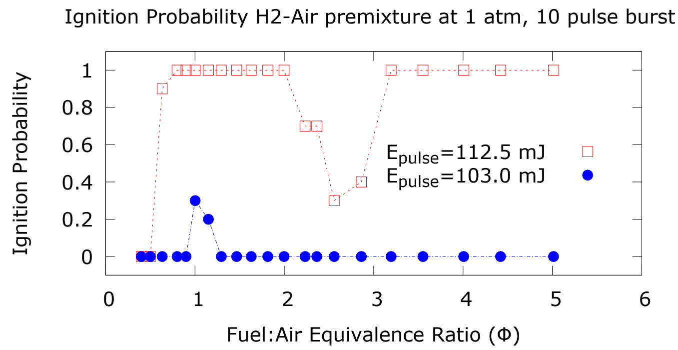

FIG. 8: Ignition probability for H_2-air mixtures at 1 atm and different ϕ and E_{pulse}.

the successful ignition in 10 attempts are 0 from ϕ=[0.4; 0.9], then it increases to 3 and 2 for ϕ=1 and 1.15, respectively. For $\phi \geq$1.3 and higher no successful ignition was achieved for this pulse energy. Also in Fig. 8 we show ignition probability for E_{pulse}=112.5 mJ, no ignition attempt is successful up to ϕ=0.65 with 9/10 successful ignitions. This probability then increases to 100% for ϕ=2.0, where it decreases to a minimum 3/10 at ϕ=2.65. Maximum probability is again reached at ϕ=3.2 staying there until ϕ=5.0.

Ignition was achieved using pulse energies smaller than the ones necessary to produce a visible spark and the rich side tends to be easier to ignite than the lean side. Both observations are in agreement with [4, 8, 10] whose observations report lower energies on the rich side comparing to the lean, with a minimum near the stoichiometric region. This is due to the formation of atomic excited highly reactive H-species, which is more difficult in the lean side. Notice that ϕ=0.4 or lower could not be ignited, despite being within hydrogen flammability limits and ignited with a lighter. One could also conclude that if the mixture is ignitable by a laser spark, by this means achieving a macroscopic spark in air ensures ignition, the reverse in not necessarily true.

It was expected, for a closed combustion vessel, that the minimum pulse energy to decreases up to stoichiometric region and then increases again. Our data does not reproduce this on the rich side of the mixture, presenting a decrease ignition probability after ϕ=2 but an increased ignition probability at ϕ=2.9. The reason for this can be relate to two phenomena, first minimum ignition energy increases far away from stoichiometric region, therefore pulse energy increases diminishing ignition probability. On the other hand, the region between ϕ=1.8 and 2.6 is characterized by high flow velocity which also increases the minimum ignition energy due to convective heat transfer on the flow, as in [18]. Another explanation for the non formation of a stable flame lies in the flame been blown out in the early stages of its formation. In the initial flame kernel the speed at which the energy and matter propagate outwards the kernel is slower than the burning velocity S_L. Since the mixture's flow velocity is four times S_L, the initial flame kernel could be blown out in a similar fashion to what occurs when the N_2 valve is open and blows out the flame. On the far rich side the flow velocity is reduced again so the flame kernel can grow and stabilize before been blown out. The reasoning behind this explanation does not deny the occurrence of ignition itself, but nullifies the flame development related to the gas dynamics and heat/mass propagation. Two observations were made during this experimental run, the air sparks stopped after ignition and the ignition timing was variable for every 10-burst shot.

From the first observation one could conclude that no major electrical breakdown occurs in a burning flame. Since the particles around the flame are in various excitation states, some of them are even ionized as they absorb the laser energy in a smoother way. Without a sudden energy deposition no shockwave is generated, meaning no audible sound is produced (or heard). The second observation relates to the ignition probability with the electric breakdown. In some shots ignition was only achieved nearly by the 10th pulse, implying that the electrical breakdown did not occur until around the 10th pulse. This is in agreement with the seed electron theory and earlier observations of air spark formation without combustible mixture. Due to the breakdown mechanism being associated with a stochastic event, low-ionization energy particles being found in the lasers focal spot, not every pulse is capable of generating an electron cascade, thus creating an electrical breakdown and igniting the mixture. The randomness of this phenomena relates to different factors: energy and spot size fluctuations on each laser pulse, local concentration of hydrogen species (easier ignition is achieved for higher hydrogen concentrations), local presence of dust and other impurities to form seed electrons. In order to study how the number of pulses influence on the ignition probability, the experiment was ran now with a 5-pulse burst instead of a 10-pulse burst. These results are presented in FIG. 9. In FIG. 9 we observe that for E_{pulse}=103 mJ the successful ignition in 10 attempts is 0 from ϕ=[0.4; 0.9], then it increases to 3/10 and 2/10 for ϕ=1 and 1.15, respectively. For ϕ ≥1.3 no successful ignition was achieved for this pulse energy, 103 mJ. In Fig. 9 is also shown the ignition probability for E_{pulse}=112.5 mJ, where ignition is unsuccessful up to ϕ=0.65, there it rises to 9/10 successful ignitions. The ignition probability increases to 100% for

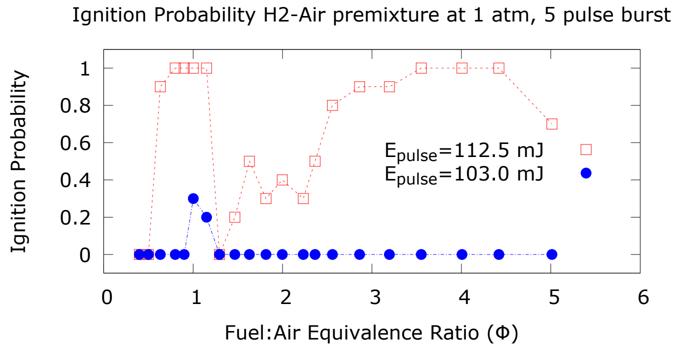

FIG. 9: Ignition probability for H_2-air mixtures at 1 atm and different ϕ and E_{pulse}.

ϕ=[0.8; 1.15], where after it abruptly decreases to a minimum of 0/10 at ϕ=1.3. The ignition probability starts to increase again in a non monotonous way, reaching a maximum probability at $\phi = 3.55$, presenting a decrease for $\phi = 5.0$ with 7/10 ignitions.

Comparing ignition with 10-and a 5-pulse burst, the general behaviour is similar in both regimes, with the 5-pulse regime having a lower success in igniting for ϕ between 1.30 and 3.20. Clearly, this is explained by the randomness of each laser pulse its interaction with the gas flow. As discussed before, spark formation is strongly related to the micro-particles presence in the air/mixture, which present a random/Brownian motion. For a pulse energy near the breakdown threshold, these fluctuations can be enough to hinder spark generation and therefore block ignition. One can also think of this as being a random event with a binomial distribution whose ignition probability is fixed between 0 a 1. After a sufficiently large number of events both results (ignition and non-ignition, in this case) will be present. Using 10-pulse burst increases the number of events and thus decreases the probability of every pulse lead to a non-ignition. Recurring to several pulses/ignition attempts for the same shot is not only a laser usual practice, as spark-plugs also recur to this technique to ensure ignition.

IV. CONCLUSIONS

A. Laser absorption at high pressure in ESTHER

As already stated, laser ignition was achieved in the ESTHER combustion chamber without focusing lens. This is a scientific novelty due to the irradiance being at least two orders of magnitude lower than the ones, until now and to the extent of our knowledge, presented in the literature. The experiment of laser absorption at high pressures lead to the following conclusions:

a) The transmittance of a gas is independent of the input laser power - In agreement with previous experiments and theoretical models, the fraction of power transmitted through a optical medium is constant, up to our laser power. As the light passes through the medium it can interact with it, this probability being governed by the absorption coefficient α. Even using the maximum power of the laser, the gas medium did not change, therefore its absorption coefficient remained constant.

b) Transmittance is, up to our experimental precision, pressure independent - The main goal of this experiment was to study how the laser energy is absorbed by the gas at high pressures. Our results show that, up to our experimental precision, no significantly transmittance difference appears between low and high filling pressures. Despite varying the pressure between 1 and 100 bar, the transmittance was nearly constant.

c) The energy fraction absorbed by a gas for an untunned laser wavelength is less than 5% - As expected for an untunned laser wavelength the absorption coefficient α is small. The Nd:YAG laser used has a wavelength of 1064 nm, belonging to the Infrared region, in contrast most of absorption wavelengths for O_2, H_2 and He gases are in the ultraviolet region. Even at filling pressures of 100 bar no significant changes on transmittance were observed, meaning that no significant fraction of energy was absorbed by the gas. The experimental precision allow a conclusion that for an unfocused beam less than 5% of the laser's energy is absorbed by the gas.

d) Seed electrons needed for electrical breakdown are generated from the ionization of dust particles and not by multiphoton ionization process - Since the gas practically does not absorb the laser light, when electrical breakdown occurs the seed electrons (first generated electrons) must come from ionization of the volatile and solid particles, not the gas. In fact, direct gas ionization is through MPI process (and is nearly pressure independent). In contrast, several experiments, including ours, the gas breakdown or ignition presents an evident dependence on the filling pressure.

e) High pressure H_2-O_2-He gas mixtures can be ignited without a macroscopic spark using a unfocused laser beam - The ESTHER setup did not produce a visible or audible spark in the combustion chamber. When an electrical breakdown occurs a sharp sound is audible due to the pressure shock-wave, and this did not occur. Besides this, if an electrical spark would be generated the laser would be more absorbed, through the inverse bremsstrahlung process. We already reported ignition without a focused beam, and also we did not observe a macroscopic spark formation on a non-combustible mixture, which allows us to conclude that ignition can be achieve at high pressure without a macroscopic spark. Other experiments thought that a macroscopic spark was needed in order to ignite the combustible mixture, therefore the necessity of focusing the laser beam. A summary of the observables for different laser and mixture conditions are shown in Tab. IV.

TABLE IV: Observables for laser-induced spark ignition in high-pressure regime

Mixture	Non-Combustible	Combustible
Focused beam	Macroscopic spark	Macroscopic spark and ignition
Unfocused beam	—	Ignition

B. Minimum pulse energy to ignite H_2-air mixtures

Looking at the data and its processing the main conclusions driven from the study and experiment done at KAUST are:

a) Laser-induced spark ignition cannot ignite H_2-air mixture at the lean flammability limit - In agreement with previous experiments from different authors, a H_2-air mixture at the lean flammability limit could not be ignited through laser ignition. The observations made during the experiment shown that, even when using maximum pulse energy (140 mJ), ignition could not be achieved for $\phi \leq 0.5$. In spite of being able to ignited mixtures with $\phi \leq 0.5$ with a simple lighter, it acted as an effective lean flammability limit for laser ignition.

b) Laser ignition is more effective at the rich side of a combustible mixture - The rich side of H_2-air mixture proved to be relatively easier to ignite than the lean side. Ignition was achieved with ease, even at ϕ=5.0, which is closer to the theoretical rich flammability limit.

c) Minimum Pulse Energy decreases in the stoichiometric region - In agreement with other experiments, the stoichiometric and near-stoichiometric regions (ϕ=[1.0; 1.15]) achieved ignition for lower pulse energy, 103 mJ corresponding to a irradiance of 1.013 $\times 10^{14}$ W/cm^2. Ignition probability decreases as ϕ goes away from stoichiometric region, meaning a rise in the minimum pulse energy.

d) Increasing the mixture's flow velocity increases the minimum pulse energy - Ignition probability presents a sudden decrease in the region with ϕ between 2.20 and 3.20. This unexpected increase in minimum pulse energy (therefore in minimum ignition energy as well) can be related to an increase in flow velocity. One can justify the increase in MPE recalling that S_L has a maximum for ϕ=[2.20;3.20], which means heat convective losses increase as well. In the rich side, flow velocity decreases again, and the ignition probability therefore increases.

e) Producing a macroscopic spark in air guarantees ignition with limits- A macroscopic and visible air spark could be formed for E_{pulse}=125 mJ. If the mixture is within the effective flammability limits for laser-induced spark ignition, forming a macroscopic electric breakdown releases enough energy to excite the surrounding species and igniting the mixture. This is observable as the mixture ignite 10/10 times and without delay for pulse energy above the spark formation threshold.

f) Ignition is a probabilistic event, function of several parameters, such as pulse energy, air:fuel equivalence ratio, pressure - Laser-induced spark ignition is highly related to initial seed electrons originated from the ionization of micro-particles. The presence of sufficiently particles and H_2 species in the laser's focal region to produce a spark and excite the hydrogen, forming reactive radicals is an aleatory event. The data comparing 10-and 5-burst pulse supports this, since the 5-pulse burst clearly presents less successful ignitions than the 10-pulse burst for similar conditions.

C. Final remarks

The principal conclusions we can draw from both experimental setups and this work are: 1) laser ignition is a process highly influenced by the filling pressure of the chamber; 2) the laser's irradiance is the critical factor to determine if ignition is successful or not. At high pressures, above to 20 bar, ignition can be achieved without focusing a laser, reducing its irradiance to values lower than the macroscopic electrical breakdown threshold. This means macroscopic electrical sparks are not created for an unfocused laser, which agrees with the second experiment where ignition was achieved with E_{pulse} inferior to the electrical breakdown threshold. Ignition is achieved through the excitation of gas molecules, in a high pressure environment the particle density is sufficiently high so these can be excited by a smaller, even microscopic spark. This small spark is generated by the ionization of volatile particles in the mixture. Richer mixture are easier to ignite, supporting the idea that hydrogen species are critical in the start of chemical chain reactions. Since no macroscopic spark is needed to ignite, the actual ignition region/point is before the focal point. Results from other authors [8, 18] already hinted towards this, as the spark was formed before the focal point.

By chance, some interesting and completely unexpected events were caught during our research, since the current scientific literature did not even considered it as a possibility. Namely, the possibility of reaching ignition with a laser irradiance at least two orders of magnitude lower than expected or starting a flame without creating a macroscopic spark.

Further research is needed to understand why and in what conditions these events can take place. The first thing that comes to mind is the comparison of focused *vs.* unfocused ignition.

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