Unfocused laser ignition of high-pressure He–H₂–O₂ combustible mixtures

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We report consistent ignition of high-pressure (p > 20 – 30 bar) hydrogen-oxygen mixtures diluted with helium, using an unfocused Nd:YAG laser. This corresponds to laser irradiances several orders of magnitude below the minimum ignition energies reported in the literature.

By placing a mirror inside a cylindrical vessel and filling it up to 100 bar with H₂–He or O₂–He non-combustible mixtures, we obtain the pressure-dependent absorptivity of the combustible He–H₂–O₂ 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). This leads credence to the theory that ignition stems from seed electrons created by autofocusing ionization of dust/impurities in the gas.

I. 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 occurs when a 10⁻³ fraction of the gas is ionized [3]. Seed electrons are accelerated by the laser’s energy, collide with other molecules, ionizing them, creating an electron avalanche, and leading to gas breakdown [3–6]. The defining parameter that ensures ignition is the laser’s irradiance I (power per unit area). This is defined in Eq. 1, where $E_{\text{pulse}}$, $\Delta t_{\text{pulse}}$ and $w$ are the pulse energy, pulse time width and beam radii at focal point, respectively. Several authors studied laser-induced spark ignition in different conditions and gas mixtures, and reported the minimum irradiance values for initiating combustion. These are summarized in Table I

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

TABLE I. Minimum breakdown or ignition irradiance reported in the literature.

| I(W/cm²) | Reference | Conditions/Model |
|---------|-----------|------------------|
| 1.93×10¹² | Bradley, [5] | Air at 1 MPa |
| 1×10¹⁰ | Phucoc, [6] | Theoretical |
| 1×10¹² | Phucoc, [7] | Air, CH₄, O₂, N₂, H₂ at 150 and 3040 Torr |
| 1×10¹⁴ | Srivastava, [8] | Drude model |
| 1×10¹² | Weinrotter, [1] | H₂-air 2.8 MPa air:fuel ratio 3 |
| 1×10¹⁰ | Lee, [8] | Propane-air at 1 atm |

The European Shock Tube for High Enthalpy Research (ESTHER) [9, 10] is a facility capable of reaching shock-speeds in excess of 10 km/s. The facility comprises a 50.3 liter 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 liter) “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 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 10⁶ W/cm². This value is two orders of magnitude below the reported minimums in the literature, which always consider a focusing lens in the setup.

Figure 2 presents the recorded pressure signal in both focused and unfocused ignition mode. A series of shots (#126, #127, #128, #135, #136) were measured for a filling pressure of 50 bar. The helium dilution is around 70% in terms of molar fractions, and the mixtures range from rich (#127 and #126 with $\varphi = 1.2$ and $\varphi = 1.1$ respectively), stochiometric (#127 with $\varphi = 1.0$), to poor (#128 and #135 with $\varphi = 0.8$ and $\varphi = 0.7$ respectively). Shots #126 and #127 were carried with a focusing lens, the others with the laser remaining unfocused.

One may further note that the combustion dynamics change significatively depending on whether the laser is focused or not. The focused shot pressure signals are considerably noisy, hinting at localized transition to detonation during the pressure rise, whereas the unfocused shots evidence smooth subsonic deflagrations. These and other phenomena (such as the shape of the pressure ride)
will not be discussed in detail here, and will be left for another upcoming publication by our group. The objective of this study is instead to report laser absorption characteristics in high pressure He–H$_2$–O$_2$ environments and infer on the possible mechanisms behind laser-induced spark ignition.

II. EXPERIMENTAL SETUP

The experimental setup comprises two main elements: a high-pressure combustion chamber a high-power pulsed laser. The combustion chamber has an associated gas filling system and the laser has an associated beam conditioning system. A cylinder was positioned inside the combustion chamber, at the opposite side of the window to support a 0º 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 [11]. The high-power laser and the beam conditioning elements were positioned on an optical breadboard, in front of the optical port of the “bombe”. The high-power laser was a Quantel Brilliant laser, a 10 Hz Q-switched Nd:YAG laser emitting 200 mJ pulses at 1064 nm, its fundamental wavelength. The laser was placed facing the opposite of the line of sight towards the “bombe” window, with two 45º mirrors, allowing height and azimuthal deviation, deflecting the laser beam towards the “bombe”.

To help the alignment, a CW He-Ne laser beam with 3 mW at 632.8 nm was combined with the Nd:YAG one. In such way, and for security reasons, the Nd:YAG laser was kept switched off whenever possible. The half-wave waveplate and the beam-splitter were used to regulate the beam power, a common procedure with linearly polarized laser beams, as is the case. The unwanted power was diverted from the beam splitter cube to the beam dump. The mirrors, beam spitter, beam combiner and half-wave beamplate were from CVI. The only two optical elements not installed in the optical breadboard were the silica window in the optical port of the “bombe” and the 0º mirror in the end of the combustion chamber. This last mirror was 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 500A. The schematics of the setup is presented in Fig. 3.

II.1. Experimental procedure

The laser was 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 \( \text{He–H}_2–\text{O}_2 \) 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 irradiances, 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 (\( \text{He–O}_2 \ [10:1] \)) is concluded, a second run is carried out in a similar fashion, considering a (\( \text{He–H}_2 \ [9:2] \)) 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 was 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 was carried out with the window and the mirror placed inside the combustion chamber in room air, yielding an “effective” transmittance of about 55%.

### III. RESULTS AND DISCUSSION

The transmittance \( T \) of the setup was calculated using Beer-Lambert’s law, see eq. (2) where \( P_t \) and \( P_0 \) are the transmitted and original beam power. The values are then corrected by the silica window transmittance, to have the gas transmittance values, presented in Fig. 4. 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.

\[
P_t/P_0 = T. \tag{2}
\]

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 multiphoton ionization. In multiphoton ionization an atom/molecule absorbs a number of photons so that it releases an electron as it ionizes. Srivastava [6] states that to generate the first electrons the irradiance should be in \( 10^{14} \) W/cm\(^2\). Ionization energy of \( \text{O}_2 \) molecules is 12.07 eV, 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 reports the intensity in the locus to be of \( 10^{12} \) W/cm\(^2\), while Phuoc in [4] even states \( 10^{11} \) W/cm\(^2\) to be sufficient. Several authors, [2, 6, 12, 13], defend that these seed electrons 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 the minimum pulse energy (MPE) with the laser wavelength, and a strong pressure dependence reported in [2, 7, 8, 13, 14], whereas multiphoton ionization predicts a very weak dependence. The strong pressure dependence is evident in this work, where ignition could be achieved with a non-focused laser, but only if sufficiently high pressures are achieved (with ignition being achieved with multiple pulses above \( p=20 \) bar, and with a single pulse above \( p=30 \) 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 \((1.336±0.115)\times10^8\) 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 [4] 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. Nevertheless, it cannot be ruled out that an ignition kernel may develop as the result of gas breakdown owing to electron release by dust/impurities, however such phenomena is not visible to our audible.

### IV. 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 pressure above 20 bar one can ignite a \( \text{He–H}_2–\text{O}_2 \) 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 I. 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 an 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, 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. The explanation for this non-focused laser ignition at high pressure may be related to the unintended presence of solid microparticles, like dust and soot, in the chamber. This explanation was already proposed by other authors [1, 4, 7, 14], 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. An experiment may be designed to measure minimum ignition energy and compare the focused and the unfocused regimes. Another possibility is to test ignition using ultra-high purity gases. The opposite option would be to seed the combustible mixture with dust/aerosol. Then the threshold pressures for unfocused ignition could be compared.

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1 A more detailed error propagation analysis may be found in [15], which means a very small portion of the laser pulse energy is sufficient for triggering ignition.

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