Normalization of absorbed energy and pressure in laser-induced breakdown in mono-atomic and molecular gases according to incident laser energy and initial pressure

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Abstract
In laser-induced breakdown experiments, the absorbed energy is one of the first measured parameters. For a given optical configuration and incident energy, the measured absorbed energy depending on pressure always exhibits a similar curve for the tested gases: argon, nitrogen, carbon dioxide and air. This work presents an empirical modelling to predict the pressure dependence of the absorbed energy in mono-atomic and molecular gas efficiently. The first series of experiments, involving Ar, N₂ and CO₂, presents its efficiency over pressure from 50 to 2400 mbar and incident laser energies from ∼15 to ∼135 mJ. The second series presents the effectiveness of this modelling on air. All experiments are conducted with a Nd:YAG laser at 532 nm and a focal radius of 4.23 μm.

Introduction
Since the first observations in the early 60’s [1, 2], laser–induced breakdown has been extensively studied and found many practical applications such as ignition of fuel-air mixtures, propulsion systems, flow control, and laser-induced breakdown spectroscopy (LIBS) [3–8]. Up to this day, the better understanding of the breakdown is still an ongoing topic. Theoretical, numerical and experimental efforts are made to determine the best conditions for laser-induced breakdown process, by particularly evaluating its dependence on pressure, laser beam characteristics and optical focusing system [9–11]. The value of the fraction of energy absorbed in the laser-induced plasma breakdown is a key parameter in the analysis of the breakdown process. To select the best model that replicates the experimental observations or to adjust parameters, a broad range of incident laser energies and gas pressures are needed.

The aim of this work is to propose a simple method to evaluate the absorbed energy with a limited series of measurements. A first step is to perform measurements over a wide range of laser energies and initial pressures on three different gases to define normalized parameters suitable for the proposed method. A second step is to test the effectiveness of this method on a limited series of measurements on a fourth gas.

Experimental setup
The mechanism of laser-induced breakdown formation is described as followed for our experimental setup: priming electrons are created via the multiphoton ionization and when the energy of the electrons exceeds the ionization energy, the multiplication of electron starts via the cascade ionization. Figure 1 depicts a scheme of the experimental setup. The laser source is a Nd:YAG (Quantel brilliant Q-smart 850) operating at the second harmonic at 532 nm over the pulse time τFWHM = 5 ns with a diameter D = 9 mm and a quality factor M² = 1.5 according to the manufacturer data sheet. A beam attenuator module (Quantel) allows to adjust the laser energy from 0 to 450 mJ. Prior to its focusing, the laser beam diameter D is expanded twice with a pair of plano-
concave \( (f = -50 \text{ mm}) \) and plano-convex \( (f = 100 \text{ mm}) \) fused silica lenses (both \( \varnothing = 25.4 \text{ mm}, 532 \text{ nm} \) coated). It is then focused with a plano-convex fused silica lens \( (f = 150 \text{ mm}, \varnothing = 50.8 \text{ mm}, 532 \text{ nm} \) coated) to achieve a beam waist \( \omega_0 = 4.23 \mu \text{m} \) and generating a breakdown in the center of the vessel.

The vessel is an octagonal prism with an internal volume of 1 l having windows to allow the laser beam to enter and exit. The internal pressure of the reactor is monitored with a 1 mbar accuracy manometer. At the exit of the vessel along the path of the laser beam, the laser energy is measured in mJ using a calibrated energy meter Ophir PE-50-DIF-C, with 3% accuracy.

The absorbed laser energy \( E_{\text{abs}} \) is deduced from the transmitted energy \( E_t \) and the incident energy \( E_{\text{inc}} \). The losses at the windows of the vessel are calibrated for each laser energy \( E \) [12].

**Experimental protocol for Ar, N\(_2\) and CO\(_2\)**

Three types of gases are selected: monoatomic with argon \( (\text{Ar}, \text{purity} \geq 99.998\%) \), diatomic with nitrogen \( (\text{N}_2, \text{purity} \geq 99.995\%) \) and triatomic with carbon dioxide \( (\text{CO}_2, \text{purity} \geq 99.7\%) \). The selected incident laser energies \( E_{\text{inc}} \), are \( \sim \{15, 30, 45, 60, 75, 90, 105, 120, 135\} \text{ mJ} \) and the initial pressures \( P \) are \( \{50, 75, 100, 125, 150, 175, 200, 225, 250, 300, 350, 400, 500, 600, 800, 1000, 1250, 1500, 1750, 2000, 2250, 2400\} \text{ mbar} \). For each couple \( (E_{\text{inc}}, P) \) 100 consecutive laser shots are performed with a burst frequency of 2.5 Hz in order to avoid some heating effect between each shot. Consequently, for each gas, the 9 \( E_{\text{inc}} \) for 22 \( P \) are studied, leading to a total of 19800 laser shots to characterize one gas. During the experiments, the laser fires continuously at a fixed energy \( E_{\text{inc}} \) to ensure a constant temperature of the lasing medium and initial pressures \( P \) are varying from the lowest to the highest with a primary vacuum done in the vessel between two consecutive pressures. All tests conducted with a single gas are performed on a single day, and for almost all the \( (E_{\text{inc}}, P) \) couples a breakdown was visible (a spark) for each laser shot (except on Ar for \( (15 \text{ mJ}, 50 \text{ mbar}) \), \( \text{N}_2 \) for \( (15 \text{ mJ}, 50 \text{ mbar}) \), \( (15 \text{ mJ}, 100 \text{ mbar}) \) and \( (30 \text{ mJ}, 50 \text{ mbar}) \), \( \text{CO}_2 \) for \( (15 \text{ mJ}, 50 \text{ mbar}) \)). The unsuccessful cases are associated with an incident energy at a given pressure below the so-called ‘breakdown threshold’ [2, 4, 13].

**Experimental results for Ar, N\(_2\) and CO\(_2\)**

All obtained results of measured absorbed energy \( E_{\text{abs}} \) as a function of initial pressure \( P \) suggests the following observations. Firstly, for all gases and all \( E_{\text{inc}} \), increasing \( P \) results in a rise of absorbed energy \( E_{\text{abs}} \) until a maximum value. This maximum value \( E_{\text{sat}} \) is known as the saturation regime and physically means that the plasma spatially grows instead of achieving higher temperature and density [13]. Secondly, all results yield same curved shapes (figures 2(a)–(c)). However, the pattern differs from gas to gas (figure 3).

From these two observations, the following assumption is formulated: if a normalized absorbed energy \( E_n \) and a normalized initial pressure \( P_n \) exist, then all results \( E_n \) on a gas can be presented as a function of \( P_n \) on a single curve. Practically, the next part of this work is then to determine the expressions of \( E_n \) and \( P_n \) allowing to scale \( P \) and \( E_{\text{abs}} \) axes to obtain a single coherent set of values.

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**Figure 1.** Main components of the experimental setup: (1) Nd:YAG laser Quantel brilliant Q-smart 850, (2) mirrors, (3) plano-concave lens \( (f = -50 \text{ mm}) \), (4) plano-convex lens \( (f = 100 \text{ mm}) \), (5) plano-convex lens \( (f = 150 \text{ mm}) \), (6) vessel and (7) energy meter.
Figure 2. Absorbed energy $E_{\text{abs}}$ (with 95% prediction bounds) as a function of the initial pressure $P$ for several fixed incident energies $E_{\text{inc}}$.

Figure 3. Example of curves for all gases for the same incident energy $E_{\text{inc}}$. 
Discussions on Ar, N₂ and CO₂

The normalization of \( E_{abs} \) is immediately found to be \( E_n = \frac{E_{abs}}{E_{sat}} \) and consequently, all curves have a unique asymptote \( E_n = 1 \) (figure 4). On this figure, \( E_{sat} \) is taken from experiments. From this normalization, when \( E_{inc} \) increases, the curve is stretched to the left with a sharper rise of normalized absorbed energy \( E_n \). However, this stretching seems to reach a limit, i.e. for high \( E_{inc} \) the amount of normalized absorbed energy \( E_n \) is independent from it. We assume that this limit is associated with the saturation regime \([13]\).

About the normalization of the pressure \( P \), figure 5 illustrates that it is possible to manually determine for each gas, a 'scaled pressure' \( P_n \), such that the plots of \( E_n \) as a function of \( P_n = P/P_c \) draw a single curve. From those observations, it is assumed that \( E_{sat} \) and \( P_c \) are unknown functions of \( E_{inc} \). However, such a procedure must be automated to be effective. Consequently, the choice of the functional \( f \) to perform it is crucial. The parameters optimization process, for all functionals tested on a single gas, is given in equation (1):

![Figure 4. Example of normalization of absorbed energy on Ar for four different \( E_{inc} \).](image)

![Figure 5. Normalized graph for Ar, with \( E_{inc} = 135.5 \) mJ as reference for \( P_n \) (manually adjusted).](image)
with x the vector of parameters involved in the functional and $E_{\text{sat}}$ and $P_c$ being considered here as parameters.

As presented in figure 6, S-shape curves clearly appear when a semi-log scale is applied on the pressure axis. This observation leads to evaluate sigmoid models such as Fisk, Dagum and Gompertz (equations (2)–(4) resp.) for the functional [14]. The Fisk model, the simplest used in this work, yields the best results among others. Contrary to others, the same function of $E_{\text{inc}}$ for all gases can model $E_{\text{sat}}$ and $P_c$. However, optimized values of $E_{\text{sat}}$ are generally higher than the observed ones, especially with N$_2$ and CO$_2$ for the highest $E_{\text{inc}}$ where the saturation phenomena is definitely observed during experiments, as shown in figure 7.

Unsatisfied by the obtained results, an effort to find out a better functional is carried out using a semi-log axis on energy (figure 8). The observed shape leads to consider a bi-exponential function on this semi-log graph and to propose the following functional (equation (5)):

$$f_4 = \exp (x_1 \exp (x_2 P_n) + x_3 \exp (x_4 P_n)) \quad \text{bi-exponential based functional}$$

Results of the normalization are presented for each gas in figure 9, and the values of all the obtained optimisation parameters are given in table 1.

Optimized $E_{\text{sat}}$ and $P_c$ are modelled as function of $E_{\text{inc}}$ via equations (6)–(7) (parameters given in table 1) and presented in figure 10.

$$E_{\text{sat}} = \varepsilon E_{\text{inc}} - \tau$$

$$P_c = \alpha E_{\text{inc}}^\beta + \gamma$$

For all the data, the correlation coefficient $R^2$ is greater than 99%.

Optimized $E_{\text{sat}}$ are in good agreement compared to observed ones, and the optimized scaled pressure $P_c$ for a given incident energy is always the pressure that leads to the same value of $E_n$ (0.28 for Ar, 0.58 for N$_2$ and 0.18 for CO$_2$). Then next part of this work is to confirm these results for air.

![Figure 6. S-shape evidence using a semi-log scale on pressure axis for Ar.](image)
Discussion: application on air

Extra-tests are performed on compressed air to confirm the results presented in the previous section using the same experimental protocol previously described but with a minimal experimental data set. To proceed, first tests are performed to find coefficients involved in equations (6) and (7) to quantify $E_{\text{sat}}$ and $P_c$ only with $E_{\text{inc}}$ for a few pressure values $P$. Each pressure condition has been tested with a minimal set of 3 different incident energies to determine $\alpha$, $\beta$ and $\gamma$ parameters in equation (7), and then used in turn to normalize other incident energies and initial pressures. The three chosen incident energies are $E_{\text{inc}} = \{15.0, 66.7, 142.6\}$ mJ and various pressures are selected during experiments to observe significant variations in the absorbed energy and also the saturation regime (figure 11). In this initial series, 32 $(E_{\text{inc}}, P)$ couples are selected with 100 laser shots for each of them.

Using equations (5), (6) and (7) leads to find $E_{\text{sat}}$ and $P_c$ as function of $E_{\text{inc}}$ for the initial series in air (figures 12(a)–(b)).

Then a second series with $E_{\text{inc}} = \{21.8, 40.1, 92.3, 125.4\}$ mJ and various pressures for 39 $(E_{\text{inc}}, P)$ couples (100 laser shots for each) is compared to the initial one: the obtained data points are displayed on the normalized graph (figure 13(a)), with those of the first series, for comparison. The figure 13(b) presents the comparison of data from the second series on air and the calculated $E_{\text{abs}}$ based on equation (5) with parameters taken from the optimization of the 1st series on air. Both show a good agreement.

Figure 7. Normalization using Fisk functional.
Figure 8. Bi-exponential evidence using a semi-log scale on energy axis.

Figure 9. Normalized presentation using bi-exponential function (equation (5)).
Conclusion

A normalization of absorbed energy $E_{abs}$ for a laser-induced spark with the incident laser energy $E_{inc}$ and the initial gas pressure $P$ has been successfully proposed for four different types of gas: mono-atomic ($\text{Ar}$), diatomic ($\text{N}_2$), triatomic ($\text{CO}_2$) and multicomponent (air). A unique 4-parameter bi-exponential based functional to

$$E_{sat} = E_{inc}^\varepsilon - \tau$$

and

$$P_c = \alpha E_{inc}^\beta + \gamma$$

were derived, as shown in Table 1. The optimized parameters are listed in Table 1 for each gas type, with $E_{inc}$ and $E_{sat}$ in mJ, $P$ and $P_c$ in mbar.

### Table 1. Optimized parameters to evaluate normalized absorption energy $E_n = E_{abs}/E_{sat}$ as a function of normalized pressure $P_n = P/P_c$ in laser-induced breakdown for different gases ($E_{inc}$ and $E_{sat}$ in mJ; $P$ and $P_c$ in mbar).

| Gas   | $x_1$  | $x_2$  | $x_3$  | $x_4$  | $\varepsilon$ | $\tau$ | $\alpha$ | $\beta$ | $\gamma$ |
|-------|--------|--------|--------|--------|---------------|--------|----------|---------|---------|
| $\text{Ar}$ | $-3.4524$ | $-1.6282$ | $-3.0346$ | $-1.6282$ | $0.9088$ | $-1.574$ | $1253$ | $-0.6651$ | $172.4$ |
| $\text{N}_2$ | $-3.7111$ | $-1.9194$ | $-2.2836$ | $-7.5600$ | $0.9898$ | $-3.045$ | $32011$ | $-1.617$ | $479.7$ |
| $\text{CO}_2$ | $-2.8847$ | $-0.7203$ | $-3.1578$ | $-2.3660$ | $0.9872$ | $-3.041$ | $3613$ | $-1.215$ | $127.1$ |
| Air   | $-5.2272$ | $-1.4938$ | $-2.9841$ | $-0.4407$ | $0.9643$ | $-4.850$ | $1574$ | $-0.9238$ | $82.5$ |

Figure 10. Optimized $E_{sat}$ and $P_c$ (full symbol) as a function of $E_{inc}$ with 95% prediction bounds dashed lines.

Figure 11. Initial air data points with 95% prediction bounds.
normalize absorbed energies and pressure of each gas is proposed and then validated with air experiments. This normalization points out that in the tested ranges of pressures and incident energies for all tested gases, their energy absorption $E_{abs}$ is experimentally predictable. The proposed normalization is useful to possibly complete data from in situ experiments or in the literature.

Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

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