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On the measurement of laser-induced plasma breakdown thresholds

Stefan Brieschenk,[1,a)] Harald Kleine,[2] and Sean O'Byrne[2]
[1]*Centre for Hypersonics, The University of Queensland, Brisbane 4072, Australia*
[2]*The University of New South Wales Canberra, The Australian Defence Force Academy, Canberra 2600, Australia*

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The breakdown threshold of a gas exposed to intense laser-radiation is a function of gas and laser properties. Breakdown thresholds reported in the literature often vary greatly and these differences can partially be traced back to the method that is typically used to determine breakdown thresholds. This paper discusses the traditional method used to determine breakdown thresholds and the potential errors that can arise using this approach, and presents an alternative method which can yield more accurate data especially when determining breakdown thresholds as functions of gas pressure. © *2013 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4819806]

I. INTRODUCTION

Plasmas generated by pulsed or continuous lasers[10,15,22] have been of particular interest in the fields of laser-induced breakdown spectroscopy,[18] chemical vapour deposition,[2] laser-driven inertially confined fusion,[5] drag reduction[19] as well as combustion augmentation and ignition[4] since they were first discovered in the 1960s. A unified theory of laser-induced breakdown does not exist at this point, and experimental measurements reported in the literature often do not contain all the details regarding the laser beam characteristics such as wavelength, linewidth, polarisation, unfocused diameter, divergence, pulse shape, and duration (transverse and longitudinal modes) as well as the characteristics of the focusing system such as focal length, aperture size, alignment to the laser beam, lens shape, lens quality, and refractive index[13,17] which would allow a comparison between different investigations. Threshold breakdown intensities for laser-induced plasmas (LIPs) reported in the literature differ greatly, for example, the threshold intensity reported in the literature for LIP in N_2 at atmospheric pressure using nano-second pulsed lasers operating at 1064 nm in a similar experimental arrangement[8,16] ranges from 6×10^{11}–3×10^{12} W/cm^2. One of the reasons responsible for the large scatter in reported threshold breakdown intensities between different experimental investigations is that the method typically used to determine this threshold depends on the subjective decision of what is considered a "regular spark event." Breakdown thresholds are functions of gas pressure, and when determining these following the traditional method, experimental errors are introduced due to changing laser properties that are typically assumed not to change. This paper discusses a different method for the determination of breakdown thresholds which can give more accurate results, particularly when determining breakdown thresholds as functions of gas pressure.

For calculating the breakdown threshold intensity, the breakdown energy, E_{th}, must be determined. The typical method of determining E_{th} is to simply increase the laser energy E_{laser} until regular spark events begin to occur. With increasing energy, the probability of a spark event at the breakdown threshold raises from zero to 100% within a certain pulse energy band, and although this band is typically small and within few millijoules, the exact threshold energy is defined by the subjective decision of the experimenter of what he considers to be a regular spark event. The energy absorbed in the plasma, E_{abs}, for a Gaussian beam is then $E_{laser}/2$.[6] For higher laser pulse energies, E_{abs} may then be written as $E_{abs} = E_{laser} - E^*_{th}/2$, where E^*_{th} is the threshold determined previously by increasing E_{laser} until regular spark events begin to occur. When measuring breakdown thresholds over a range of pressures, a change in E_{th} is observed, and it is argued in this work, that the observed change can result partially from changing E_{laser}, rather than the physics of the laser-gas interaction. This is caused by the fact that increasing the pulse energy in a laser often changes the laser behaviour in terms of transverse and longitudinal laser modes, pulse intensity distribution, pulse length, beam intensity distribution, diameter and divergence angle. A change in pulse frequency or pulse energy in a solid state laser affects the temperature of the lasing medium, and subsequently its refractive index, thereby changing the beam characteristics.[9] While it is typically assumed that these changes are negligible, the results presented here show that they can have a significant effect on the measured breakdown thresholds.

II. EXPERIMENTAL SETUP

In the example experiment presented in this paper, LIP is generated in a gas cell using a Q-switched ruby laser (JK Lasers HLS-2, wavelength $\lambda = 694$ nm), focused using a plano-convex spherical lens (made from fused silica) with a focal length of $f_l = 100$ mm. The lens is positioned "normally," with the convex surface facing the laser to yield a minimum in spherical aberration. The LIP is generated non-resonantly, there are no molecular or atomic transitions at the laser wavelength to assist breakdown[10,22] for the gases tested. The laser was operated with the oscillator aperture set to produce a 4-mm-diameter beam. The laser operates at multiple transverse and longitudinal modes as specified by

a)Electronic mail: s.brieschenk@uq.edu.au

the manufacturer. The divergence of the laser is specified as $\Phi = 0.5$ mrad. The experimental configuration is shown in Fig. 1. A beam expander was placed outside the cell to reduce the laser irradiance below the damage threshold of the window. A beam diameter of $d_b = 40$ mm behind the $\times 10$ expander yields a focusing f-number of $\approx f/2.5$. The divergence- and diffraction-limited waist diameters D_w^Φ and D_w^d, respectively, are estimated to be lower than the spherical-aberration-limited waist diameter, which calculates to $D_w^s \approx 400\ \mu$m.

$$D_w^\Phi = f_l\ \Phi \approx 50\ \mu\text{m}$$

$$D_w^d = 2.44\ \lambda\ \frac{f_l}{d_b} \approx 4\ \mu\text{m}$$

$$D_w^s = 0.067\ f_l \left(\frac{f_l}{d_b}\right)^{-3} \approx 400\ \mu\text{m}.$$

Pre- and post-focal energies were measured simultaneously for each pulse, to determine both the laser pulse energy absorbed in the LIP, E_{abs}, and the laser pulse energy E_{laser}. Following this methodology, E_{th} can be determined as a function of pressure without changing E_{laser} and therefore without changing the laser beam characteristics. A fused silica plate orientated at 45° to the beam operates as a beam splitter, with the reflected beam passing though a diffuser and neutral density filter to a photodiode that records the energy of each pulse. Transient photodiode signals were recorded using a Tektronix TDS 420 oscilloscope and have been calibrated against an Ophir 30A-P thermopile energy meter. The temporal laser intensity distribution is close to Gaussian and the laser pulse duration slightly increases with higher pulse energies, as shown in Fig. 2. At a pulse energy of 500 mJ, the pulse duration of the laser source, determined using a photodiode, measures 41 ns (FWHM). For lower pulse energies, the pulse duration reduces slightly and measures 28 ns (FWHM) for a pulse energy of 100 mJ. The breakdown threshold is an intensity threshold, I_{th}, and the breakdown energy thresholds E_{th} derived from the transient photodiode signals for laser energies between 100 and 500 mJ for this laser are similar due to slightly increasing pulse durations with higher pulse energies (Fig. 2). In order to measure the fraction of energy absorbed in the LIP, the energy meter is placed behind the focal spot during the experiment. Values for E_{laser} and E_{abs} given in this document are corrected for optical transmission and reflection losses and therefore represent the energy immediately before and immediately behind the focal spot. The transmission and reflection losses of the optical elements were determined in separate experiments. E_{th} can be significantly reduced if the optical axis of the laser is perfectly aligned with the optical axis of the focusing lens. Such an arrangement, however, is difficult to maintain and requires frequent re-alignment because even small displacements have a strong effect on the focal spot size. In the current investigation, the focusing lens was deliberately misaligned at a slight angle, approximately 1°, to minimise the effect of variations in the focal geometry over the course of the experiments. The gas cell is filled with hydrogen gas (purity 99.99%) at pressures up to $p = 30$ atm.

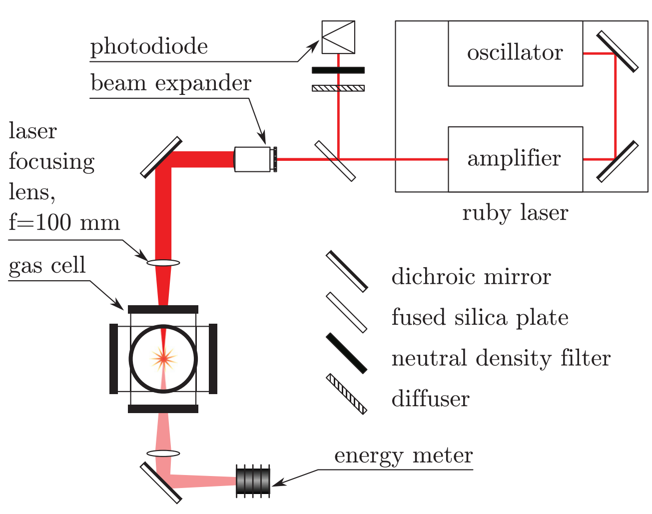

FIG. 1. Experimental arrangement for parametric study of LIP characteristics.

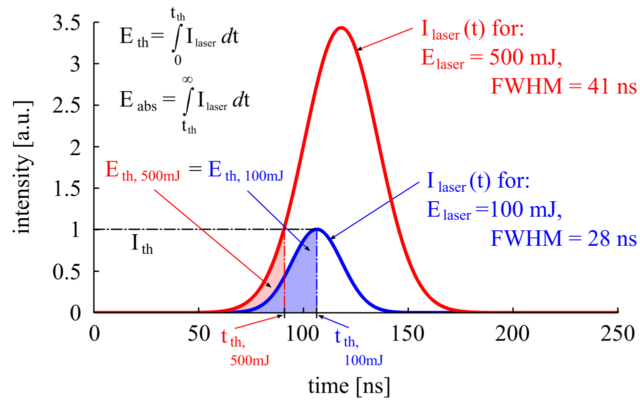

FIG. 2. Transient laser intensities for a 100 mJ and a 500 mJ laser pulse.

The key process for the formation of a LIP is the liberation of initial electrons by multiphoton ionisation (MPI).[3,7] In the electric field of the laser, these free electrons can gain enough energy through the inverse Bremsstrahlung process to ionise neutral particles by electron-impact ionisation. This results in an avalanche-like, cascade ionisation process, increasing the plasma temperature and electron density until the end of the laser pulse.[11,12] Although initial electrons are typically generated by MPI, both cascade ionisation and MPI contribute to the growth of the plasma.[15,16] Depending on the gas pressure, one of the two processes typically dominates the plasma formation process, for high pressures, cascade ionisation dominates the plasma formation process whereas for low pressures, MPI dominates the plasma formation process.[15,21] The absorption cross-sections for both the inverse Bremsstrahlung process and MPI are relatively low, but with photon pressures of the order of $p = 100$ atm in the focal spot[13] the gas becomes opaque to the laser beam. The fraction of scattered laser energy, i.e., laser photons scattered away from the plasma at the focal spot, to E_{abs} is typically negligible.[20] Incident laser photons are either transmitted through the focal region before the breakdown threshold is reached (early in the laser pulse) or are absorbed once the plasma has started to form. Experiments where we attempted to measure the energy scattered away from the plasma have confirmed this over the range of pressures and laser energies investigated in this work. The portion of the laser pulse that reaches the energy meter located behind the focal waist (Fig. 1) represents the early portion of the pulse before the MPI process initiates the plasma formation and is therefore equal to the threshold energy E_{th}. Following this methodology, E_{th} can be determined as a function of pressure without changing E_{laser} and therefore without changing laser beam characteristics. Breakdown threshold energies determined using this approach can vary significantly from breakdown thresholds determined by simply increasing E_{laser} until regular spark events occur as shown below.

III. RESULTS AND DISCUSSION

Figure 3 shows the laser pulse energy absorbed in the LIP over a range of pressures at a laser energy of $E_{laser} \approx 480\,\text{mJ} \pm 2\%$ in hydrogen. For a laser with a symmetric temporal profile, the portion transmitted through the focal region is equal to the portion contributing to the LIP when operating at threshold breakdown conditions. Once lasing is initiated in the laser, the photon flux density increases until the maximum irradiance is reached, where half of the photons have been lost through the focal waist.[6] At maximum laser irradiance, the intensity threshold is reached and MPI renders the focal waist opaque to the laser beam due to the generation of free electrons and hence, the second half of the photon flux is contributing to the LIP formation. More intense laser radiation allows the plasma to form earlier in the pulse history and a greater proportion of E_{laser} can thus be absorbed. Figure 4 shows the energy absorbed in the plasma as a function of pressure for various laser pulse energies. These data clearly show that the energy absorbed in the plasma, and therefore the breakdown threshold energy and intensity, strongly depend on the laser pulse energy setting. For example, at a gas pressure of p = 2 bar, a pulse energy of $E_{laser} = E^*_{th} = 80$–$100\,\text{mJ}$ is required to observe regular spark events. The probability of regular spark events is 100% for laser energies above 100 mJ, and sparks begin to form infrequently for a laser energy of 80 mJ. At the breakdown threshold, half the energy, here 40–50 mJ, is absorbed in the plasma, and the pre- and post-focal energies recorded in the experiment confirm this for laser pulse energies up to 130 mJ. Higher laser energies change the properties of the laser beam, and the threshold energy E_{th} transmitted through the focal region significantly increases from 40–50 mJ to 150 mJ for a laser pulse energy of $E_{laser} = 470\,\text{mJ}$. For LIP where $E_{laser} > E_{th}$, the energy transmitted through the focal spot $E^*_{th}/2$ following the traditional method, yields values that differ by a factor of up to three compared to those found by measuring pre- and post-focal energies. It is clear from these measurements, that the dependence between threshold energies and gas pressure cannot be accurately measured when following the traditional method of determining breakdown thresholds by changing the laser pulse energy. The laser pulse energy must be kept constant for the laser properties to remain unchanged throughout the experiment, and the measurements to be accurate. The threshold energy curves for 130, 270, and 470 mJ in Figure 4 are, as expected, similar to each other and their shape is solely due to the physical dependence between the laser-gas interaction and the gas pressure.

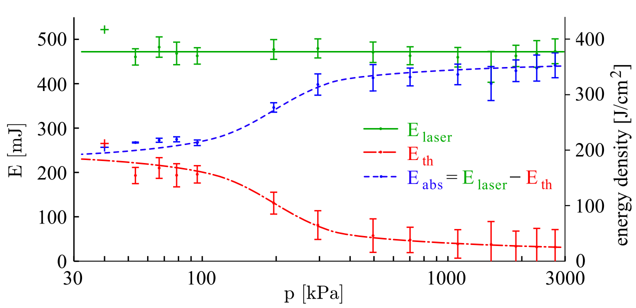

FIG. 3. Laser energy absorbed in LIP in hydrogen gas. Data points with error bars represent the average of nine individual measurements and their standard deviation.

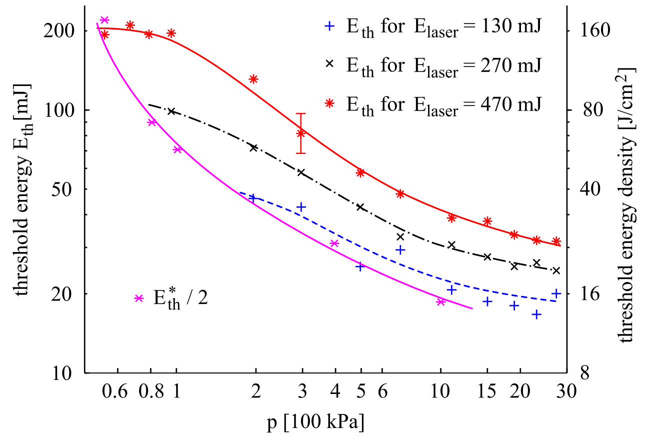

FIG. 4. E_{th} as a function of gas pressure for different laser pulse energies determined by measuring post-focal energies compared to the threshold laser energy $E^*_{th}/2$ using the traditional approach of increasing the laser energy until regular sparks occur to form. Measurement standard deviations indicated at the data point at 300 kPa and 470 mJ.

It should also be noted that a common approach[1,16] to measuring the wavelength dependence of the breakdown threshold is to use the fundamental wavelength of a laser source and compare the measured thresholds to experiments where the second or third harmonic frequency of the laser is used. Following this methodology, however, is difficult, since non-linear optical processes such as frequency doubling or sum-frequency generation typically result in temporally and spatially distorted laser beam profiles. This can go as far as the incident pulse being split into two, virtually separate pulses and a spatial intensity distribution far from a Gaussian shape, as has been reported in the literature before.[14] A solution for comparing breakdown thresholds at different wavelengths would be using a dye laser, where the wavelength can be changed without changes in the temporal and spatial beam profiles.

IV. CONCLUSIONS

The presented work has shown that significant errors can arise when determining laser breakdown thresholds as functions of gas pressure, by the traditional method of adjusting laser pulse energies until regular spark events begin to occur. Changing a single laser property, such as pulse energy, or frequency by harmonic generation, can have a significant effect on secondary parameters that are typically assumed not to change, causing significant errors in the measurement.

For the experiments presented in this paper, the energy transmitted through the focal spot $E^*_{th}/2$ following the traditional method, where the laser pulse energy is increased until regular spark events begin to occur, yields values that differ by a factor of up to three compared to those found by measuring pre- and post-focal energies, where the laser pulse energy has been held constant. The presented data have shown that determining breakdown thresholds by measuring pre- and post-focal energies can give more accurate results, as the laser pulse energy, and therefore the laser beam characteristics, do not change when generating the LIP at different gas pressures. The laser is set to a fixed pulse energy and the only parameter that changes in the experiment is the pressure in the gas cell.

A series of proper measurements need to be conducted to ensure that changing one property of the laser does not affect any other property of the laser output if the traditional method of determining breakdown thresholds is to be used. For the ruby laser used in this study, the laser pulse energy setting had a significant influence on the beam characteristics, introducing significant errors when the traditional method of determining breakdown thresholds versus gas pressure was used.

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