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Comparison of the electron density measurements using Thomson scattering and emission spectroscopy for laser induced breakdown in one atmosphere of helium

E. Nedanovska,^{1,a)} G. Nersisyan,¹ T. J. Morgan,² L. Hüwel,² C. L. S. Lewis,¹ D. Riley,¹ and W. G. Graham¹

¹Department of Physics and Astronomy, Centre for Plasma Physics, Queen's University Belfast, BT71NN, United Kingdom

²Department of Physics, Wesleyan University, Middletown, Connecticut 06459, USA

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Thomson scattering from laser-induced plasma in atmospheric helium was used to obtain temporally and spatially resolved electron temperature and density profiles. Electron density measurements at $5\ \mu\text{s}$ after breakdown are compared with those derived from the separation of the allowed and forbidden components of the 447.1 nm He I line. Plasma is created using 9 ns, 140 mJ pulses from Nd:YAG laser at 1064 nm. Electron densities of $\sim 5 \times 10^{16}\ \text{cm}^{-3}$ are in good agreement with Thomson scattering measurements, benchmarking this emission line as a useful diagnostic for high density plasmas. © 2011 American Institute of Physics. [doi:10.1063/1.3672817]

Laser produced plasmas have found applications in a variety of circumstances, including fast triggering in gaseous environments¹ and elemental analysis of solid and liquid samples.² In the latter context, the role of the ambient gas in general and that of helium in particular^{3–5} as well as gas dynamic effects⁶ have been the focus of recent studies. The purpose of the current work is to expand on recent experiments⁷ and to demonstrate with the help of Thomson scattering (TS) that certain emission based diagnostic techniques used in these applications are reliable and set the stage for progressing into the low electron density regime ($10^{15}\ \text{cm}^{-3}$ and below) characteristic of micro-plasmas. Results for the electron density presented in the current paper test the reliability of commonly applied spectroscopic methods, in particular the HeI line at 447.1 nm which can be used to determine electron density by monitoring the separation and intensity ratio of the allowed ($2p\ ^3P_0 - 4d\ ^3D$) and forbidden ($2p\ ^3P_0 - 4f\ ^3F_0$) components. Several authors have published equations relating the electron density to the spectral separation and intensity ratio of the allowed and forbidden components of this particular line.^{8–10} It is important that such a diagnostic is tested against an independent measurement. This has been done in discharges for the 447.1 nm line, for example, using interferometry for densities between $(1.9\text{--}5.9) \times 10^{15}\ \text{cm}^{-3}$ (Ref. 11) and between $(1\text{--}7) \times 10^{17}\ \text{cm}^{-3}$.⁹ Both experiments were performed without spatial resolution along the axis of low pressure pulsed discharges. In the current work, we have employed Thomson scattering to obtain both temporally (20 ns window) and spatially (6.9 mm) resolved electron temperature and electron density profiles to make a direct comparison with electron density measurements derived from emission spectroscopy (ES). In particular, we present an assessment of the HeI 447.1 nm methods for the intermediate range of $(1\text{--}5) \times 10^{16}\ \text{cm}^{-3}$ in atmospheric pressure laser produced plasmas.

Our experimental arrangement is shown schematically in Fig. 1. A vacuum chamber is evacuated to approximately 10^{-4} mbar and backfilled to 1 atm with He. Using a 10 cm focal length lens, a 9 ns 140 mJ pulse from an Nd:YAG laser (GCR 4) with an initial spot diameter of ~ 9 mm is focused into the chamber centre by a lens of $f/12.5$. The baffle arms are fitted with Brewster angle windows to reduce stray light scattering. Light emitted from the induced plasma at 90° to the direction of the breakdown beam is collected with an $f/5$ lens, and a second lens is used to image the plasma with unity magnification onto the slit of an imaging double grating spectrometer (SPEX 750, with 1200 l/mm gratings providing $5.7\ \text{\AA}/\text{mm}$ at the detector plane). The diverging beam, transmitted through the helium, exits the chamber via a BK7 window and into a light trap. The spectrometer provides spatial resolution in the direction perpendicular to the direction of the breakdown beam (lateral direction). The $100\ \mu\text{m}$ slit provides effective spatial resolution along the axial direction. For Thomson scattering, a separate laser is used (7 ns, 80 mJ at 532 nm). This beam is directed perpendicular to the

FIG. 1. (Color online) Schematic of set-up for Thomson scattering.

^{a)} Author to whom correspondence should be addressed. Electronic mail: enedanovska01@qub.ac.uk.

FIG. 2. (Color online) Typical data for $5 \mu\text{s}$ delay: (a) Thomson scattering and (b) emission spectroscopy. The upshifted, blue wing of the Thomson scattering is fitted to the Salpeter approximation. The image insets show the spatial region over which the line-outs are averaged.

breakdown pulse, and so the Thomson scattered signal on the spectrometer slit provides lateral resolution. The probe pulse is loosely focused with a full width half maximum of approximately $400 \mu\text{m}$ in order to avoid heating by the probe. The spectrometer was fitted with an intensified CCD camera that was gated to 20 ns for both Thomson scattering and emission spectroscopy. The two lasers and the CCD were synchronized using a Stanford Delay Generator (SDG). The system was calibrated by filling the chamber with 10 mbar of Argon and measuring the Rayleigh intensity.¹²

In Fig. 2(a), we show typical Thomson scattering data and a spectral line-out of the central 0.54 mm of the breakdown at $5 \mu\text{s}$ delay. In Fig. 2(b), a sample emission spectrum of the 447.1 nm line for the same conditions is shown. As can be seen, at this time, the Thomson scattering is in the collective regime which manifests with a plasmon feature in the spectrum, and fitting to the Salpeter approximation¹³ yields both density and temperature.

By $5 \mu\text{s}$, the plasma has expanded up to ≈ 4.3 mm in the direction parallel to the probing laser beam (y axis); later in time, it develops a “torus” structure probably as a result of radially propagating shock formation,¹⁴ and the Thomson scattering moves into non-collective mode. Here, we use the lowest electron density where the plasma is uniform, at the early time of $5 \mu\text{s}$, to compare emission and scatter data. In

FIG. 3. (Color online) Electron density and temperature lateral profile (y axis) for $5 \mu\text{s}$ delay from the Thomson scattering signal. The uncertainties for the density and temperature are within 13% and 10%, respectively, for $|y| < 1.5$ mm and within 22% and 20% for $|y| > 1.5$ mm. The experimental data have reproducibility of 95%.

Fig. 3, we show the lateral profiles of the electron temperature and density at this time delay deduced from the Thomson scattering signal, and the uniformity of the plasma is evident, apart from the elevated electron temperature at the extreme edges. The latter is possibly an artifact from the initial blast wave.¹⁴

The emission spectra, although resolved spatially in the direction of the probe laser, originate from a line of sight through the plasma, and Abel inversion is needed. By assuming cylindrical symmetry of the induced plasma, we can Abel invert the signal to obtain the radial intensity profile. For the Thomson scattering, Abel inversion is not needed since we do not scatter through a line of sight through the plasma but from a limited region defined by the 0.4 mm probe beam diameter, which is small compared to the ≈ 4.3 mm diameter of the plasma at this stage of its evolution. Aside from the cylindrical symmetry of the plasmas, the other main assumption for the Abel inversion is low optical density of the plasma. A rough analysis of the Rayleigh peak, at the central 0.54 mm of the plasma region, assuming only the presence of He neutral atoms and HeII in the plasma region, yields He atom density of the order of $4.8 \times 10^{18} \text{ cm}^{-3}$. This value enables us to set an upper limit on the opacity at $\tau = 0.1$. In He plasmas,¹⁵ He metastable ($\text{He}(2^3\text{S})$), positively charged molecular helium ions (He_2^+), as well as excited molecular helium (He_2^*) are being created. The presence of the aforementioned species can only lead to lowering the density of He atoms which in turn results in lower opacity of the plasma.

In Fig. 4(a), we show the radially resolved profiles of electron density inferred from the emission data and from Thomson scattering. We have used three different formulae to determine the density from the separation of the allowed and forbidden lines, i.e., those of Perez *et al.*,⁸ Ivković *et al.*,⁹ and Czernichowski and Chapelle¹⁰ and one formula that uses the forbidden to allowed line ratio (F/A).¹⁰ We have used two separate Abel inversion routines, a numerical integration¹⁶ and the Nestor Olsen approach,¹⁷ and get consistent results. The degree of agreement of the electron density data from the emission spectroscopy methods with the

FIG. 4. (Color online) (a) Comparison of electron density, at different radial positions r , from the 447 nm line forbidden-allowed separation methods and intensity ratio (F/A) method with TS data. (b) Ratio of the electron densities from the ES approaches and the TS, after performing Abel inversion. The dashed line represents the Thomson scattering data (TS/TS).

Thomson scattering measurements, in radial direction r , is shown in Fig. 4(b). We can see that the empirical formula by Czernichowski and Chapelle¹⁰ for the forbidden-allowed separation, in particular, agrees very well with the Thomson scattering data. Thus, this emission methodology, for the 447 nm line at atmospheric pressure, can be used in the density region of $1\text{--}5 \times 10^{16} \text{ cm}^{-3}$ with 30% confidence, and consequently, we have expanded the prescribed validity range in their paper to higher electron densities. The 447 nm line shapes before and after performing Abel inversion are identical, confirming its homogeneous profile.

To conclude, in the electron density range of $(1\text{--}5) \times 10^{16} \text{ cm}^{-3}$, we have used optical Thomson scattering as an independent check on the validity of three formulae that give electron density from the allowed-forbidden component separation in the 447 nm line of helium and one that is based on the intensity ratio of the forbidden and allowed line. We find in the present electron density regime that there is a better agreement with the line separation than the line ratio method, with the best agreement from the Czernichowski and Chapelle approach. This is an important check as there are many plasma types where diagnostics other than emission spectroscopy are difficult to implement. The open geometry used in our experiment has allowed us to make the comparison. We are now in a position to compare the various emission based techniques with the Thomson scattering in laser-initiated plasma at later times, in the presence of other gases and at electron density regimes believed to be present in electrically produced microplasmas.

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