Hydrogen Alpha Self-Absorption Effects in Laser-Induced Air Plasma

C G Parigger, L D Swafford, D M Surmick, M J Witte, A C Woods, and G Gautam
University of Tennessee Space Institute, 411 B.H. Goethert Parkway, Tullahoma, TN 37388, USA
E-mail: cparigge@tennessee.edu

Abstract. Time-resolved spectroscopy measurements of the hydrogen alpha Balmer series line following laser-induced optical breakdown in laboratory air are designed to investigate in detail the determination of electron density from Stark-broadened spectral line shapes. Comparisons of results obtained from H_β and H_γ lines indicate higher electron density inferred from H_α early in the plasma decay, suggesting self-absorption occurs. However, detailed comparisons for time delays of 300 and 400 ns after optical breakdown reveal the minute extent of self-absorption in air breakdown experiments from (i) differences of electron density determined from the N^+ lines and the H_α line, and/or from (ii) differences in recorded data sets with/without the mirror for the various time delays in the experiments.

1. Introduction
In this work, we systematically vary the time delay from 100 - 1000 ns, and employ a gate width of 10 ns. Optical breakdown is generated by focusing pulsed Nd:YAG laser radiation parallel to the spectrometer slit using f/# = 4 optics. The emission from the decaying plasma is spatially and temporally resolved with a Czerny-Turner spectrometer and an ICCD camera [1, 2]. Rather than using a spherical mirror [3], we utilize a flat mirror and a plano-convex lens in the self-absorption studies. Typical results for the full-width half-maxima for H_α and the two N^+ lines on either side of the H_α line lead to an electron density of $\approx 10 \times 10^{23}$ m$^{-3}$, indicating hardly any H_α self-absorption.

The self-absorption analyses include comparisons of recorded data with and without the duplicating mirror for both the continuum radiation and the measured plasma emissions of atomic lines above the continuum [3]. This approach works well for line-of-sight measurements, for homogeneous distributions, and in optically thin conditions [3, 4, 5]. For optically-thick conditions, the observed line will no longer be linearly related to the absorption coefficients. Determination of the electron density from ionized nitrogen lines [6] and comparison with the inferred electron density from hydrogen alpha emissions will allow us to investigate the level of self-absorption as well. In previous studies, H_α self-absorption was indicated by comparisons with H_β and H_γ measurements [1].
2. Experiment
Measurements of spatially and temporally resolved spectra with/without mirror are reported here. The laser-induced plasma is generated by focusing the radiation from the top. The emission spectra are captured using a 10 ns gate width in steps of 100 ns from the optical breakdown events. A set of 276 spectra was recorded along the 7.15 mm slit height that was utilized for this experiment. The recorded data from 81 consecutive breakdown events in air are portrayed as images of wavelength versus slit height.

3. Results
In the recorded spectra, hydrogen alpha emissions begin to emerge for time delays of 300 ns, and are clearly recognizable for the explored range of 400 to 1000 ns. For time delays of 100 ns to 700 ns, ionized nitrogen lines are clearly recognizable at 648.2 nm and 661.1 nm, i.e., at the low and high wavelength side of the 656.28 nm H$_\alpha$ line.

The addition of a mirror allows us to investigate self-absorption effects of the line-of-sight emission spectroscopy diagnostic. Optical radiation from the plasma is retro-reflected and imaged as well onto the spectrometer slit. Detailed comparisons are aimed to reveal the level of self-absorption due to (i) differences of electron density determined from the N$^+$ lines and the H$_\alpha$ line, and/or due to (ii) differences in recorded data sets with/without the mirror for the various time delays in the experiments.

Addition of a mirror and subsequent comparisons of continuum radiation and of recorded emission lines is a well-accepted method for determination of the level of self-absorption [3]. The experimental factor, $K_{\lambda,\text{corr}}$, to correct for self absorption [3, 4, 5] in the optically-thin case, can be evaluated from the data independent of the detector’s sensitivity. The ratio of the continuum radiation, $R_C$, is determined from data with and without the mirror. The wavelength dependent ratio, $R_\lambda$, is obtained as ratio of signal above the continuum for data w/ and w/o the mirror. The self-absorption correction factors at line centers of the two N$^+$ lines amount to $\simeq$ 1.0 for the 300 ns data sets. At a time delay of 300 ns, the hydrogen alpha line is too broad for application of the correction factor method; therefore, comparisons of electron densities determinend from N$^+$ and H$_\alpha$ widths is discussed in the next sections. For a time delay of 800 ns, Equation (1) is used to evaluate $K_{\lambda,\text{corr}}$ for H$_\alpha$. Figure 1 shows the recorded, filtered data w/ and w/o the mirror. The illustrated data have been detector-background subtracted, sensitivity corrected and wavelength calibrated. The line center correction factors amount to $\simeq$ 1. Self-absorption of H$_\alpha$ is insignificant for 800 ns time delay.

![Figure 1](image)

**Figure 1.** Spectra w/ and w/o mirror (Left) time delay 800 ns, slit height: 5.15 mm; (Right) time delay 300 ns, slit height: 6.15 mm. Smoothed line shapes are Savitzky-Golay filtered.
Measurements of electron density from ionized nitrogen lines utilize published tables for Stark widths and shifts [6]. In previous analysis we utilized the N\(^+\) 399.5 line [7], here we use the two N\(^+\) lines at 648.2 nm and 661.1 nm. Figure 1 shows spectra at slit heights of 6.15 mm. For the 648.2 nm line, the width is 0.86 nm, leading to an electron density of \(12.7 \times 10^{23} \text{m}^{-3}\). For the 661.1 nm line, the width is 0.98 nm, leading to an electron density of \(12.1 \times 10^{23} \text{m}^{-3}\). The accuracies of the electron densities from the N\(^+\) lines amount to 30% [6].

Electron density from Stark widths and shifts of the hydrogen alpha Balmer series line can be inferred using Stark width tables [8], results from convergent theory [9, 10] and empirical formulae [11]. Comparisons of the accuracy of H\(_\alpha\) and of H\(_\beta\) fitting shows it is generally better to use H\(_\beta\) lines [12]; yet for measurements of electron density larger than \(7 \times 10^{23} \text{m}^{-3}\) the H\(_\beta\) line cannot be used [10]. For optical breakdown in air, this implies that H\(_\beta\) diagnostic will be useful for time delays larger than \(1.4 \mu \text{s}\). Figure 1 also shows broad H\(_\alpha\) emission spectra. The inferred widths from the broad profiles are 8 to 10 nm, the shifts are 0.8 to 1.0 nm. H\(_\alpha\) widths of 9 nm and H\(_\alpha\) shifts of 1 nm imply electron densities of \(14 – 20 \times 10^{23} \text{m}^{-3}\) and \(10 – 20 \times 10^{23} \text{m}^{-3}\) [10]. In turn, the N\(^+\) line widths show electron densities of \(12 – 13 \times 10^{23} \text{m}^{-3}\), suggesting significant self-absorption of H\(_\alpha\) occurs for time delays on the order of 300 ns.

4. Conclusions
Detailed investigation and comparisons of time-resolved spectroscopy measurements of the N\(^+\) and H\(_\alpha\) lines allow one to determine the hydrogen alpha self-absorption effects in Laser-induced air plasma. The N\(^+\) lines yield smaller electron densities than the H\(_\alpha\) line for early time delays, yet the differences diminish for data that is averaged along the spectrometer’s slit dimension. Results show differences in electron density along the plasma with noticeable H\(_\alpha\) red-shifts for early time delays. There are indications that significant self-absorption of H\(_\alpha\) indeed occurs for a time delay of 300 ns from optical breakdown, for 400 ns hardly any self-absorption is evident, and for 800 ns time delay self-absorption effects are insignificant for optical breakdown in laboratory air. From the recorded data sets, one can infer plasma expansion speed on the order of 300 to 100 \(\mu \text{m}\) in 100 ns or 3 to 1 km/s, in the range of 300 to 1000 ns after optical breakdown.

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References
[1] Parigger C G 2010 Int. Rev. At. Mol. Phys. 1 129
[2] Parigger C G 2013 Spectrochim. Acta Part B 79-80 4
[3] Moon H, Herrera K K, Omenetto N, Smith B W, Winefordner J D 2009 Spectrochim. Acta Part B 64 702
[4] Konjević N 1999 Phys. Rep. 316 339
[5] El Sherbini A M, El Sherbini Th M, Hegazy H, Cristoforetti G, Legnaoli S, Palleschi V, Pardini L, Salvetti A, Tognoni E 2005 Spectrochim. Acta Part B 60 1573
[6] Konjević N, Lesage A, Fuhr J R, Wiese W L 2002 J. Phys. Chem. Ref. Data 31 819
[7] Parigger C G, Woods A C, Surmick D M, Swafford L D, Witte M J 2014 Spectrochim. Acta Part B 99 15
[8] Griem HR. 1964 Plasma Spectroscopy, McGraw-Hill, New York 1964
[9] Oks E. 2006 18th ICSLS: AIP Conf. Proceedings 874 19
[10] Parigger CG, Plemmons DH, Oks E. 2003 Appl. Opt. 42 5992
[11] Konjević N, Ivković M, Sakan N. 2012 Spectrochim. Acta Part B 76 593
[12] Parigger C G, Swafford L D, Woods A C, Surmick D M, Witte M J 2014 Spectrochim. Acta Part B 99 28