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Laser plasma diagnostics and self-absorption measurements of the H_β Balmer series line

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ABSTRACT

In this work, the peak-separation of the Balmer series hydrogen beta line was measured to determine the electron density of laser-induced plasma from spatially and temporally resolved spectra collected in laboratory air at standard ambient temperature and pressure. The self-absorption phenomenon is investigated by using a mirror that retro-reflects the emitted radiation through the plasma. The experimental data with and without the mirror were analyzed with available hydrogen beta computer simulations. Hardly any self-absorption was found as indicated by the correction factors that only marginally differ from unity. The obtained electron density values are also compared with the electron densities from nearby nitrogen lines. The hydrogen beta H_β peak-separation method yields reliable results for an electron density of the order of 1×10^{17} cm^{-3} for time delays of 5 μs from plasma generation, which confirms that self-absorption is insignificant for such electron densities.

1. Introduction

Laser-induced breakdown spectroscopy (LIBS) diagnostics in gases are of general interest in atomic emission spectroscopy experiments. Hydrogen Balmer series atomic lines like H_α and H_β are frequently used to determine the plasma electron density, N_e. Measurement of Stark broadening allows one to determine the electron density and this diagnosis has been employed by many researchers [1–9]. Stark-broadened atomic emission profiles for hydrogen beta are mostly utilized to determine characteristics of the transient plasma for specific time delays; however, it is desirable to evaluate the extent of potential self-absorption of atomic emission profiles. When radiation propagates through the plasma active volume, self-absorption or line-reversal effects may occur and cause the profile to appear broader and/or possibly show a spectral dip at the central emitted wavelength. In this work, an additional mirror and lens are used to determine the extent of plasma self-absorption [10–14]. Previous studies of optical breakdown in laboratory air [10–13] indicated that there is a significant amount of self-absorption at a time delay of 0.3 μs, at which electron densities are $N_e \geq 30 \times 10^{17}$ cm^{-3}, determined from the H_α line and by comparisons with N^+ lines. For electron densities in the range from 1 to 10×10^{17} cm^{-3}, the use of the H_β line is preferred for plasma diagnostics [15,16].

Ionization of air species occurs after heating by nanosecond laser pulses and, subsequently, free electron radiation and atomic lines of hydrogen, nitrogen, and

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oxygen dominate the emission spectra. Electron densities between 10^{15} cm^{-3} and 10^{18} cm^{-3} [15–19] are usually determined from analysis of the hydrogen Balmer series beta line. The H_β peak-separation method [20,21] can also be applied to accurately determine the electron density and, equally, serve as H_β self-absorption test. The application of the Balmer series encompasses methods that rely on spectral line shape parameters and/or the evaluation of the peak separation of the beta line, with line center at 486.14 nm. Data are collected from the integration of radiation along the line of sight.

Self-absorption due to the plasma will be discussed primarily for the hydrogen Balmer series beta line. In the assessment of self-absorption, the radiation emitted from the side opposite to the spectrometer is retro-reflected from a mirror to pass back through the plasma prior to imaging onto the spectrometer slit. For time delays smaller than 1.4 μs, the H_β has not yet emerged from the background radiation and typically the H_α line is utilized for diagnostic purposes [12]. Effects of the hydrogen Balmer series beta line due to self-absorption are of interest; however, the assessment method for the H_β line is identical to the one applied for the H_α line [10]. The extent of self-absorption is determined from the intensity profile without and with the doubling mirror. The correction factor is evaluated by using the equation,

$$K_\lambda = \frac{\ln\{y_\lambda\}}{y_\lambda - 1}, \quad \text{with } y_\lambda = \frac{R_\lambda - 1}{R_C - 1}. \tag{1}$$

The continuum radiation ratio, R_C, and the signal ratio above the continuum, R_λ, are obtained from the recorded data with and without the mirror. The value for the continuum ratio is determined after fitting symmetric, computer simulated profiles to the recorded data. Ideally, the R_C value would be determined approximately 5× FWHM from the line, but we extracted this ratio approximately 1.5× FWHM from the H_β center. The signal ratio R_λ is evaluated from the fitted profiles which diminishes excursions due to noise superimposed to the data.

2. Experimental details

In the experimental arrangement for the optical breakdown in the laboratory air, a Q-switched Nd:YAG laser is operated at the fundamental wavelength of 1064 nm and at a repetition rate of 10 Hz generating 13 ns, 190 mJ pulses. Fig. 1 illustrates the general schematic for air breakdown studies.

The residual component of 532 nm is removed by passing the beam through a dichroic beam splitter and 1064 nm is used for the optical breakdown. Three mirrors are used to align the beam parallel to the spectrometer slit. A laser-line glass lens coated for the 1064 nm IR radiation is used to focus the beam. The micro-plasma is imaged onto the spectrometer slit by two uncoated, fused silica lenses. The lens for focusing onto the slit is positioned to match the spectrometer's f-number of 5.2. The laser beam direction is parallel to the spectrometer slit yet the line of sight is perpendicular to the direction of the incident laser radiation. A plane mirror, referred to as the doubling mirror, of 90% reflectance is used to retro-reflect the radiation from the plasma for the purpose of self-absorption studies. The plasma radiation passes twice through the fused silica lens of 94% transmittance, positioned in front of the doubling mirror, as indicated in the photograph displayed in Fig. 1. Furthermore, some minor losses can occur due to scattering from the mirror surface. Therefore, the experimental signal for the mirror-case will not be double the no-mirror-case, as it would be expected in an ideal condition.

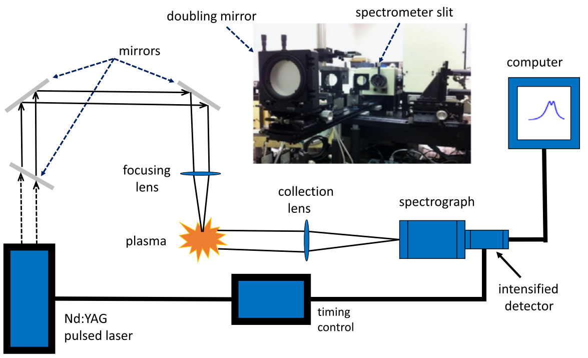

Fig. 1. Experimental schematic for laser-induced breakdown spectroscopy with the Nd:YAG laser source and its component. Photograph of the arrangement for self-absorption studies with the doubling-mirror in the foreground (Middle–top).

For the experiments, a 1800 grooves/mm grating is selected to disperse the radiation from the plasma for various time-resolved spectroscopic studies with the 0.64 m Jobin-Yvon spectrometer, resulting in a spectrometer-detector system resolution of 0.10 nm. Temporally resolved hydrogen Balmer series emission profiles are measured by varying the time delay from optical breakdown. For the recording of spatially resolved images along the slit height, a 2-dimensional intensified charge-coupled device (ICCD; Andor technology model iStar 334 T) is used [22]. Spectral measurements of radiation from the plasma are spatially resolved by grouping the vertical lines of the ICCD. For the experiments reported here, a grouping of 8 pixels was selected resulting in 0.109 mm zones, and with a 1.05:1 imaging an effective plasma zone resolution of 0.114 mm could be achieved. Each of these zones is in principle described by different plasma parameters like electron density, electron temperature and ion temperature. We note that an implementation of an Abel inversion would certainly allow the determination of the plasma distribution along the line-of-sight for an optically thin and cylindrically symmetric plasma.

3. Results

Fig. 2 displays the recorded images of optical breakdown and plasma formation with H_β and N^+ lines at a 5 μs time delay without and with the doubling mirror. Optical breakdown is generated by focusing the laser radiation from the top, in the image. Comparisons of the two images indicate higher signals for the recorded data with the mirror in place. For completely transparent