



## Research Note

Diode laser absorption measurements at the  $H_{\alpha}$ -transition in laser induced plasmas on different targets

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## ABSTRACT

The diode laser atomic absorption spectroscopy (DLAAS) technique has been utilized to assess the degree of optical opacity of plasma at the wavelength of the  $H_{\alpha}$ -line. The plasma is produced at atmospheric conditions by focusing a 6 ns Nd:YAG laser pulse at 1.064  $\mu\text{m}$  on different solid target materials including aluminum, iron and titanium as major elements as well as flat pieces of plastic and wood characterized by a high content of hydrogen. The optical depth was investigated as a function of delay times ranging from 0 to 5  $\mu\text{s}$ , and at laser fluences ranging from 7 to 19  $\text{J}/\text{cm}^2$ , all at a fixed gate time of 1  $\mu\text{s}$ . The results show that the plasma associated with metallic targets is almost optically thin at the  $H_{\alpha}$ -line over all fluences and at delay times  $\geq 1 \mu\text{s}$ , but rather thick for hydrogen-rich targets (plastic and wood) over all delay times and fluences.

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## 1. Introduction

Laser induced breakdown emission spectroscopy (LIB) proved to be a powerful tool in spectrochemical elemental analysis. Plasma parameter measurements make use of well defined optically thin spectral lines of excited neutral and ionic species. One promising line often appearing in the plasma produced by laser irradiation of solid targets in atmospheric conditions is the hydrogen  $H_{\alpha}$ -line at 656.27 nm [1–8]. In an earlier publication it was suggested that the optically thin  $H_{\alpha}$ -line can be used as a reference measure of the electron density in the plasma [9]. Moreover, it assisted in evaluating the optical depth  $\tau$  through the coefficient of self-absorption (SA) of lines that appear in the emission spectrum under the same experimental conditions and are suspected to be optically thick [1]. However, in all the published works [1–9], that make use of the assumption of negligible optical depth of the plasma to the  $H_{\alpha}$ -line there was no direct measurement that supported this assumption.

The process of re-absorption of radiation exists for every system capable of emitting radiation, predominantly strong with resonance transition lines as well as for intense ones [1]. This was attributed to the existence of a relatively large concentration of atoms in lower states often located at the colder regions of plasma where the atomic absorption is the prevailing process [3,4]. Moreover, the appearance of the strong continuum in LIBS experiments especially at the early delay times or very near to the target surface cannot be neglected in the process of re-absorption. Inversebremsstrahlung (free–free transi-

tions) mechanism and/or photo-ionization (bound–free transitions) might play an important role at the early time of plasma evolution [10]. The combination of these absorption processes leads, in general, to a nonlinear relationship between the measured line intensity and element concentration (curve of growth COG), as well as an enlargement to the measured line width (FWHM). The overestimated line broadening yields serious errors in the measured plasma electron density.

Several methods were suggested to quantify the amount of absorption via different techniques and the reader is referred to the review by Konjevic [11]. A precise and plasma state independent method was recently verified by H.-Y. Moon et al. [12] in which the method of a duplicating mirror was used to measure and correct against the amount of re-absorption by duplicating the path travelled by the light from the plasma. This method yielded a precise and simultaneous measurement of the absorption as well as temporal evaluation of the optical depth from the early delay to the end of the plasma, allowing outliers in the Boltzmann plot to be eliminated and to improve the linearity of the calibration curve [12]. Another technique based on imaging absorption was utilized by Bushaw et al., [13] in order to monitor the space resolved atomic absorption.

On the other hand, the diode laser atomic absorption spectroscopy (DLAAS) technique was utilized to probe the plasma in a variety of applications [14–27]. The high intensity of the laser light is used to overcome the luminosity of the plasma, while the monochromaticity can provide a high selectivity for exciting a definite transition and the narrow bandwidth of the laser provides a high spectral resolution [14].

Unfortunately, not too much work was done at the wavelength corresponding to the hydrogen  $H_{\alpha}$ -line. The ratio of different

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elements (C, H, and Cl) as a function of plasma conditions at the wavelength corresponding to the  $H_{\alpha}$ -line was measured by Koch et al. [28].

In this paper, we report the results of our attempt to employ the diode laser atomic absorption spectrometry (DLAAS) in a direct measurement of the plasma optical depth at the wavelength corresponding to the  $H_{\alpha}$ -line under different experimental conditions.

## 2. DLAAS-technique

In the presence of an external probe light source (e.g. laser diode of sufficiently low power), the solution of the radiative transfer equation can be written in the following form [29–31];

$$I_{LP}(0, \lambda_o) = I_L(\lambda_o) + I_P(\lambda_o) \quad (1)$$

where  $I_{LP}(0, \lambda_o)$  is the spectral (specific) intensity of plasma in the presence of the external light source at the specific wavelength ( $\lambda_o$ ) corresponding to the central emission of the  $H_{\alpha}$ -line. The spectral intensity of the diode laser light after passing through the plasma is given by the Lambert relation,

$$I_L(\lambda_o) = I_{oL}(\lambda_o) \exp(-\tau) \quad (2)$$

$\tau(\lambda_o) = -\int_0^L \kappa(x, \lambda_o) dx$  is the optical depth of the plasma at the wavelength corresponding to the  $H_{\alpha}$ -line. Thus, Eq. (1) can be rearranged to yield,

$$\tau(\lambda_o) = \text{Ln} \left[ \frac{I_{oL}(\lambda_o)}{I_{LP}(\lambda_o) - I_P(\lambda_o)} \right] \quad (3)$$

Expression (3) shows that the optical depth can be measured in terms of the spectral intensity of the  $H_{\alpha}$ -line emission of the probe diode laser, the plasma as well as the attenuated diode laser beam intensity, i.e. the optical thickness on resonance which is the maximum optical thickness that can be achieved for an  $H_{\alpha}$ -transition and it is independent of the frequency of the probe light source used [32].

## 3. Experimental setup

The experimental setup is shown in Fig. 1. A Nd-YAG laser (type Quantel, model Brilliant-B) was used at a fundamental wavelength of 1.06  $\mu\text{m}$  and a FWHM of 6 ns duration. The laser energy delivered to the target was varied in the range from 210 to 610 mJ using a set of glass sheet absorbers and was monitored with the help of a 6.3% reflection glass beam splitter by a power-meter (type Ophier, model 1z02165). The target was positioned on an xy-stage at a distance of 9.5 cm from the 10 cm focal length laser focusing lens. The 2 mm diameter circular laser spot area was estimated using a thermal paper and from the burn marks (crater) on the different targets surfaces. This enabled us to calculate the fluences at the target surface, which were in the range from 7 to 19 J/cm<sup>2</sup>. The light from the plasma plume was collected by an optical fiber and analyzed by an echelle type spectrograph (type Catalina, model SE200) with the VIS-UV module of resolving power of 2400, in conjunction with a time controlled ICCD camera (type Andor, model iStar). This enables us to change the delay in the range from 0 to 5  $\mu\text{s}$  at a fixed gate time of 1  $\mu\text{s}$ . A low cost commercially available diode laser (type SANYO, model DL3147-060, bandwidth of 5 pm) was used. The saturation limit of our diode laser power was calculated at the wavelength corresponding to the  $H_{\alpha}$ -line and was found to be  $\sim 9.5$  mW [38]. However, the laser output power of the diode was measured using a previously absolutely calibrated echelle spectrograph and was found on the average  $1.75 \pm 0.02$  mW at the peak emission wavelength centered at 656.36 nm at an operating temperature of 25 °C and a constant biasing current of 18 mA. A quartz optical fiber of inner diameter of 25  $\mu\text{m}$  (solid angle 76°) with metal caps at its ends for limiting the solid angle to 7.2° was used to collect radiation from the illuminated plasma area  $\sim 4 \times 3$  mm<sup>2</sup>. Spatially integrated plasma transparency data points were averaged over three consecutive laser shots taken at the same position with the first laser shot delivered on a fresh- polished target. The measurements of the plasma optical depth were carried at the peak emission of the diode laser probe beam centered at the shifted  $H_{\alpha}$ -line emission at wavelength  $\lambda_o = 656.36$  nm. It is worth noting that, the scattering of the probe laser was examined by positioning the optical fiber off the diode laser axis up to angle of 45°, and no scattered radiation was detected.

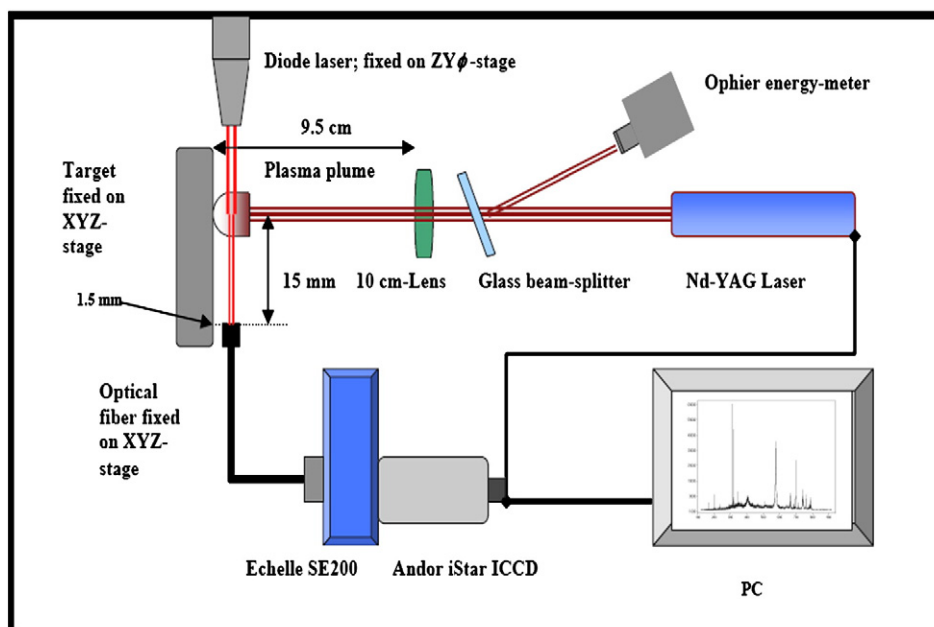


Fig. 1. A schematic diagram of the experimental setup.

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