

Technical note

Real time measurement of the electron density of a laser generated plasma using a RC circuit

F. Bredice^{a,*}, H. Sobral^b, M. Villagran-Muniz^b, H.O. Di Rocco^c, G. Cristoforetti^d,
S. Legnaioli^d, V. Palleschi^d, A. Salvetti^d, E. Tognoni^d^a Centro de Investigaciones Opticas, P.O.Box 124, (1900) La Plata, Argentina^b Laboratorio de Fotofísica, Centro de Ciencias Aplicadas y Desarrollo Tecnológico, Universidad Nacional Autónoma de México, Apartado Postal 70-186 México Distrito Federal, P. O. Box 04510, Mexico^c Instituto de Física “Arroyo Seco”, Facultad de Ciencias Exactas, U.N.C.P.B.A., Pinto 399, 7000 Tandil, Argentina^d Applied Laser Spectroscopy Laboratory IPCF-CNR, Area della Ricerca di Pisa, Via G. Moruzzi 1, 56124 Pisa, Italy

Received 31 January 2007; accepted 24 June 2007

Available online 3 July 2007

Abstract

A Nd:YAG laser pulse was focused, in air or on a Cu target, between the plates of a planar charged capacitor. The plasma generates a transient redistribution of the electrical charges on the plates that can be easily measured as a voltage drop across a resistor connected to the ground plate. At the same time, the Stark broadening of the H_{α} spectral line (656.3 nm) obtained from the optical emission spectrum of the plasma was measured. In this work, we show that the peak of electrical signal measured on the resistor is, in the energy range of our laser (30 mJ to 220 mJ) and at time delays typical of Laser-Induced Breakdown Spectroscopy applications (500–5000 ns), univocally related to the temporal evolution of the Stark broadening of the H_{α} line. Therefore, after a proper calibration depending on the material and the experimental geometry, the peak of the electrical signal can be used to predict the temporal evolution of the electron density of the generated plasma.

© 2007 Elsevier B.V. All rights reserved.

Keywords: Laser-induced breakdown spectroscopy; Plasma diagnostic techniques; Electron density measurement; H_{α} Stark broadening

1. Introduction

The spectroscopic measurement of the electron density of a transient plasma is generally based on the analysis of the profile of spectral lines with known Stark broadening parameters [1–14]. This implies the use of high resolution spectrographs and acquisition systems with time resolution such as box-cars or ICCD [6,7,13,14]. In this work we show that the response obtained from a very simple RC circuit can be used for measuring the plasma electron density.

The characterization of laser-induced plasmas through their electron density (and temperature) has a high interest because it allows improving applications in elemental analysis, quality control, in situ planetary exploration, environmental diagnos-

tics, culture heritage conservation, etc. [1,2]. In particular, laser-induced plasmas are characterized by high electronic density which are of great interest for laboratory or astrophysical studies [15,16].

In previous works [4,5], the authors showed that the peak of the electrical perturbation produced on a uniform electric field when a laser-generated plasma is created inside of a plane plate charged capacitor can be used as a tool for breakdown characterization. This perturbation can be measured as a voltage drop on a resistor connected to the ground plate of the capacitor. A similar method for characterizing the laser ablation and laser processing was also used in references [6,7].

In this work we show that the peak of the electrical signal measured as indicated in the above cited references is univocally related with the time evolution of the full width at half maximum (FWHM) of the Balmer H_{α} line at 656.3 nm; we present the cases where the plasma is created in air or on a metallic (copper) target. Therefore the peak of the electrical

* Corresponding author. Tel.: +54 221 484 0280/2957, fax: +54 221 471 2771.
E-mail address: faustob@ciop.unlp.edu.ar (F. Bredice).

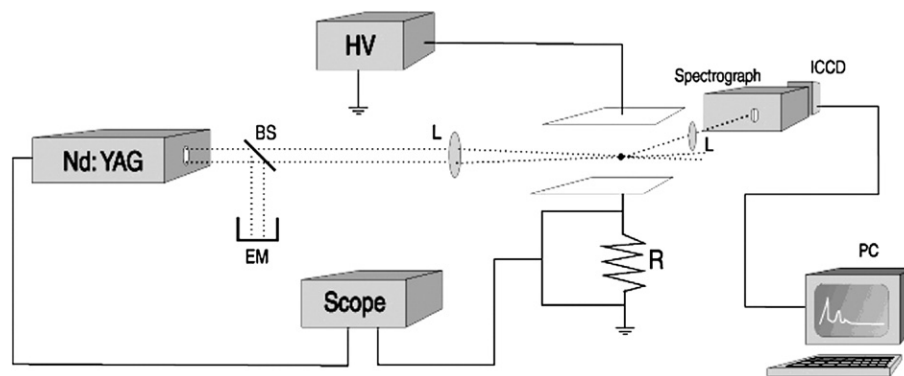


Fig. 1. Experimental setup for the measurement of the H_{α} line profile and the electrical signal. BS=Beam Splitter; EM = Energy Monitor; HV = High Voltage generator; L = lens.

signal can be used, after a proper calibration depending on the material under study, for measuring – in real time – the time evolution of the electron density of the laser-produced plasma. The proposed method is not perturbative, is characterized by a high signal-noise ratio and allows measuring in a simple way both the energy absorbed by the plasma (see Ref. [4]) and the electronic density of the plasma without the need of analyzing the lineshapes of the spectral emission lines.

2. Experimental setup

The experimental set up used was similar to that described in Ref. [4]. A diagram of the experimental device is shown in Fig. 1. The plasma was obtained by focusing the emission of a pulsed Nd:YAG laser (Surelite I from Continuum), operated at $\lambda = 1.06 \mu\text{m}$. The laser delivers pulses from 30 mJ up 220 mJ at a 3 Hz repetition rate, with a 7 ns pulse width (FWHM). The beam was focused into the middle space of the capacitor by a lens of 7.5 cm focal length with anti-reflecting coating.

The capacitor consisted of two plane circular aluminum plates of 100 mm diameter spaced by 3 cm. The applied voltage was 6 kV. The electric field produced inside the capacitor was perpendicular to the laser beam. One of the plates was connected to the ground through a 10 k Ω resistor. The voltage drop across the resistor was registered by a 500 MHz digital oscilloscope (TDS 540 from Tektronix) synchronized with the emission of the Nd:YAG laser. Fig. 2 shows the shape of a typical electrical signal observed on the oscilloscope.

At the same time, the profile of the Balmer H_{α} line at 656.3 nm was resolved using a 50 cm monochromator (Spectra Pro 500i from Roper Scientific, diffraction grating of 1200 lines/mm), with a slit width of 10 μm . A 20 cm focal lens was used to form the image of the plasma onto the entrance slit of the spectrometer. In a typical laser-induced plasma, the temporal variations of electron temperature and density are very steep; therefore, large temporal acquisition windows cannot be used, since in this case very different values of these parameters would be integrated on the detector, and the resulting profile would be affected by this integration. In our experimental configuration, the plasma spectral emission was acquired by a ICCD camera

(PI-MAX:1024 UV from Princeton Instruments) with a fast 100 ns gate, starting from 500 ns up to 5000 ns after the arrival of the laser pulse, with a step of 500 ns. In order to improve the signal-noise ratio, fifty measurements of the emission profiles were averaged.

3. Results and discussion

3.1. The electrical signal

As was pointed out in the introduction, the electronic density of a laser generated plasma can be determined in real time and in easy way through the perturbation that the plasma cause when is generated between the plates of a planar charged capacitor [4,5]. The laser induced plasma produces a transient spatial redistribution of the charges in the plates, changing the capacity of the circuit that is temporarily modified due to the presence of the plasma inside the electric field. This redistribution can be measured as a voltage drop across the resistor.

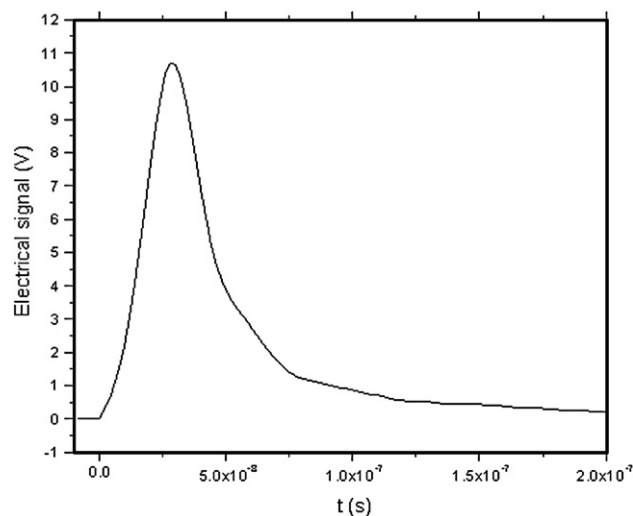


Fig. 2. Temporal evolution of a typical electrical signal. The zero of the time scale corresponds to the start of the laser pulse.

In Ref. [4] the authors showed that the absorbed laser energy used in the generation of the plasma is related to the peak of the electrical signal through the relation:

$$h = \frac{k_c E_{bd} V}{d^3} \quad (1)$$

where h is the maximum voltage drop across the resistor (peak of the signal); E_{bd} is the absorbed energy by the plasma (i.e. the difference between the incident and the transmitted laser energy, see Ref. [4]), k_c is a constant that depends on the circuit parameters, V is the applied voltage and d is the distance between the plates.

According to Ref. [5], a spherical plasma of radius a in a uniform electric field with intensity E_0 behaves as a floating conductor in the external field, acquiring a dipolar moment

$$p = 4\pi\epsilon_0 E_0 a^3$$

where ϵ_0 is the vacuum dielectric permittivity. The corresponding polarizability constant of the plasma is thus independent on the applied electric field strength and orientation. At the early stage of the breakdown, electrons and positive ions are in fast dynamics and their charge centers are not in the same place, so a transient dipole is generated. For this reason the electrical signal is detected few nanoseconds after the breakdown and this time delay is independent of the distance between the plasma and the plates of the capacitor and the energy of the laser pulse. According to Eq. (1), the dependence of the peak of the electrical signal with d^3 suggests that the charge distribution inside the plasma has a dipolar moment, whose dynamics is governed by the applied electric field and the recombination processes. From this consideration we conclude that the peak of the electrical signal is due to the electric field generated by the transient dipole inside the capacitor. These facts seem to contradict the conclusions reported in the paper by Madjid et al. [7], where the electrical signal is attributed to the electrons generated by ionizing radiation reaching the capacitor plates.

Fig. 3 shows the variation of the peak of electrical signal with the laser energy when the breakdown is performed in air and on copper target.

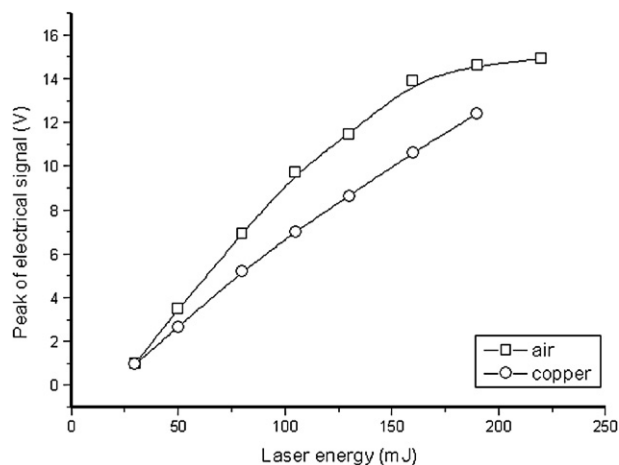


Fig. 3. Peak electrical signal (V) measured across the resistor vs. the laser energy. Squares: Breakdown in air. Circles: Breakdown on Cu target.

A very good linear correlation between the peak of the electrical signal and the laser energy is observed, especially at relatively low laser energies. The saturation effect which is evident at higher laser energy is related to the screening effect of the plasma, preventing the last part of the laser pulse to reach the target above a given threshold [17].

The calibration curves obtained suggest that in actual experiments in air the operating energy of the laser should be maintained around 130 mJ, in order to stay in the linear part of the curve, while for copper targets the linear zone of the calibration curve extends up to 200 mJ.

3.2. Measurement of electron density by Stark broadening

3.2.1. Effect of the self-absorption

Self-absorption is one of the most important source of errors in the measurement of Stark parameters. Laser-induced plasmas are often characterized by a high optical depth due to the high density of atoms and ions that can be found in them. However, in our case, the H_α is originated from the natural humidity present in the atmosphere, which represent a very small concentration of water. At these levels of concentration, the hydrogen spectral lines can be safely considered optically thin [13,14] and consequently the self-absorption is not present in our case. Furthermore the H_α profiles are very well fitted by Voigt functions, showing no sign of self-reversal. The self-reversal phenomenon is revealed by the presence of a 'dip' at the maximum of the emission line, generated by the absorption of the light emitted by the hot core of the plasma by the colder outer layers at the plasma boundary. This phenomenon has not been observed on the recorded spectral profiles of the H_α line.

3.2.2. Calculation of Stark broadening

The approximation most frequently used for the calculation of Stark broadening due to the collision of the emitters with the charged particles in the plasma is in the semiclassical impact approximation. The effect of the electronic collision is supposed to be predominant, while the effect of the collision with the ions is much weaker. Griem [10] suggested the following expression for the measurement of the electron density using the H_α line:

$$n_e(H_\alpha) = 8.02 \times 10^{12} (\Delta\lambda_{1/2}/\alpha_{1/2})^{3/2} (\text{cm}^{-3}) \quad (2)$$

where n_e is the electron density (cm^{-3}), $\Delta\lambda$ is the measured FWHM of the H_α line (in Angstrom) and $\alpha_{1/2}$ is the half width of the Stark profile, tabulated in [11]. Then, the functional dependence of the electron density with the Stark width can be written as

$$n_e = C(n_e, T) \Delta\lambda^{3/2}$$

In this equation $C(n_e, T)$ is a coefficient that is only a weak function of the electron density and the temperature but its dependence on n_e is more significant.

A more accurate determination of the electron density can be achieved using the tables compiled by Gigosios et al. [12]. The authors used computer simulations, including ion dynamics

effects, obtaining a series of tables that provide the Stark FWHM of the Balmer- α , - β and - γ line profiles depending on the electron density, electron temperature and reduced mass of the emitter-ion system. However, in the conditions of our experiment, the values calculated for the electron density using the Gigos tables almost coincide with the Griem estimation (3% maximum difference). In our calculations, we used the values published in reference [12].

Under our experimental conditions, the obtained spectrum and the temporal evolution of the H_α line show no differences in the presence or absence of the electric field up to applied voltages of 10 kV/cm.

The H_α line profiles were fitted using Voigt function; the gaussian contributions to the line broadening (instrumental broadening and Doppler broadening) as well as the Lorentzian one, directly related to the electron density, were obtained by deconvolution. In our experiment, Lorentzian broadening is typically one order of magnitude larger than Gaussian broadening.

In the case of plasmas generated on Cu target, it is more difficult measuring the H_α line width, since at short times after the breakdown this line is blended with several Cu II lines which only disappear for delay times $t > 1 \mu\text{s}$.

In Fig. 4A it is shown the variation of the H_α line emission obtained from the breakdown in air, at delay times ranging from 0.5 to 4.5 μs . The energy of the laser was fixed at 130 mJ.

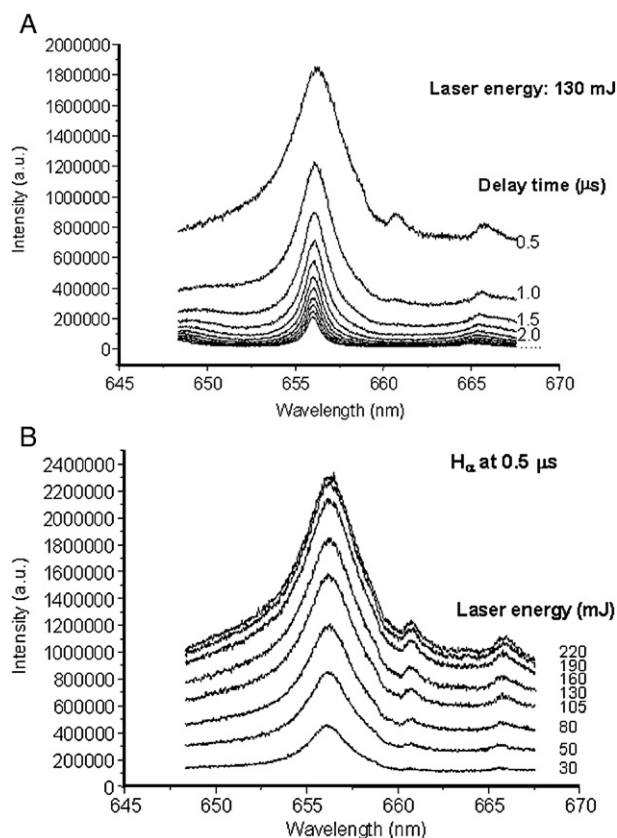


Fig. 4. A— H_α emission profile obtained from the breakdown in air, at different delay times after the laser pulse. The laser energy was fixed at 130 mJ. B— H_α emission profile obtained from the breakdown in air, at different laser energies. The acquisition delay was fixed at 0.5 μs after the laser pulse.

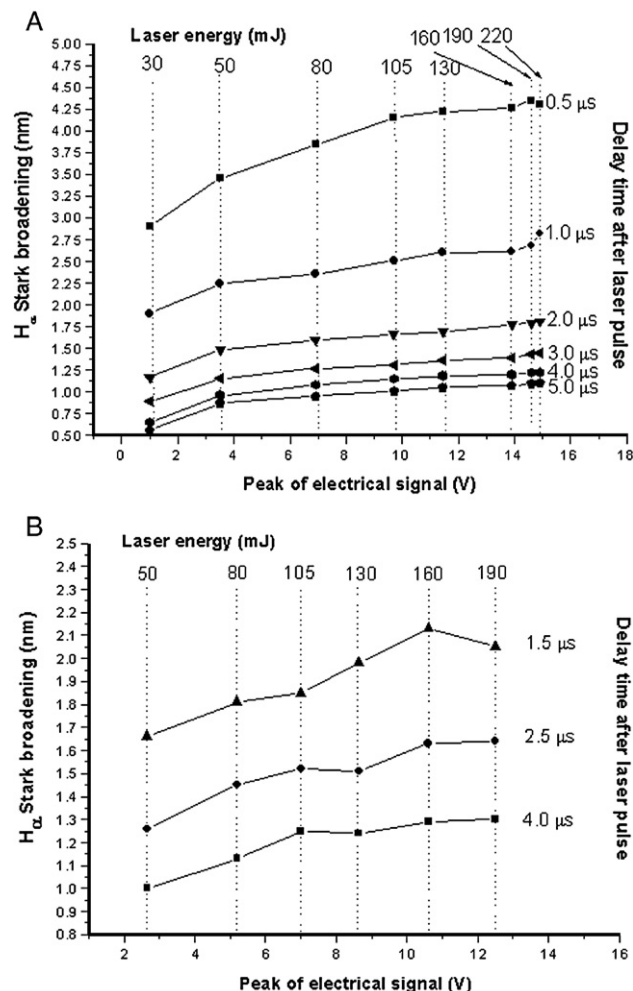


Fig. 5. A—FWHM of the H_α emission line obtained from the breakdown in air, recorded at different delay times after the laser pulse, vs. the electrical signal. B—FWHM of the H_α emission line obtained from the breakdown on Cu target, recorded at different delay times after the laser pulse, vs. the electrical signal.

Fig. 4B shows the variation of the profile of the H_α line obtained from the breakdown in air, versus the energy of the laser. The acquisition delay time was fixed at 0.5 μs .

In Fig. 5A it is shown that, when the laser is focused on air, the time evolution of the FWHM of the H_α line is univocally related with the peak of the electrical signal. Similar situation occurs when the laser is focused on a Cu target (Fig. 5B). Then, based on the correspondence between the values of the FWHM of the H_α and the peaks of the electrical signal, the latest can be used as a measure of the plasma electron density temporal evolution.

In Fig. 6A it is shown the electron density of the plasma produced in air, measured at delay times ranging from 0.5 to 4.5 μs from the FWHM of the H_α according to the tables of Ref. [12], vs. the electrical signal. In Fig. 6B the temporal evolution of the electron density, produced by focusing the laser on a Cu target, is plotted against the electrical signal recorded.

From Fig. 6A and B one can see that the electron density generated on a Cu target is similar to the electron density generated in air for the same laser energy. This fact can be explained by several reasons. In the case of breakdown in air, the laser beam is focused by a focal lens of 7.5 cm; according to Ref.

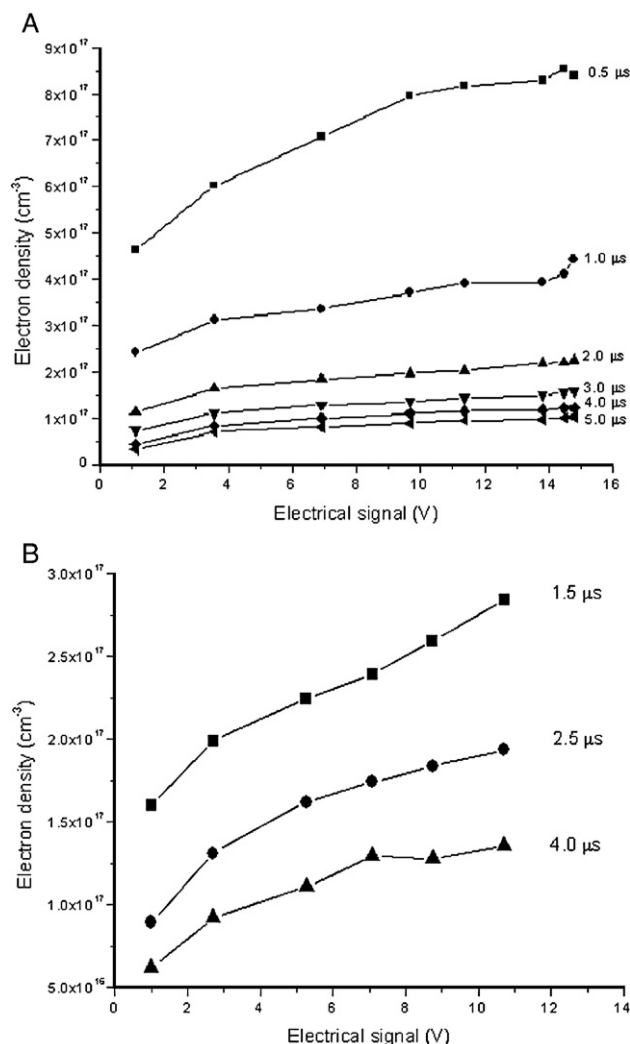


Fig. 6. A—Breakdown in air. Electron density at different delay times after the laser pulse vs. the electrical signal. B—Breakdown on copper target. Electron density at different times delay times after the laser pulse vs. the electrical signal.

[4] the ratio between the laser energy absorbed in the breakdown process vs. the incident energy reaches about 90%. When the same energy arrives to a copper target, an important fraction of it is reflected and diffused by the target, and less energy remains for the plasma generation [5].

4. Conclusion

The most important experimental result of this work is that the temporal evolution of the FWHM of the H_{α} line obtained from a plasma generated in air or in metallic target is related with the peak of the electrical signal measured on the resistor of a simple RC circuit.

Therefore, the results obtained in this work together with those reported in Ref. [4] allow the determination, in real time, of the energy deposited in the laser-produced plasma as well as the temporal evolution of its electron density. This kind of method is not perturbative, has a high signal-noise ratio, and allows building calibration curves (depending on the material and the experimental geometry) for quantitative measurements

of the mentioned parameters without the use of spectral and temporal analyzers.

Acknowledgments

This work was supported by the National Autonomous University of Mexico GAPAUNAM and the National Council of Science and Technology of Mexico CONACyT.

The authors also want to thank the Comisión de Investigaciones Científicas de la Provincia de Buenos Aires, Argentina, where one of the authors (F.B.) works as researcher.

This activity was performed in the framework of the SAILORMAN (Southern American-Italian LIBS-Oriented Research for Material Analysis Network).

References

- [1] A.W. Miziolek, V. Palleschi, I. Schechter (Eds.), *Laser Induced Breakdown Spectroscopy*, Cambridge University Press, Cambridge, 2006.
- [2] D.A. Cremers, L.J. Radziemski (Eds.), *Laser spectroscopy and applications*, Marcel Dekker, New York, 1987.
- [3] M.A. Uman, R.E. Orville, Electron density measurement in lightning from Stark-broadening of H_{α} , *J Geophys. Res.* 69 (1964) 5151–5154.
- [4] F.O. Bredice, D.J.O. Orzi, D. Schinca, H. Sobral, M. Villagrán-Muñiz, Characterization of pulsed laser generated plasma through its perturbation in an electric field, *IEEE Trans. Plasma Sci.* 30 (2002) 2139–2143.
- [5] A. Robledo Martinez, H. Sobral, M. Villagrán-Muñiz, Measurement of the electrical size of a laser induced plasma in a uniform field, *Appl. Surf. Sci.* 248 (2005) 32–35.
- [6] M.H. Hong, Y.F. Lu, Plasma diagnostics at early stage of laser ablation, *Appl. Phys., A* 69 (1999) S605–S608 (Suppl.).
- [7] S.N. Madjid, I. Kitazima, T. Kobayashi, Y.I. Lee, K. Kagawa, Characteristics of induced current due to laser plasma and its application to laser processing monitoring, *Jpn. J. Appl. Phys.* 43 (2004) 1018–1027.
- [8] J. Bengoechea, J.A. Aguilera, C. Aragón, Application of laser-induced plasma spectroscopy to the measurement of Stark broadening parameters, *Spectrochim. Acta Part B* 61 (2006) 69–80.
- [9] A.M. El Sherbini, H. Hegazy, Th.M. El Sherbini, Measurement of electron density utilizing the H_{α} -line from laser produced plasma in air, *Spectrochim. Acta Part B* 61 (2006) 532–539.
- [10] H.R. Griem, *Spectral Line Broadening by Plasmas*, Academic Press, New York, 1974.
- [11] H.R. Griem, *Principles of plasma spectroscopy*, Cambridge University Press, Cambridge, 2005.
- [12] M.A. Gigoso, V. Cardeñoso, New plasma diagnosis tables of hydrogen Stark broadening including ion dynamics, *J. Phys. B: At. Mol. Opt. Phys.* 29 (1996) 4795–4838.
- [13] A.M. El Sherbini, Th.M. El Sherbini, H. Hegazy, G. Cristoforetti, S. Legnaioli, V. Palleschi, L. Pardini, A. Salvetti, E. Tognoni, Evaluation of self-absorption coefficients of aluminum emission lines in laser-induced breakdown spectroscopy measurements, *Spectrochim. Acta Part B* 60 (2005) 1573–1579.
- [14] F. Bredice, F.O. Borges, H. Sobral, M. Villagrán-Muñiz, H.O. Di Rocco, G. Cristoforetti, S. Legnaioli, V. Palleschi, L. Pardini, A. Salvetti, E. Tognoni, Evaluation of self-absorption of manganese emission lines in Laser Induced Breakdown Spectroscopy measurements, *Spectrochim. Acta Part B* 61 (2006) 1294–1303.
- [15] L.C. Popović, M.S. Dimitrijević, “The electron impact broadening parameters in hot star atmospheres: Mn II, Mn III, Ga III, Ge III and Ge IV lines”, *Astron. Astrophys., Suppl. Ser.* 128 (1998) 203–205.
- [16] S. Djenize, S. Bukvić, A. Srećković, D. Nikolić, “The first measured Mn II and Mn III Stark broadening parameters”, *New Astron.* 11 (2006) 256–261.
- [17] X. Mao, R.E. Russo, Observation of plasma shielding by measuring transmitted and reflected laser pulse temporal profiles, *Appl. Phys., A* 64 (1997) 1–6.