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Citation for published version (APA):

Torres, J., Mullen, van der, J. J. A. M., Gamero, A., & Sola, A. (2010). Multiple diagnostics in a high-pressure hydrogen microwave plasma torch. *Applied Physics Letters*, 96(5), 051501-1/3. [051501].
<https://doi.org/10.1063/1.3306731>

DOI:

[10.1063/1.3306731](https://doi.org/10.1063/1.3306731)

Document status and date:

Published: 01/01/2010

Document Version:

Publisher's PDF, also known as Version of Record (includes final page, issue and volume numbers)

Please check the document version of this publication:

- A submitted manuscript is the version of the article upon submission and before peer-review. There can be important differences between the submitted version and the official published version of record. People interested in the research are advised to contact the author for the final version of the publication, or visit the DOI to the publisher's website.
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Multiple diagnostics in a high-pressure hydrogen microwave plasma torch

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(Received 14 September 2009; accepted 11 January 2010; published online 3 February 2010)

We present an experimental study of a hydrogen plasma produced by a microwave axial injection torch, launching the plasma in a helium-filled chamber. Three different diagnostic methods have been used to obtain the electron density and temperature as follows: The Stark intersection method of Balmer spectral lines (already tested in argon and helium plasmas); the modified Boltzmann-plot showing that the plasma is far from the local thermodynamic equilibrium but ruled by the excitation-saturation balance; and a study by the disturbed bilateral relations theory. All of these diagnostic techniques show a good agreement. © 2010 American Institute of Physics.

[doi:10.1063/1.3306731]

We have attempted to generate a hydrogen discharge using a *torche à injection axiale*; TIA device¹ to create a two-temperature plasma generated by microwaves. The study has been focused from different diagnostic techniques, which has permitted the comparison and validation of their results. The plasma is created using a TIA working at 2.45 GHz microwave energy at two high frequency (HF) powers: 600 and 1000 W. The chamber was initially designed as a reaction chamber for organic compound destruction.² A stable, pure hydrogen plasma flame of typically some millimeters in diameter and a few centimeters long was produced expanding in the surrounding helium atmosphere occupying the discharge chamber. Only spectroscopic lines of atomic hydrogen were observed. Further details on the experimental setup and working conditions can be found in previous works.³

1) *Crossing-point or Stark intersection method* (SIM) of spectral lines: The basis of the diagnostic method determining simultaneously n_e and T_e by Stark broadening lies on the study of two or more lines broadened under the same working conditions.^{3–6} We use the microfield model method, a computational simulation theory due to Gigosos *et al.*⁷ ap-

plied to the three first Balmer series lines (H_α , H_β , and H_γ) in a nonequilibrium (two-temperature) plasma. The broadening of different spectral lines depends differently on n_e and T_e . In a coherent experiment, all the values of n_e related to different broadenings of different spectral lines, coincide at a specific T_e (the so called *crossing or intersection point*).

The experimental profile must be cleaned by deconvoluting different broadening mechanisms other than the Stark broadening. In this way, we can show the relation between electron density and electron temperature, where the Stark broadening is an external parameter, obtained experimentally in terms of the full-width at half-maximum (FWHM) (Fig. 1). H_α is not useful, because the large self-absorption that H_α suffers in pure hydrogen plasmas (however, H_α was used in other experiments with Ar or He plasmas to have a diagnostic involving three lines simultaneously⁵). For H_β and H_γ , self-absorption is not so important under the experimental conditions of our hydrogen plasma. The simultaneous diagnosis of electron density and has been done, in a point of the discharge very close (1 mm) to the TIA nozzle tip.

2) *Modified Boltzmann-plot*: As we have measured up to

FIG. 1. Intersection point, n_e vs T_e , using Stark H_β and H_γ FWHM as a parameter, in the 600 and 1000 W experiments.

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FIG. 2. Boltzmann-plot in the 600 and 1000 W experiments.

FIG. 3. Relative populations vs effective quantum number p , in the 600 and 1000 W experiments.

seven hydrogen spectral lines of Balmer series, we have thought about a kind of Boltzmann-plot analysis, which is the study of the excited state populations of the atomic levels versus their excitation energies. If the plasma is in equilibrium, the electron temperature T_e can be obtained experimentally from a logarithmic representation of the level populations per statistical weight $[\ln n(p)/g(p)]$ against their ionization potentials (I_p) as the slope of a straight line. However, it can be expected that the atomic state distribution function (ASDF) will deviate from the Saha–Boltzmann equation in the observed ionizing zone of the hydrogen plasma. Under the excitation-saturation balance (ESB) the excited level populations can be described by

$$\frac{n^s(p)}{g(p)} = \frac{n(p)}{g(p)} \frac{1}{p^{-x}} \exp\left(\frac{I_p}{k_B T_e}\right), \quad (1)$$

where the x exponent value must be around 6. From the experimental spectral lines, the integrated intensity (area) I_{exp} is calculated (after calibration), and from this, the relative populations are determined. A Boltzmann-plot is constructed and represented in a semi-log plot (Fig. 2)—the plasma is not in local thermodynamic equilibrium (LTE) (*not* a straight line). But in the double-logarithmic representation of the populations against the principal quantum number p (Fig. 3), the populations of the departure levels corresponding to each Balmer transition are linearly distributed—with the exception of the first line in the series, H_{α} ; this is consistent with the idea of the H_{α} self-absorption in this pure hydrogen plasma, but not for the rest of the Balmer lines. Excluding H_{α} , a linear fit can be obtained, which results in slope values that are close to the theoretical value of the p^{-x} law as follows: 5.6 ± 0.1 for 600 W (correlation coefficient,

FIG. 4. Numerical solution of the equation for the electron temperature T_e , based on the temperature of heavy particles T_h , in the dBR theory.

TABLE I. Experimental results.

Experiment TIA-H		T_e	n_e
		[(K)–(eV)]	(cm ^{−3})
Stark intersection method (SIM)	600 W	9500–0.82	4×10^{14}
	1000 W	10 000–0.86	5×10^{14}
Modified Boltzmann-plot in ESB	600 W	10 500–0.90	...
	1000 W	10 700–0.94	...
Disturbed bilateral relations (dBR)	600 W	10 300–0.89	1×10^{14}
	1000 W	10 300–0.89	2×10^{14}

$r \sim 0.9991$) and 5.9 ± 0.1 for 1000 W ($r \sim 0.9994$). So, we can conclude that the plasma is very close to the ESB, and using this experimental value for the exponent, it is possible to correct the experimental populations to obtain the equilibrium values. From the populations of the excited levels in a theoretical LTE situation, the excitation temperature is obtained. This so-called *modified Boltzmann-plot method* has been used before in middle-pressure surface-wave argon discharges.⁸ This modified Boltzmann-plot does not provide any information about the electron density.⁹

$$\hat{T}_e = \frac{\hat{I}^*}{\ln \left\{ \frac{C_i(H) \cdot \hat{n}_1^2 \cdot \hat{\Lambda}^2 \cdot \hat{\sigma}_{ia} \cdot \sqrt{A} \cdot \left(\frac{R}{I^*} \right)^2}{5.521 \times 10^3} \times f(T_e/T_h) \right\} - 0.0854}, \quad (2)$$

where the average dimensionless diffusion length $\hat{\Lambda}$ has been considered according to the measurements of Thomson scattering in TIA argon plasma ($\hat{\Lambda} = \Lambda/1 \text{ mm} = 0.1$), and $f(T_e/T_h)$ is a function of the electron and heavy particle temperatures given by

$$f(T_e/T_h) = \sqrt{T_e/T_h} [1 + T_e/T_h]^{-1}. \quad (3)$$

Thus Eq. (2) for the electron temperature is recurrent. In order to solve it we have performed an iterative process. The results of T_e for different initial values from the temperature of heavy particles T_h are shown in Fig. 4. T_h reasonably ranges from 0.3 eV—the temperature for molecular hydrogen dissociation at atmospheric pressure (there are no molecular traces in our discharge), to about 0.6 eV (melting temperature of the experimental assembly of TIA). For that interval, the variation in the electron temperature is only 1%. The dependence of this result on the HF power provided by the generator is not very important.¹⁰ It is interesting to indicate that the case with $T_e/T_h = 2$, the ratio between temperatures that was used for the diagnosis based on the SIM of spectral lines, is within this interval of possible temperatures considered by this dBR study.

On the other hand, from the electron energy balance equation it is possible to obtain the electron density [Eq. (4)] using the mean values for our plasma as follows:¹⁰

$$n_e = \frac{\varepsilon \cdot n_1}{S_{\text{heat}}(k_B T_e - k_B T_h)}. \quad (4)$$

From the different methods and analysis we have used, Table I shows the results obtained for the diagnosis of the pure hydrogen TIA plasma.

Furthermore, we have validated the SIM by comparing its results with those obtained by other independent techniques. The pure hydrogen plasma produced by TIA is a very

3) *Disturbed Bilateral Relations* (dBR): The general formulation of the dBR theory can be applied to estimate the values of electron density and temperature in the hydrogen plasma produced by TIA.¹⁰ This method permits us to establish the equation of balance of the electron density and the equation of energy balance for electrons. We assume that the studied zone of the plasma is highly ionizing, since this has been widely verified with the modified Boltzmann-plot technique. Finally,¹⁰ the expression for the electron temperature is obtained by the following:

delicate experiment, but under our experimental conditions, seven different spectral hydrogen Balmer lines have been measured. The hydrogen plasma is a two-temperature non-equilibrium plasma. Using the modified Boltzmann-plot technique, the results show that the hydrogen plasma is not in LTE. The overpopulation of excited states follows the p^{-x} law, so, the TIA hydrogen plasma can be considered to be ruled by the ESB theory in the observed zone, and the Boltzmann-plot can be modified so that the equilibrium values of the populations are obtained and the value of the electron temperature can be deduced. In all the three diagnostic studies, values of around one electron-volt (10 000 K) were found for the electron temperature, which are in good agreement with those we can expect according to other studies in TIA produced plasmas.

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