A simple and accurate analytical model of the Stark profile and its application to plasma characterization

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Abstract:

Optical Emission Spectroscopy techniques are among the most employed to perform the characterization of laboratory plasmas. The analysis of the obtained data is based on the convolution of three different types of profiles: Lorentzians, Gaussians and Starks. While analytic expressions are available for the first two types, the Stark profile has been traditionally obtained through theoretical calculations using different models. In this paper, we propose is a simple and accurate analytical function that can be employed as approximation of a Stark profile. The application of this new model may simplify the analysis of plasmas.

Keyword: Plasma; Spectroscopy; Simulated Profiles; Stark Effect

1. Introduction

During past decades, plasmas have found application in a wide range of fields such as gas detoxification, materials processing, catalysis, elemental analysis, treatment of liquids (water), sterilization... [1-6]. This increasing interest in plasmas has led to the need of a good understanding of its internal mechanisms and governing parameters, since the optimization for its different applications depends on it. The knowledge of the electron density is crucial because it plays an important role in the ability of plasmas to induce reactions.

Optical Emission Spectroscopy (OES) is a passive spectroscopy technique widely used in the study of plasma parameters, being a non-perturbing method that does not alter the internal kinetics of the discharges during the measurement process [7-10]. This technique is based on the collection of the radiation coming from the plasma and its subsequent molecular and atomic emission spectra. Experimental atomic spectral lines have a distribution of intensity around their central wavelength (line profile). The profile shape and its characteristic parameters such as its width (full width at half maximum, FWHM) and intensity (area under this profile) are of great importance since they depend on the internal processes taking place in the discharge. More specifically, these processes contribute to the total width of a spectral line in an independent manner [11, 12]. Among them, Hydrogen Balmer series lines (and more concretely the H_β line) are usually used for determining the electron density in laboratory plasma since the electron density is related to their width [7].

The most relevant processes to be taken into account are the collisions of the emitter hydrogen atom with the charged particles in its surroundings (Stark broadening) [13-16], the dipole moment induced by neutral atom perturbers in the instantaneous oscillating electric field of the excited emitter atom (van der Waals broadening) which

generates the line shape described by a Lorentzian function [17], the movement of emitter atoms (Doppler broadening) [18] and the error induced by the device used for the plasma radiation registration (Instrumental broadening) which both generate a Gaussian function [7]. All these effects produce a deformation in the measured spectral profiles from the plasma, which allow us to determine the values of its characteristic parameters: electron density, electron temperature and gas temperature [12].

There are two different kinds of methods to analyze this Hydrogen Balmer series lines. First, the methods assuming a Lorentzian function for the Stark profile like as, commonly used for commercial software which discriminates, by means a Levenberg-Marquardt non-linear fitting algorithm for minimum squares, the Lorentzian and Gaussian contributions of the profile shape. The main advantage of these techniques lies in their readiness and quickness of calculus. However, the assumption of Lorentzian shape for Stark profile is valid only when broadening by the collisions with ions is negligible or for temperatures high enough that the impact approximation is valid not only for electrons but also for ions. If ions are quasistatic, their contribution results in asymmetry of line shape [10]. Also ion dynamics effects modify lineshapes which is the consequence of the kinetics of the emitter and perturber [16]. Additionally, if at high densities the no-quenching approximation is violated, the coupling of levels with different principal quantum numbers contributes to the asymmetry of spectral line [18, 19]. The error introduced by non-Lorentzian ion broadening has been studied by Konjević *et al.* [20] and this error is most significant in an electron density range below the "low n_e limit", when the separation between Fine Structure Components becomes larger than the Stark broadened line width. As an example, this occurs in argon plasmas generated at pressures higher than 100 Torr, where the electron density is of the order of 10^{14} cm^{-3}.

The second method is based on the models and theories which describe Stark broadening taking into account ion dynamics and other effects producing asymmetry of the line profile and departure from the Lorentzian shape [13-16]. This method consists of comparison between the experimental profile shape and the profile calculated through the convolution of different functions corresponding to the different phenomena provoking its broadening. In this sense, there are several works using this method for the H_β line: Ranson *et al.* [21] and Thomsen *et al*. [22] applied it to a plasma with electron densities around 10^{15} cm^{-3}, Acon *et al.* [23] applied this method to calculate the electron density values in an Ar Inductively Coupled Plasma (ICP) (10^{15} cm^{-3}), a He ICP (10^{13} cm^{-3}), a glow discharge (10^{13} cm^{-3}) and a High Voltage (HV) spark (>10^{15} cm^{-3}) and Žikić *et al.* in [24] used plasmas with electron density within the range between 10^{15} and 10^{17} cm^{-3}. Among them, the most used model explaining the Stark broadening of hydrogen is the one developed by Gigosos *et al*. (CS model) [16] based on the inclusion of the non-equilibrium conditions existing in two-temperature plasmas, (plasma with electron temperature, T_e, higher than the gas temperature, T_g). However, this model does not offer an analytical function which can be employed to generate a Stark profile.

In this paper we propose an approximate method for the Stark profile which approximates the CS profile when it is convoluted with the profiles due to other broadening mechanisms. If there are no more sophisticated calculations, or a quick check during experiment is needed, this method may be useful. So, the structure of the paper is as follows: in Section 2, we explain the previous models used to describe Stark profiles and our new model is proposed together with the corresponding coefficients deduced by means of statistical procedures; in Section 3, this new model is applied to

the characterization of two real plasmas in order to validate it; and finally are presented the conclusions obtained in this work.

2. Theory

2.1. Modeling the Stark Profile

As mentioned in the introduction, experimental profiles (P_{exp}) obtained from plasma measurements may be described as the combination of various individual profiles that take into account the impact of each process acting into the system. All these contributions can be basically reduced to the convolution of a Lorentzian (van der Waals broadening), a Gaussian (Instrumental and Doppler broadenings) and a Stark profile (Stark broadening) [16]:

$$P_{exp}(\omega_L, \omega_G, \omega_S) = P_L(\omega_L) \otimes P_G(\omega_G) \otimes P_S(\omega_S) \quad (1)$$

where ω_L and ω_G are the width of the Lorentzian and Gaussian profiles respectively, providing information about the gas temperature [17], while ω_S is the Stark width that contains information about the electron density and temperature [13-16].

While there are analytical expressions for the Lorentzian and Gaussian profiles, there is not such formula for the Stark one if effects like the influence of ion dynamics, quasistatic ions or breakdown of no-quenching approximation have a non negligible influence on the lineshape. The best efforts directed in the past to that end have led us to some more or less sophisticated models that can approximate the Stark profile with high accuracy, but at an expensive computational cost.

In this work we use as starting point the CS model [16], which consider a weakly coupled, globally neutral, homogeneous and isotropic plasma, where the particles (ions and free electrons) are independent classical particles that move along rectilinear paths with constant velocity. Velocities are given by the Maxwell–Boltzmann distribution. In order to take into account the emitter kinetics, a relative

movement between emitter-ion pairs introducing the reduced mass of the pair, μ (corresponding to plasmas in thermodynamic equilibrium) is used in the previous model (μ-ion model [15]) and later, Gigosos *et al.* [16], by means of the Computer Simulation CS model, reinterpreted the μ-ion model in order to include the non-equilibrium conditions existing in two-temperature plasmas ($T_e > T_g$). This is controlled by means of the parameter $\mu_r = \mu\, T_e/T_g$, which is a fictitious reduced mass used in order to adjust the perturbing ion mobility to that of the emitter. This model provides Stark theoretical profile simulated with different values of μ_r, T_e and n_e [16].

In order to find an analytical expression which fits these simulated profiles from the CS model, we took into account that when electric fields, influencing the emitter, is varying slowly in comparison with the time of the atom emission, they produce "a typical Stark shift of the spectral line" [25]. Consequently the profile can be considered as a superposition of individually shifted spectral lines similarly to Doppler broadening, which is the result of superposition of Doppler shifted lines. So, a sum of the symmetrical Lorentzian profiles has been employed for fitting the Stark profile given by the CS model. Each pair of symmetrical Lorentzian profiles has amplitude (A_i), width (ω_{Li}) and central wavelength (λ_{oi}).

$$P_S \backsim \sum_{i=1}^{N} [P_L(A_i, \lambda_{oi}, \omega_{Li}) + P_L(A_i, -\lambda_{oi}, \omega_{Li})] \tag{2}$$

Where,

$$P_L(A_i, \pm\lambda_{oi}, \omega_{Li}) = \frac{2A_i}{\pi} \frac{\omega_{Li}}{4(\lambda \mp \lambda_{oi})^2 + \omega_{Li}^2} \tag{3}$$

The minus sign in the second Lorentzian in Eq. (2) is due to the symmetrical character of the paired profiles from the central wavelength of the original Stark profile. As the number of couples increases, a higher precision is expected to be achieved but with the corresponding increment of computational cost.

2.2. New proposed model

The starting point of our work is the CS model. This model has proven its validity in many studies, so we have chosen seven Stark profiles given by this model to be used as *theoretical* cases in our comparative process. As can be seen in Table 1, we have selected these profiles over an interval of temperatures going from 6000 to 8000 K, and a range of densities from 1×10^{14} to 1×10^{16} cm^{-3}, and a reduced mass of 4.

	1	**2**	**3**	**4**	**5**	**6**	**7**
n_e (cm^{-3})	1.00×10^{14}	2.14×10^{14}	4.67×10^{14}	1.00×10^{15}	2.14×10^{15}	4.67×10^{15}	1.00×10^{16}
T_e (K)	6001	7751	6952	6597	6523	6657	6964

Table 1. Theoretical Stark profiles obtained by means of the CS model.

The next step was to apply this fitting to approximate these Stark profiles as a sum of Lorentzian ones. In order to find the optimal number of pairs, we carried out a fitting process of every theoretical case as sum of 2, 4, 6 and 8 Lorentzians respectively. The Levenberg-Marquardt nonlinear regression has been used, calculating the Root Mean Square (RMS) and Pearson coefficient *r* to establish the goodness of the fittings.

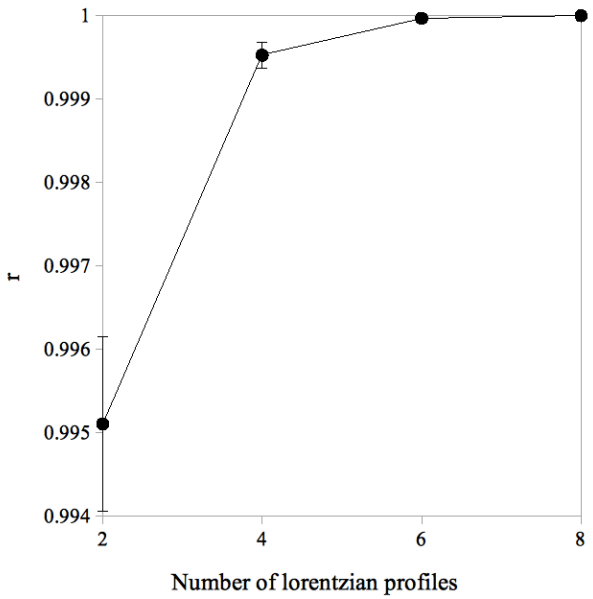

Figure 1. Averaged RMS of the six theoretical cases for the Stark broadening profile approximation as sum of 2, 4, 6 and 8 Lorentzian profiles.

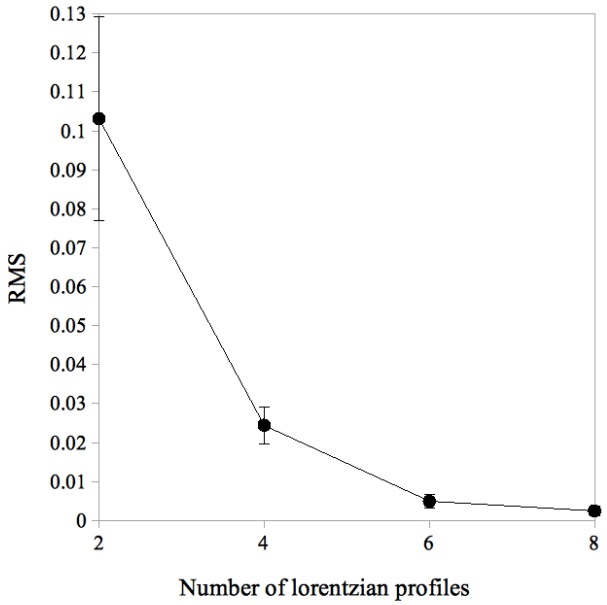

Figure 2. Averaged Pearson coefficient of the six theoretical cases for the Stark broadening profile approximation as sum of 2, 4, 6 and 8 Lorentzian profiles.

Figures 1 and 2 show the obtained average values of RMS and Pearson coefficient for the different sums of Lorentzians; the lower the RMS and the nearer to 1 the Pearson coefficient, the better approximation would be. As could be seen, there is a qualitative leap between the two Lorentzian approximations and the rest of cases, so we have discarded the two Lorentzians case. The four Lorentzians expansion shows slight differences with the six and eight cases, but taking into account that the computational cost duplicates with every increasing in the number of profiles, and that these differences are second order in the RMS and third order in *r*, we have chosen the four Lorentzians sum as the optimal compromise between accuracy and computational cost for the Stark profile:

$$P_S(A_1, A_2, \pm\lambda_{o1}, \pm\lambda_{o2}, \omega_{L1}, \omega_{L2}) \backsim \frac{2A_1}{\pi}\frac{\omega_{L1}}{4(\lambda-\lambda_{o1})^2+\omega_{L1}^2} + \frac{2A_2}{\pi}\frac{\omega_{L2}}{4(\lambda-\lambda_{o2})^2+\omega_{L2}^2} +$$

$$\frac{2A_1}{\pi}\frac{\omega_{L1}}{4(\lambda+\lambda_{o1})^2+\omega L_1^2} + \frac{2A_2}{\pi}\frac{\omega_{L2}}{4(\lambda+\lambda_{o2})^2+\omega L_2^2} \quad (4)$$

The seven theoretical Stark profile cases have been fitted to four Lorentzian. Figure 3 an example (corresponding to the profile 3 of Table 1) of a CS model profile compared with the one given by equation (4), sum of four Lorentzian profiles is shown.

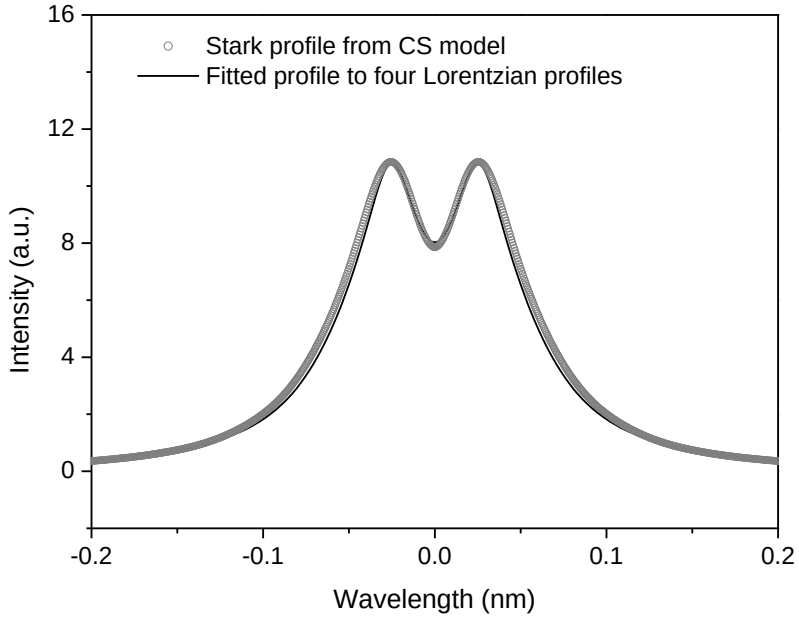

Figure 3. CS model profile compared with our analytical function, sum of four Loretzian profiles from equation (4)

The fitted values ($\omega_{L1}, \omega_{L2}, \lambda_{o1}, \lambda_{o2}, A_1$ and A_2) present a great variability, so it would be desirable to find an expression that could fit these data in an analytic expression. Moreover, we would like that this function depends on the electronic density (n_e), the parameter we want to measure from the plasma; and, since the range of temperatures is relatively small, we can suppose that its influence would be negligible.

In this sense, if we represent the widths (nm), ω_{L1} and ω_{L2} and the central wavelength (nm), λ_{o1} and λ_{o2} versus the electronic density (cm^{-3}), we can observe an exponential relation between them; the same applies to the central positions. A fit of the values gives rise to the following relations,

$$\omega_{L1} = (1.8 \pm 0.4) \cdot 10^{-11} \times (n_e)^{(0.657 \pm 0.007)} \quad (5)$$

$$\omega_{L2} = (1.1 \pm 0.6) \cdot 10^{-10} \times (n_e)^{(0.582 \pm 0.015)} \quad (6)$$

$$\lambda_{o1} = (6.6 \pm 0.6) \cdot 10^{-12} \times (n_e)^{(0.669 \pm 0.005)} \quad (7)$$

$$\lambda_{o2} = (7.2 \pm 0.8) \cdot 10^{-12} \times (n_e)^{(0.650 \pm 0.004)} \quad (8)$$

with Pearson coefficient higher than 0.997. It must be recalled that the validity of this approximation is restricted to the mentioned interval of temperatures going from 6000 to 8000 K.

The amplitudes A_1 and A_2 however are very similar for all of the seven profiles, so we have checked the mean value can be employed with a good approximation (A_1=0.3205 a.u. and A_2=0.2018 a.u.). Substituting eqs. (5)-(8) and the amplitudes into eq. (4), we obtain a simple and accurate function for the Stark profile (P_S) that depends only on electron density (n_e).

Therefore, our final expression for the line profile based on equation (1) will be the result of the convolution of the three profile; Stark profile P_S (n_e), Gaussian profile P_G (ω_G) and Lorentzian profile P_L (ω_L) what gives us an analytical expression with significantly lower computational cost. So, our total analytical function for the profile has this dependence:

$$P_{Simulated}(\omega_L, \omega_G, n_e) = P_L(\omega_L) \otimes P_G(\omega_G) \otimes P_S(n_e) \quad (9)$$

And with that, we can fit three parameters, ω_L, ω_G, n_e at the same time.

3. Results: Experimental verification and discussion

The final objective of this work was the characterization of a plasma through the measurement of its electronic density. Since we now dispose of a *friendly* approximation to the Stark profile that is function of this parameter, we would like to

prove the goodness of this new model by determining the characteristics of one real plasma.

In order to do this, two hydrogen Balmer series lines from an argon plasma column at atmospheric pressure were measured. This plasma was created in a quartz tube with one of its ends opened to the air. The inner and outer diameter dimensions of the discharge tube were of 1 and 4 mm, respectively. The electron density decreases longitudinally as the wave propagates away from the launcher (surfaguide) transferring its energy to the plasma to an axial position.

The optical system used for the spectroscopic measurements consisted of a Jobin-Yvon Horiba 1000M ® spectrometer (Czerny-Turner type), with 1 m of focal distance and a holographic diffraction grating of 2400 lines/mm. A Hamamatsu R928P photomultiplier with a spectral output interval of 200-750 mm was used as the detector.

The radiation emitted by the plasma was picked up across the column by a vertical optical fiber and guided to the entrance slit of the spectrometer. In these experiments, slits of 50 μm width were used and the spectra were taken with a step of 1 pm obtaining the discrete experimental profile. The position z was measured from the end of the plasma column, introducing for these conditions an instrumental broadening of (0.021 ± 0.001) nm.

On the other hand, because the gas temperature was equal, the electron temperature for the plasma column was equal to ≈ 1450 K and 6000-7000 K respectively [26], the parameter μ_r used for calculating the Stark profiles from the CS model is approximately 4.0 ($\mu_{\text{Ar-H}}$ = 0.975 a.m.u). And the value of the electron density is of the order of 10^{14} cm^{-3}.

Within the method considered here an experimental profile can be expressed as the convolution of a Lorentzian, a Gaussian and a Stark profiles. The characterization of the two experimental profiles, H_β for z equal to 4 and 12 cm, would consist on the search for the width of the Lorentzian profile due to Van der Waals and Gaussian profile due to Doppler and Instrumental broadening as well as the electronic density for the Stark profile. Once we have the three theoretical profiles, we convolute them to obtain the theoretical profile and compare it with the measured data to check the

accuracy of the fitting. The minimum RMS and Pearson coefficient are the criteria to choose the optimal fitting profile.

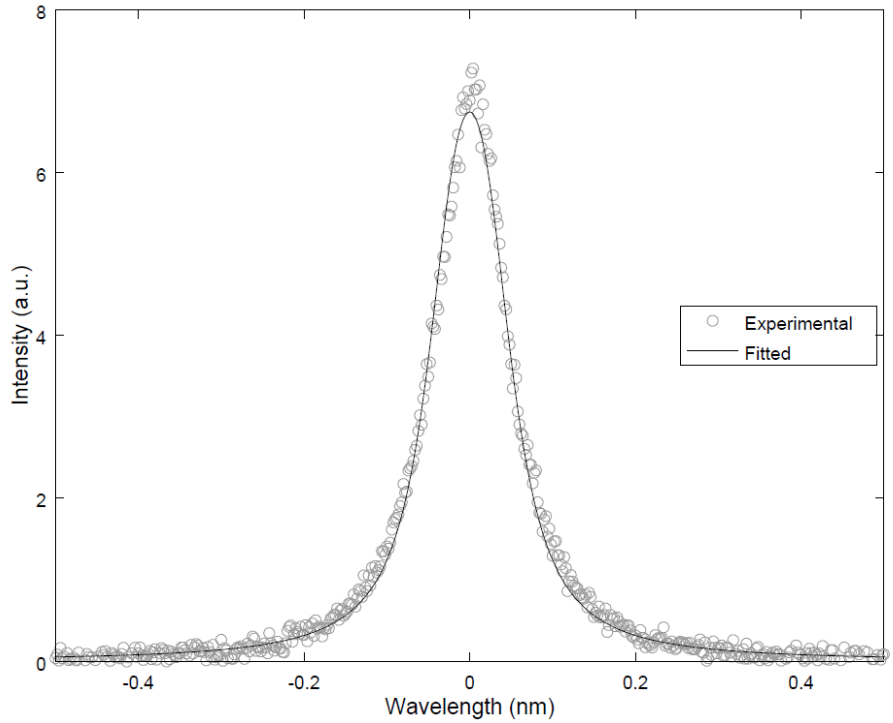

Figure 4. H_β experimental and theoretical profile for a column of plasma of z = 4 cm. The experimental profile has been centered to simplify the calculations.

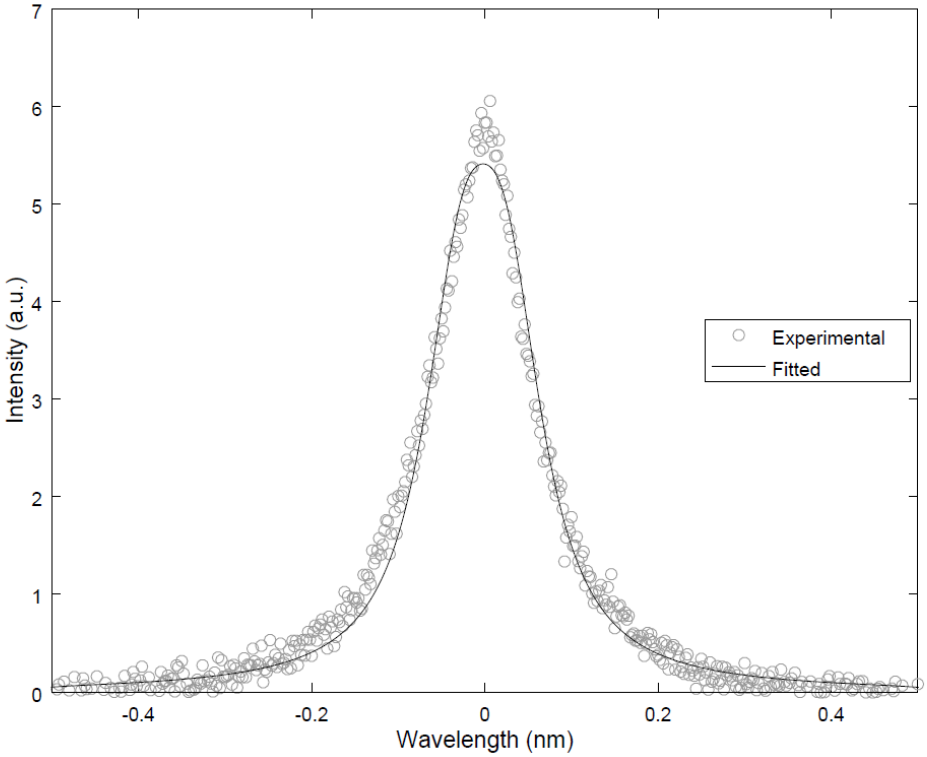

Figure 5. H_β experimental and theoretical profile for a column of plasma of z = 12 cm. The experimental profile has been centered to simplify the calculations.

Figs. 4 and 5 show the cases of z = 4 cm and z = 12 cm, where z is the plasma column length. As can be seen, the agreement between the experimental and theoretical curves is large in both cases, with a RMS value of 0.1173 for the first case and 0.1811 in the second. Table 2 contains the experimental results [26] and calculated values of this work:

z = 4 cm	**ω_L (nm)**	**ω_G (nm)**	**n_e ($\cdot 10^{14}$cm^{-3})**
Experimental [26]	0.035 ± 0.004	0.025 ± 0.001	1.42 ± 0.18
Theoretical	0.039 ± 0.006	0.029 ± 0.004	2.0 ± 0.9
z = 12 cm	**ω_L (nm)**	**ω_G (nm)**	**n_e ($\cdot 10^{14}$cm^{-3})**
Experimental [26]	0.035 ± 0.004	0.025 ± 0.001	3.7 ± 0.3
Theoretical	0.040 ± 0.005	0.030 ± 0.004	3.5 ± 1.1

Table 2. Experimental [26] and theoretical values for the z = 4 cm and z = 12 cm cases in the same experimental conditions for an argon plasma at atmospheric pressure

As the tables show, the widths and electronic density values expected are very close to the theoretical ones. Thus, we can affirm not only the proposed Stark profile model fits in good attendance with the previous models (with a significantly lower computational cost), but also allow us to identify the characteristic parameters of real plasmas with high precision.

4. Conclusions

An analytical model to fit a Stark profile, non-Lorentizan due to the influence of ion broadening, has been proposed. This model consists of the sum of Lorentzian profiles, and can be expressed as a function of the electronic density, the most important parameter that characterizes a real plasma.

Seven Stark theoretical profiles from CS model have been fitted using various numbers of Lorentzians, finding that the optimal number of Lorentzians is three. This number enable optimal ratio of the quality of fitting and the computing time.

The advantages of the proposed method are: 1) the possibility to express the non-Lorentzian Stark profile as a function of the electronic density and its simplicity; 2) the simultaneous calculation of three parameters at the same time, ω_G, ω_L and n_e. Both factors made of this new model a simple but sufficiently accurate tool to analyze real plasmas.

To that end, two real cases with previously known characteristic parameters have been studied. The agreement between the expected values and the ones obtained using the proposed method is acceptable, demonstrating the reliability and applicability of new method.

It must be remembered that the deduced functions in this paper are restricted to the considered ranges of density and temperature, but there are works in progress to obtain similar expressions in different ranges of applicability.

Acknowledgements

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