The Stark-crossing method for the simultaneous determination of the electron temperature and density in plasmas

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Abstract. The use of the Stark broadening of Balmer lines spontaneously emitted by atmospheric-pressure plasmas as a method to determine both the electron density and temperature in high-pressure plasmas is discussed in this paper. This method is applied to argon and helium plasmas produced in microwave discharges. Especially for Ar plasmas, valuable and reliable results are obtained.

1. Introduction

The basis of the diagnostic method determining at the same time \( n_e \) and \( T_e \) by Stark broadening rests on the idea of the study of two or more lines broadened under the same working conditions. The results of applying this diagnostic method to some cases were previously presented [1,2]. The first idea was raised earlier [3]. By superposing the information obtained for such lines, it is possible to determine not only \( n_e \), but also \( T_e \), in the discharge, which in the end provides a diagnostic technique that is a diagnostic equivalent to Thomson scattering. In fact, it is a first, rapid and approximated estimation of these magnitudes that can be sufficient for a large number of cases in which only a rough value of these magnitudes is needed. So, advantages and disadvantages are reasonably balanced in this discharge diagnostic method versus the more complicated, expensive and slow Thomson scattering. However, for some specific cases in which accuracy and precision in the measurements is needed, Thomson scattering will prevail over Stark broadening for the simultaneous determination of \( n_e \) and \( T_e \). In the future, this Stark broadening method must be contrasted with the Thomson scattering method.

In gaseous high-pressure discharges, microwave discharges have been used more and more because such discharges are very reproducible and stable, of easy handling and low cost in most cases. In this work we shall describe the experiments performed on the discharge produced by a TIA (Torch à Injection Axiale) device [4] using argon and helium at 2.45 GHz at moderated HF powers (300-1000 watts). In this way a microwave-torch flame is produced. It should be noted that in this kind of microwave discharge, we have a two temperature (2-T) plasma. The heavy particle (gas) temperature, \( T_{\text{gas}} \), is much less than the electron temperature, \( T_e \). In this way, the simultaneous measuring of \( n_e \) and \( T_e \) by Stark broadening of spectral lines is extended to a wider range of work conditions. Apart from discussing advantages and disadvantages of this novel technique we will also discuss the limitations.
that are for instance present in the case of He plasmas. In the future we will try to apply this technique to other type of plasmas. It is also possible to try our diagnostic method in high-pressure discharges other than microwave ones.

2. Brief review on the Stark broadening theories
The first statistical approach to explain the phenomenon, initially developed by Holtsmark [6], is a quasi-static theory. This theory mainly pays attention to the interaction of the ions over the light emitting atom with which give more broadening than the plasma electrons. Historically, different theories have been developed later to explain this broadening mechanism quantitatively better. Such theories took into account the influence of the static ions and also of the electron collisions. Some of these theories have a semi-empiric character and/or apply to some specific spectral lines. As an example, this was the case for the $H_{\beta}$ line of the hydrogen Balmer series. At present, the most important models for this line are the merely quasi-static approach due to Hill [7], as well as later approaches that also keep in mind the electron impact contribution to some extent, namely the GKS theory [8], the Greig’s theory [9], and the VCS theory [10].

For a measured Stark broadening of a line emitted by the plasma, the different theories explaining this broadening mechanism provide different values for the electron density $n_e$ at a given electron temperature $T_e$ in the discharge. If those values of $n_e$ are historically sorted, the result is a list of decreasing values. That is to say, the theoretical values are improved with each new theoretical contribution taking into account more and more causes of perturbation. In effect, by adding the perturbation effect of the plasma free electrons to the quasi-static theory, less density of ions will be necessary to produce the same effect (broadening) on the same profile shape.

More recent micro field model methods (MMM) take into account the ionic dynamics, which has supposed a new correction and improvement of the theoretical predictions compared with the experimental data. As was expected, by considering that the ions also move during the interaction time, the result obtained for the ion density is even lower. In this work we use the computational simulation theory due to Gigosos et al (or Gig-Card theory, [11]), which includes the more relevant processes in the plasma and constitutes one of the more accurate approximations at the moment. It becomes a method for calculating line shapes with the scheme of MMM, but with the electric micro field more correctly obtained from an ideal experiment [12]. In this approach the calculations have been made for three of the hydrogen Balmer series lines, namely $H_\alpha$, $H_{\beta}$ and $H_{\gamma}$, and their analogous lines of the Lyman series. This ion-dynamic correction has not the same relevance for each concrete line.

In short, the Gig-Card model is based on the computational numerical simulation of the behavior of all the particles in the plasma. The electric field created by the charged particles, electrons and ions, perturbing the emission of a hydrogen atom is obtained. The plasma is considered as a globally neutral, homogeneous and isotropic system in thermal equilibrium. Atoms, ions and electrons are moving (quasi-) randomly in the plasma, with rectilinear and uniform velocities given by the Maxwell-Boltzmann distribution. These particles move in a Debye sphere centered in the emitter whose size is related to the average distance between electrons, which is determined by the electron density. Inside this sphere of electric influence that surrounds the central (emitter) atom, the position (impact parameter) and the velocity (related to the temperature) of each particle with respect to the central hydrogen atom are calculated at each instant. In this situation, the time-dependent electric field $E(t)$ is obtained and introduced in the evolution quantum equations.

The habitual way to determine $n_e$ by Stark broadening is to experimentally measure the broadening of a line, and by knowing approximately the value of $T_e$ (usually thanks to some other complementary method of plasma diagnostics), to obtain the corresponding value of $n_e$. It is not easy to know $T_e$ in the discharge accurately and, as a consequence, the calculated values for $n_e$ are also affected by uncertainty. In some extent, the temperature is a free parameter in the conventional Stark broadening diagnostic method, i.e. once its value is fixed, $n_e$ is determined.
The broadening of different spectral lines depends differently on \( n_e \) and \( T_e \). Generally, the experimental broadening of different lines corresponds to slightly different electron densities depending on an arbitrary fixed \( T_e \). However, we shall show that is possible to obtain coherent results (same electron density) using different Balmer lines, by varying the free parameter (electron temperature). In this way, all the \( n_e \) values related to different broadenings of different spectral lines coincide at a specific \( T_e \). Finally we get a diagnosis of both quantities in the plasma simultaneously from these lines. The cross-lines diagnostic method of both the electron density and temperature that is proposed here lies in the use of two (or more) lines at the same time in order to look for a point (cross-point) in which the predicted electron density and temperature are coincident for these lines.

### 3. Experimental and diagnostic aspects

This spectroscopic method was already applied to a capillary argon plasma column produced by surfatron (SWD) [1], and to plasmas produced in argon and helium at atmospheric pressure by coupling microwave energy at 2.45 GHz with the aid of a TIA device [2]. This structure produces a plasma flame typically of one millimeter of diameter and a few centimeters long expanding in the open air. The electron temperature and density are considerably higher than in the surfatron plasma. As in the previous case, for the spectroscopic diagnostics of this discharge standard optical arrangements are used to increase the image and focus the light emitted by the brightest zone of the plasma flame onto the entrance slit of a THR 1000 (Jobin-Yvon) monochromator. At its exit slit, a double optical detection system is mounted, that alternatively permits us to use by means of a rotating mirror: a) an iCCD (intensified Coupled Charge Device, FlameStar II, LaVision), and b) a phototube (Hamamatsu), both in the visible region.

The light collected in the radial direction by the lens - that focuses an image of the plasma of about one millimeter diameter at the entrance slit - comes from different zones inside the plasma for a fixed axial position (see Figure 1). For a proper diagnosis of the discharge, this is in principle not a desirable feature spatially speaking due to the existence of relevant gradients in the discharge. The light coming from different points with spectral profiles having different widths is difficult to be treated. For that reason, the diagnostics presented here is representative (or apparent) for a \( z \) position in the plasma and not for any specific radial point inside the plasma at this \( z \). This position is measured from the beginning of the discharge (\( z = 0 \) at the tip of the TIA’s nozzle, as appears in Figure 2). It is also possible to move the system properly to make the diagnosis at different \( z \) using the light coming from different plasma positions (\( z \) heights). Even when radially resolved information is needed, this method is also suitable by using standard inversion techniques, for instance the Abel inversion, provided that cylindrical symmetry conditions are fulfilled by the discharge. This is the habitual situation when significant gradients exist in the discharge, although at the moment this improvement is not yet implemented in this experiment.

To properly observe the Balmer series lines, a small amount of about 1% hydrogen is introduced into the gas discharge. We can control the amount of hydrogen introduced into the discharge by using a flux meter (Tylan General, Dynamass Flow System). So, when we say “gas”, we mean plasmogenic gas (\( i.e. \) argon) plus a small amount of \( H_2 \). This amount is enough to measure the intensities of the Balmer series hydrogen lines, but does not disturb the discharge significantly. We can assume that our plasma is optically thin for radiation of the Balmer lines, so the light from Balmer lines may go through the whole plasma in its radial direction before it escapes the plasma. Self-absorption is not a problem for these hydrogen lines we are interested in because: \( i \) the hydrogen - due to its low concentration - is a negligible perturbation for the plasma, \( ii \) they are non-resonant lines and \( iii \) the light path is really short.
Results of the Stark broadening by the computational model arising from the Gig-Card theory are obtained at specific values of electron temperature, electron density and the reduced mass parameter $\mu$. The TIA plasma is typically a two-temperature (2-T) plasma: a higher temperature for the electrons - that receive the energy from the microwave field - and a lower temperature for the heavy particles (ions and atoms), that mainly receive the energy by electron collisional processes. This deviation from the thermodynamic equilibrium can be taken into account using the reduced mass as a parameter (i.e., if the electron temperature is twice the heavy particle temperature, a reduced mass two times higher must be used). In Figure 3, we can see this relationship in a 3-D representation in which the Stark broadening versus the electron density and the electron temperature is displayed for the case of $H_\alpha$ (at a given value of $\mu$). For a given electron temperature, we can fit the dependence of the Stark broadening on the electron density by using $\log n_e = a - b \ln(\Delta \lambda + c)$, where in fact the parameters $a$, $b$ and $c$ are functions of the electron temperature $T_e$. Of course, they behave differently with $T_e$ for each hydrogen line.

4. Some results and discussion

With the TIA device, experiments with two different plasmogenic gases were performed, namely argon and helium. As it has already been explained before, for both rare gases it has been necessary to add a controlled quantity of hydrogen to the discharge to make the used lines of the hydrogen Balmer series sufficiently visible. The gas flow remained fixed at one liter per minute with the addition of 1% of hydrogen. Two different positions in the discharge have been checked: 1) at approximately one centimeter above the tip of the TIA’s nozzle, and 2) just above this tip, in order to compare the results spatially. Since a radial study of the lines emitted by the plasma has not been carried out, the obtained measurements are apparent radial averages of the whole plasma in that position in which higher $T_e$ zones in the plasma are more important because there the emission are higher. Two different microwave powers, 600 and 800 watts, have been used in the experiments.
The theoretical dependence between Stark broadening $\Delta \lambda$, electron density $n_e$ and electron temperature $T_e$ is depicted in Figure 3. For an experimental measured $\Delta \lambda$, a horizontal plane is determined. The intersection between this plane and the color surface provides, after projection, a curve in the $n_e$ and $T_e$ plane (for the fixed Stark broadening) as appears in Figures 4, 5 and 6. By doing
this for two (or more) different spectral lines, two curves and thus two relations between \( n_e \) and \( T_e \) are found, and the intersection of these curves provides the \( n_e \) and \( T_e \) values (it is not a single point, but rather a little area of values, because experimental and theoretical uncertainties).

After repeatedly recording spectral profiles of \( H_\alpha \), \( H_\beta \) and \( H_\gamma \) lines under given experimental conditions (HF power and position in the flame), the different broadening mechanisms other than Stark broadening were eliminated from the total experimental profiles by using a simplified deconvolution method, where Lorentzian profiles were assumed for the Stark broadening and Gaussian profiles were assumed for the other contributions (thermal Doppler and instrumental broadening). By applying the Gig-Card theory to the Stark broadenings determined by this way, concerning the different studied lines of the hydrogen Balmer series, we have obtained tables of values for the electron density as a function of the electron temperature.

This has been possible for the three first Balmer series lines used in our study, but some problems have arisen with \( H_\gamma \). Due to its low intensity, we have had a very high scattering in the Stark broadening values of \( H_\gamma \) (that is because its profile is not sufficiently different from the background signal of the spectrum). The different measurements were not reproducible and they did not arise as a set of homogeneous broadening values. Due to this difficulty, at best we are able to obtain not a crossing point among these three Balmer series lines (which is the ideal situation), but rather a triangular-shaped crossing zone. This crossing zone has a very small area (see Figure 4) indicating a very low spread of the obtained result. But in other cases, this area is much larger and the results are not definitive by using three lines in the cross-point methods. In these same sets of measurements however, \( H_\alpha \) and \( H_\beta \) present a very stable behavior with hardly any scatter and provide a very stable value of the Stark broadening of the lines. At the moment, these two lines will provide the most useful combination for the simultaneous diagnostics of the electron density and temperature by means of this technique.

The most desirable would be to have a diagnostic based on a single-crossing of the three lines, which has been achieved in the particular case illustrated in Figure 4. But before, we should eliminate the great dispersion in the measurement of \( H_\gamma \). For that, we believe that it will be necessary to add more hydrogen to the discharge in future experiments in order to have a higher intensity of \( H_\gamma \). And furthermore, its profile would be better defined against the background signal.

![Figure 4](image-url)
4.1. Results on the TIA argon plasma

The results of the experiments on the TIA argon plasma are shown in Figures 5.a and 5.b at HF powers of 800 and 600 watts respectively. For the experimental full-width at half-maximum, FWHM, Stark broadening values obtained with $H_a$ and $H_\beta$ indicated in the legend of figures, the relationship between $n_e$ and $T_e$ are depicted as two different curves (position 1 cm approx. above the nozzle’s tip). As it is shown, the FWHM of $H_\beta$ is almost independent on the electron temperature. That is the main reason to use preferably $H_\beta$ if one is interested in the electron density value only. However, in the case of $H_a$, the influence of $T_e$ is more pronounced. So, in contrast to the $H_\beta$ case, the $H_a$ curve is not so horizontal. By crossing both curves, it is possible to find the values of $n_e$ and $T_e$ simultaneously at which the Stark broadenings predicted by the Gig-Card theory coincide with those obtained experimentally. This is the essence of the cross-point method. The results of the diagnostic for the electron density and the electron temperature in this case are $1.51 \times 10^{21} \text{ m}^{-3}$ and 21,100 K (800 watts), and $1.46 \times 10^{21} \text{ m}^{-3}$ and 20,800 K (600 watts) respectively. These are reasonable results compared with other works [13].
inferior in both \( n_e \) and \( T_e \) magnitudes for the lower power. An error of \( \pm 200 \) K in the electron temperature (and its corresponding error in density which is relatively lower) is estimated in this determination due to the fit process, the experimental dispersion in the measured FWHM values concerning \( H_\alpha \) and \( H_\beta \) profiles being very low. In addition, we also assume in this work the possible error inherent to the theory.

4.2. Results on the TIA helium plasma

For the case of the TIA helium plasma we have found an additional difficulty: for the conditions of interest, not only for the \( H_\beta \) but also for the \( H_\alpha \) case the influence of \( T_e \) is found to be limited. The values of the electron density and the electron temperature compatible with the measured Stark broadens concerning the involved lines are even less dependent than for the argon \( H_\beta \) case. Therefore, in the corresponding representations, we obtain curves quasi-horizontal and quasi-parallel to each other. To try to find a cross-point between these lines with such a behavior is very difficult.

As illustrative, the results for the TIA helium plasma at a microwave power of 600 watts (position 1 cm approx. above the nozzle’s tip) are represented in Figure 6, in an identical scale to that used in the TIA argon plasma. By comparing Figures 5 and Figure 6, the difficulty of obtaining a diagnostic cross-point is appreciated clearly. In He case, more appropriately we should speak of a wide temperatures range (19000-24000 K). However, this range supports results obtained by scattering Thomson [13] and furthermore, the representation determines with high precision the value of the electron density in the discharge, since a considerable coincidence exists between the values obtained by both \( H_\alpha \) and \( H_\beta \) lines. These experiments for the TIA helium plasma study were also carried out at two different positions, 1 cm approx. above the nozzle’s tip and just above this tip. For both spatial positions the same difficulty was found for different HF powers. By using the \( H_\gamma \) line in this set of experiments, we could not obtain satisfactory results (similarly to the argon case), because the TIA helium plasma was more unstable.

![Figure 6](image)

5. Conclusions

We have studied an experimental method to diagnose both the electron density and temperature simultaneously in high-pressure, microwave-excited plasmas. Compared with other methods of simultaneous diagnostics (Thomson scattering, for instance), this method is really simpler and low-cost, spectroscopic tools are used. It is based on an easy idea although there are assumptions about the
plasma equilibrium state and/or the distribution of the species existing in it, as well as the particle interaction responsible for the broadening mechanism. By studying simultaneously different spectral lines spontaneously emitted by the plasma in terms of their Stark broadenings, we have shown that it is possible to determine the two main magnitudes in the discharge (the electron density and the electron temperature) at the same time, according to the Stark broadening theories existing in the bibliography, namely one of the most recent (the Gig-Card theory). This theory takes the ion dynamics into account, whereas previous theories do not, and that fact affects and is reflected in the results.

The experiments have been carried out to diagnose two different discharges, namely the TIA plasma and the surfatron plasma, and have been satisfactory in both cases. The method is suitable under different working conditions and the results are consistent if they are compared with previous works, although a further comparison of this method with Thomson scattering should be made in the future. Hydrogen gas must be present in the discharge in order to make the Stark broadening of Balmer series lines intense enough. We have shown that the necessary amount of hydrogen gas is very low (around 1% of the main gas flux in the plasma discharge), and does not change the plasma conditions drastically.

It would be desirable to use more than two lines in order to gain coherence in the diagnostics, which could cause the existence of a small cross-region instead of a unique cross-point, but at the moment we have found some difficulties to obtain this in every case. This is an evident improvement in which we shall continue working on. In this article, it has been shown that such a possibility has been successfully performed in a particular case.

No other elements are so well studied as the Balmer series of hydrogen lines with respect to the Stark broadening of lines spontaneously emitted by plasmas. Therefore, the possibility of using any other lines (isolated argon or helium lines, for example) is discarded at present. So, we are restricted to hydrogen lines, but here we could have a future possibility with the ultraviolet Lyman series of hydrogen lines, namely the analogous Lyman-\(H_{\alpha}\), -\(H_{\beta}\) and -\(H_{\gamma}\) lines which are also well modeled by the Gig-Card theory.

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