Study of Lorentz and Gauss broadening of the argon lines with respect to the excited levels energy in a plasma at atmospheric pressure

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Abstract

We have analyzed the profile wavelength of several arg spectral lines respect to their broadening with the aim of finding a method to measure both the electron density and the gas temperature when the experimental conditions do not allow use the usual method. This method is based on using the Lorentzian and the gaussian broadening of the line profiles to measure these parameters. This one combined with a suitable set of spectral lines provides possibilities for reliable experimental results for electron density and later for the gas temperature.

1. Introduction

Surface-wave discharges are used in an increasing number of applications including, among others, spectrochemical analysis [1-3]. Such applications involve knowledge on the plasma processes, which ensure the atomic level excitation and, thus, require data for the plasma parameters. Two parameters, electron density (n_e) and gas temperature (t_g) control the kinetics of the plasma. The electrons are responsible for the energy transfer to the atoms by collisions. The gas temperature is a measure of the heavy particle energy.

Usually the electron density is determined from the stark broadening of the H_β hydrogen line. In many works the value of the gas temperature is obtained from the rovibrational spectra of the OH radical. Both the hydrogen atoms and the OH radicals are present in the plasma as impurities of the carrier gas. However, under certain experimental conditions the H_β line is not enough intensive and the OH radical is not a good thermometric species for determining gas temperatures below 1800 K [4]. In such situations it is reasonable to use the lines of the plasma neutral atoms and by measuring their broadening to determine n_e and t_g [5].

A spectroscopy diagnostics study of surface-wave-sustained discharges at atmospheric pressure in an argon gas is presented. The method applied here, and described in [6], is based on the analysis of the wavelength profile of several Ar I spectral lines respect to their broadening mechanisms.

2. Method of investigations

It is known that the shape of the emitted spectral lines is governed by the plasma processes: stark effect, van der waals interactions of heavy particles and also doppler effect connected with the heat motion of the particles. The influence of the natural spectral line broadening is negligibly small under the conditions of plasmas at atmospheric pressure. The registered spectral line profile is a convolution of two distributions: that of the light emitted from the plasma and the
instrumental one due to the detection system. When the monochromators are used, the profile depends on both the linear dispersion of the monochromator and the slit widths (entrance and exit). In some cases it can be approximated with a gaussian profile. The spectral line profile emitted from the plasma results a convolution of gauss and lorentz components (with full widths at half maximum $\Delta\lambda_g$ and $\Delta\lambda_l$ respectively), represented by a voigt function. The broadening $\Delta\lambda_{v}$ comes out from the doppler effect and, thus, $\Delta\lambda_{v} = \Delta\lambda_{d}$. The doppler broadening depends on the kinetic temperature and the atomic mass of the radiating atoms. The broadening $\Delta\lambda_{d}$ is due to the van der waals and stark broadening.

under our experimental conditions – low degree of ionisation and high neutral atom density – van der waals broadening plays the main role among the plasma broadening mechanisms. The interaction potential between excited and neutral atoms is presented according to the theory developed by hindmarsh et al. [7]. Here, we interpret our results with the aid of this potential and the hindmarsh formula [7] is used for obtaining the van der waals broadening width

$$2\gamma = 8.16 \, n \, (c/\hbar)^{3/5} \, v^{3/5} \quad (1)$$

Where $2\gamma$ (rad/s) is the full broadening, $n$ the number density of neutral ground state, with $\Delta\lambda_{w} = (\lambda^2/2\pi) \, 2\gamma$.

For the spectral lines involving high excited states i.e. Those corresponding to large values of the effective quantum number $n_{eff}$ it should be expected also a large broadening. Therefore, the spectral lines chosen for plasma diagnostic here correspond to the $3p^44p - 3p^5nd$ ($n = 4 - 7$) levels (table 1). According to [8-9], for the high values of the effective quantum number $n_{eff} \geq 5.5$ the potential involved in the formula for obtaining $\Delta\lambda_{w}$ width is not adequate for the description of the collisional effects. It is also known that in the case when the line profiles are registered with a monochromator, the value of the instrumental broadening $\Delta\lambda_{v}$ is of the order of $\Delta\lambda_{d}$. Thus, it is important to have a proper choice of spectral lines used for obtaining the plasma parameter.

<table>
<thead>
<tr>
<th>$\lambda$ (nm)</th>
<th>$3p^44p - 3p^5nd$</th>
<th>Level energy (cm$^{-1}$)</th>
<th>$N_{eff}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>737.212</td>
<td>4p - 4d</td>
<td>105436 - 119024</td>
<td>3.67</td>
</tr>
<tr>
<td>518.757</td>
<td>4p - 5d$^*$</td>
<td>104102 - 123373</td>
<td>4.61</td>
</tr>
<tr>
<td>603.213</td>
<td>4p - 5d</td>
<td>105463 - 122036</td>
<td>4.63</td>
</tr>
<tr>
<td>549.587</td>
<td>4p - 6d</td>
<td>105463 - 122036</td>
<td>5.62</td>
</tr>
<tr>
<td>522.127</td>
<td>4p - 7d</td>
<td>105436 - 124610</td>
<td>6.62</td>
</tr>
</tbody>
</table>

Table 1.

according to the griem's approximation, the stark broadening width $\Delta\lambda_{s}$ depends not only on the electron density, but also on the electron ($w$) and ion ($\alpha$) impact parameters (both in [10]), the stark broadening has been calculated using the approximated expression [10]

$$\Delta\lambda_{s} = 2w [1 + 1.75 \alpha(1-0.75)r] \quad (2)$$

1454
Where \( w \) is the electron impact broadening parameter customarily given as a half-\( \lambda \)-width, \( \alpha \) is the parameter for quasi-static broadening by ions and \( r = \rho_d / \rho_0 \) with \( \rho_0 \) is the mean inter-ion distance and \( \rho_d \) is the debye length. The stark profile can be approximated to lorentzian profile.

3. Experimental arrangement

The field applicator is a wave launcher of the waveguide surfatron type [11]. The microwave power (varied between 100 and 200 w) is provided by a 2.45 ghz generator (sairem gmp 12 ke/i), the reflect power is below 5%. The maximum length of the plasma column is 9 cm. The discharge produced in a tube with an inner diameter of 1 mm (4 mm o.d. Of a fused silica tube). The discharge gas is 99.99% pure argon, with a flow of 0.25 slpm; controlled by a flow meter.

An optical fiber is used for collecting the light emitted from the plasma at different axial positions \( z \) (denotes the end of the column). The light is analysed by a czerny-turner monochromator (jobin-yvon thr1000s) of 1 m focal length equipped with a 1200 grooves/mm holographic grating, the spectral resolution is 0.08 nm/mm. The monochromator slits used in the experiment are of the same order (100 \( \mu \)) width. The registered system includes a photomultiplier r636 sensitive for the region 200-930 nm.

4. Results and discussion

The spectral line profiles experimentally obtained are approximated with a symmetric voigt function, which is a convolution of gaussian (doppler and instrumental broadening) and lorentzian (stark and van der waals broadening). Then the procedure described in [6] is applied for obtaining \( t_e \) and \( n_e \).

We have found for all lines that full gaussian broadening is almost constant along the plasma column. Since the instrumental broadening does not depend on axial position \( z \), neither does the doppler broadening. This is in an agreement with the result for \( t_e \) obtained by using oh radical [12]. But the \( \Delta \lambda_e \) is equal to \( (2-3) \times 10^3 \) nm for \( t_e = 1400 \) k [12] and the fit error is the same order, thus it is not possible to use this method to measure the gas temperature at the moment. It is necessary to improve the spectroscopy techniques.

The results for the \( \Delta \lambda_e \) broadening of the lines with \( \lambda = 549.6 \) nm and \( \lambda = 603.2 \) nm obtained at different axial positions \( z \) and different values of the applied power \( p = 100, 150 \) and 200 w are presented in fig. 2(a,b). The \( \Delta \lambda_e \) values obtained for all lines from the table 1 show that \( \Delta \lambda_e \) increases with the \( n_{eff} \) increase (fig.3). A slight increase of \( \Delta \lambda_e \) towards the wave launcher is also obtained. The \( \Delta \lambda_e \) broadening of the \( \lambda = 522.1 \) nm \( (n_{eff} = 6.62) \) has been also measured. Although the highest values of \( \Delta \lambda_e \) obtained from this line are not used here for obtaining data for \( n_e \) since the data for \( w \) and \( \alpha \) are not found in the literature.

The van der waals broadening widths \( \Delta \lambda_w \) are calculated according to formula (1) for \( t_e = 1400 \) k. The errors of such obtaining are in the order of \( 15 - 20 \% \) [7]. The values of \( \Delta \lambda_w \) calculated for \( \lambda = 549.6 \) nm and \( \lambda = 603.2 \) nm are \( 2.7 \times 10^{-2} \) nm and \( 2.3 \times 10^{-2} \) nm, respectively. The van der waals broadening found for this line is also highest compared to the other lines \( (3.2 \times 10^{-2} \) nm). Therefore, this line should be used as a good test for future investigations, clarifying the mechanism which is the most important factor causing the broadening of the lines origin from the upper levels with an effective quantum number \( n_{eff} \geq 6 \).

The axial \( z \) dependants of the electron density obtained from the measurements for the spectral lines \( \lambda = 549.6 \) nm and \( \lambda = 603.2 \) nm are presented in fig. 4(a,b). The obtained data are for different applied power \( p = 100, 150 \) and 200 w. The data for \( t_e \) and \( t_e \) are taken from [12]. At the end of plasma column the electron density decreases as it is expected from the theory [13].

1455
The values measured by hρ measurements between (1.9 - 4.1)×10^{20} m^{-3} and smaller than those obtained with ari lines.

In conclusion the method presented here combined with a suitable set of spectral lines provides possibilities for reliable experimental results for electron density (and its axial variation) of surface wave sustained discharges at atmospheric pressure and it is possible later to can use its to measure the gas temperature.

Figure 2. A) Δλ_i broadening of 549.6 nm and b) Δλ_i broadening of 603.2 nm, both at different axial positions z and applied power (100, 150 and 200 w)
Figure 3. Variation of $\Delta \lambda_1$ broadening versus $n_{\text{eff}}$ for ari lines from the table 1.

Figure 4a: 
Ari 549.6 nm
- 100W
- 150W
- 200W

$n_{\text{eff}}$ vs. $z$ (cm)
Figure 4. The axial dependence (z) of the electron density obtained from different powers for a) \( \lambda = 549.6 \) nm and b) \( \lambda = 603.2 \) nm

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