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Stark broadening of Ar I spectral lines emitted in surface wave sustained discharges

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Abstract. Stark broadening parameters (widths and shift) of three Ar I spectral lines in pure argon: 522.1, 549.6, and 603.2 nm (nd → 4p, for n = 7-5) and 696.5 nm (4p'→ 4s) have been calculated within the semi-classical perturbation approach. The provided data, needed for example for the plasma diagnostics in surface wave sustained discharges, are given as a function of the temperature, for an electron density of 10^{14} cm^{-3}.

Key words. Atomic processes: Stark broadening – Stars: line profiles: Plasma: diagnostics – Discharges: surface waves

1. Introduction

Surface wave sustained discharges (SWDs) form a particular class of high frequency (HF) discharges, which have been successfully employed in various fields of science and technology, including materials processing, elemental analysis, spectrochemical applications, abatement of harmful gases, and more recently, sterilization of medical devices.

The broadenings of the spectral lines emitted in SWDs are poorly discussed in experimental and theoretical studies. The shape of a spectral line emitted (absorbed) by plasma is a convenient test for theoretical models of the interactions between the emitters and perturbers (neutrals and charged particles). One of the first works dedicated to the broadenings of spectral lines of SWDs (Moussounda & Ranson 1987) treated the broadening and shift of argon lines ($3p^5 4p \rightarrow 3p^5 4s$ transitions) in high-pressure plasmas. Under these conditions, the lines of the studied transitions are broadened principally by the neutrals. A successful modelling of the spectral profile of the argon 430.0 nm line ($3p^5 5p \rightarrow 3p^5 4s$ transition) at atmospheric pressure has been made in Dobreva et al. (1990). The electron density and the gas temperature have been obtained by the comparison between the simulated and registered line profiles. The results are in agreement with those deduced from other methods. A spectroscopic diagnostic method based on the detailed analysis of the profile of the Ar I 696.5 nm line excited in a non-stationary SWD has been presented in Christov et al. (1999) and Christova et al. (2000). In the pressure range of 20-140 Torr the discharge has a filamentary structure. Increasing the pressure, the evolution of the gas temperature and electron density has been obtained while the

Send offprint requests to: M. Christova discharge contracts into filamentary structures. The proposed method ensures perturbation-free access to plasma parameters, which is not the case when adding hydrogen and thermometric molecules to the discharge, even in a small amount. This method has been applied to determine the electron density and gas temperature of a microwave discharge at atmospheric pressure (Christova & Calzada 2000), (Christova et al. 2004a)

First of all, we are motivated here to study the interactions between the emitters and the perturbers. Second, we are interested in using Stark broadening of the argon lines to determine the electron density without any perturbation or contamination of the discharge. There are some peculiarities of the SWDs. On one hand, the higher energy levels are more densely populated in comparison with DC and RF discharges with the same density of power deposited into the plasma. The observed lines emitted from the high lying levels are more intense in microwave discharges even if the average electron energy is lower, compared to RF and DC discharges. On the other hand, the high energy levels are in particular thermal local equilibrium that favours the usage of the Stark broadening theory. A comprehensive study of the experimental Stark broadening published over the period of 30 years have been reported in Pellerin et al. (1996) yielding for the Ar I 696.5 nm line $\Delta\lambda_{St} = 0.0814$ nm ± 5.0% normalized to $n_e = 10^{17}$ cm^{-3} and $T = 13\,000$ K. The good agreement with the value of 0.0856 nm ± 30% calculated using S. Sahal-Bréchot theory and corresponding to the above n_e and T (Christova et al. 2004b; Christova et al 2004c) has encouraged us to continue the calculations of the Stark parameters of several argon lines.

In the present paper, we give results for Stark widths and shifts for three visible lines of argon that have not been calculated before for one of them, i.e. 522.1 nm.

2. Theory

Using the semiclassical perturbation formalism and the corresponding computer code (Sahal-Bréchot 1969a,b), which was updated and optimized several times (see Dimitrijević & Sahal-Bréchot (1996) and Ben Nessib et al. (1996), as well as references therein), we have calculated the full half width (W) and the shift (d) of the line emitted between the initial level i and the final level f by the following formulae (Sahal-Bréchot 1969a,b):

$$W = 2n_e \int v f(v) dv [\sum_{k \neq i} \sigma_{ik}(v) + \sum_{j \neq f} \sigma_{fj}(v) + \sigma_{el}] \quad (1)$$

$$d = n_e \int v f(v) dv \int_{R_3}^{R_d} 2\pi\rho d\rho \sin 2\phi_p, \quad (2)$$

where k and j are the perturbing levels, n_e and v are respectively the electron density and the velocity of perturbers, and $f(v)$ is the Maxwellian distribution of the electron velocities.

The inelastic cross sections $\sigma_{ik}(v)$ (respectively $\sigma_{fj}(v)$) can be expressed by an integration of the transition probability P_{ik} over the impact parameter ρ as

$$\sum_{k \neq i} \sigma_{ik}(v) = \frac{1}{2}\pi R_1^2 + \int_{R_1}^{R_d} 2\pi\rho d\rho \sum_{k \neq i} P_{ik}(\rho, v). \quad (3)$$

The elastic collision contribution to the width is given by:

$$\sigma_{el} = 2\pi R_2^2 + \int_{R_2}^{R_d} 8\pi\rho d\rho \sin^2 \delta \quad (4)$$

$$\delta = (\phi_p^2 + \phi_q^2)^{1/2}. \quad (5)$$

The phase shifts ϕ_p and ϕ_q are due to the polarization and quadrupole potential respectively (Sahal-Bréchot 1969a,b). The cut-offs R_1, R_2, R_3, the Debye cut-off R_d and the symmetrization procedure are described in Sahal-Bréchot (1969a,b).

The impact approximation is valid when the strong collisions are separated in time, or when the duration of collisions is much shorter than the separation time between strong collisions: $n_e\pi\rho_{typ}^3 << 1$, where ρ_{typ} is the typical impact parameter for strong collisions. This condition is well fulfilled by the electronic collisions in a wide range of densities. For ionic

Table 1. Present calculations for the transitions 7^3F^o - 4^3D, 6^3F^o - 4D, and 5^3F^o - 4^3D (in Tables 1-3, respectively) of argon - T: electronic temperature in K, electronic density is $10^{14}cm^{-3}$, - W and d: impact width and shift in Å.

		Ar I 522,1 nm	7^3F^o-4^3D			
T =	2500 K	5 000 K	10 000 K	20 000 K	30 000 K	50 000 K
W, Å	0.804e-1	0.886e-1	0.992e-1	1.137e-1	1.202e-1	1.255e-1
d, Å	0.571e-1	0.588e-1	0.515e-1	0.431e-1	0.393e-1	0.343e-1
C_e^1	0.538e-2	0.373e-2	0.265e-2	0.196e-2	0.158e-2	0.113e-2
C_e^2	0.556	1.002	1.655	2.148	2.382	2.583
C_e^3	308	427	436	352	299	245
C_e^4	0.427	0.390	0.361	0.319	0.284	0.247
C^5	0.012	0.013	0.015	0.017	0.018	0.019
C_i^1	0.184	0.123	0.833e-1	0.568e-1	0.455e-1	0.346e-1
C_i^4	0.115	0.115	0.114	0.110	0.110	0.114

Table 2.

		Ar I 549,6 nm	6^3F^o-4^3D			
T =	2500 K	5 000 K	10 000 K	20 000 K	30 000 K	50 000 K
W, Å	0.437e-1	0.479e-1	0.531e-1	0.606e-1	0.645e-1	0.675e-1
d, Å	0.312e-1	0.321e-1	0.299e-1	0.252e-1	0.229e-1	0.203e-1
C_e^1	0.184e-2	0.126e-2	0.879e-3	0.649e-3	0.526e-3	0.382e-3
C_e^2	0.481	0.875	1.470	1.984	2.216	2.435
C_e^3	137	194	201	164	141	117
C_e^4	0.432	0.393	0.360	0.325	0.293	0.254
C^5	0.004	0.004	0.005	0.005	0.006	0.006
C_i^1	0.648e-1	0.433e-1	0.291e-1	0.198e-1	0.158e-1	0.120e-1
C_i^4	0.116	0.116	0.116	0.112	0.112	0.116

Table 3.

		Ar I 603,2 nm	7^3F^o-4^3D			
T =	2500 K	5 000 K	10 000 K	20 000 K	30 000 K	50 000 K
W, Å	0.222e-1	0.243e-1	0.267e-1	0.302e-1	0.322e-1	0.341e-1
d, Å	0.164e-1	0.171e-1	0.160e-1	0.136e-1	0.127e-1	0.113e-1
C_e^1	0.500e-3	0.341e-3	0.234e-3	0.171e-3	0.140e-3	0.103e-3
C_e^2	0.419	0.763	1.304	1.817	2.036	2.282
C_e^3	51.944	74.895	78.915	65.460	56.410	47.244
C_e^4	0.438	0.400	0.359	0.331	0.301	0.262
C^5	9.8e-4	1.1e-3	1.2e-3	1.2e-3	1.4e-3	1.5e-3
C_i^1	0.184e-1	0.122e-1	0.818e-2	0.553e-2	0.441e-2	0.333e-2
C_i^4	0.118	0.118	0.118	0.115	0.114	0.117

collisions the impact approximation fails, especially for high densities, that is not our case. 3. Results

3. Results

The calculations have been made for a grid of temperatures (10^4 to 10^5 K) and electron density of 10^{14} cm^{-3}. The results are displaced in Tables 1-3 for three studied argon neutral spectral lines 522.1 nm, 549.6 nm, and 603.2 nm, corresponding to multiplet transitions in LS coupling: 7^3F^o - 4^3D, 6^3F^o - 4^3D and 5^3F^o - 4^3D. We give the calculated impact width W and shift d for collisions with electrons (We, de) and Ar+ ions (Wi, di). We use the TOPbase data: energy and oscillator strength values. We give also the various validity criterions useful for the following discussion. The validity criterion for the impact approximation is given by:

$C_1 = \tau W_{strong} << 1$,

where τ is typical collision duration and W_{strong} is the strong collision contribution to the collisional width.

$C_2 = W_{in}/W_{el}$ gives the ratio of the contribution of inelastic collisions versus elastic ones for the width.

C_3 is the criterion for the "one state" approximation validity.

C_4 is the ratio of the contribution of strong collisions to the total impact width and thus gives a validity criterion for the perturbation theory approximation.

C_5 is the criterion for the isolated line approximation, defined by Dimitrijević & Sahal-Bréchot (1984) and Baranger (1958).

$(\Delta E_{ii'})_{min}$ is the energy distance between the upper level and the nearest perturbing level. The impact criterion is satisfied, the resulting profiles are lorentzian, and the total widths and shifts are given by:

$W = W_e + W_i;\ d = d_e + d_i$

The inelastic collisions by the electrons dominate over elastic ones for the temperatures closed and greater then 10 000 K for the studied transitions. The ionic collisions are entirely elastic. This is the reason why we don't include in the Tables 1-3 the ratio C_2 and the criterion C_3 concerning the ions. The "one state approximation" is valid for the three transitions since the contribution of the collisions with upper state is dominant. The validity criterion for the perturbation theory approximation is satisfied for the both types of collisions, i.e. electron and ion collisions. The studied argon neutral spectral lines are isolated. The positive red shift is obtained for the examined spectral lines. It decreases with the temperature.

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