PHYSICAL REVIEW E VOLUME 51, NUMBER 1 JANUARY 1995

Search for ion dynamics effects on the shift and width of plasma-broadened C I and O I spectral lines

Z. Mijatović
Institute of Physics, Trg Dositeja Obradovića 4, 21000 Novi Sad, Yugoslavia

N. Konjević
Institute of Physics, P.O. Box 57, 11001 Belgrade, Yugoslavia

R. Kobilarov and S. Djurović
Institute of Physics, Trg Dositeja Obradovića 4, 21000 Novi Sad, Yugoslavia

(Received 14 March 1994; revised manuscript received 9 June 1994)

We report measured Stark shifts and widths of the neutral carbon 5052- and 4932-Å lines and the neutral oxygen 4368-Å line in the plasma of an atmospheric pressure wall stabilized electric arc. Electron densities of $(1.4-3.1)\times10^{16}$ cm^{-3} are measured from the width of the H_β line while electron and gas temperatures in the range from 9300 to 10 300 K are determined from plasma composition data. Experimental Stark widths and shifts are compared to theoretical results obtained from semiclassical calculations of Stark broadening parameters using quasistatic and ion-dynamic treatment of the ions. Some indications are found that the inclusion of ion dynamics in the calculation of the shifts of the C I 5052-Å line slightly improves the consistency between theory and experiment.

PACS number(s): 52.70.Kz, 32.70.Jz

I. INTRODUCTION

Study of the influence of ion dynamics to the shape and shift of plasma broadened spectral lines has been concentrated to hydrogen and hydrogenlike ions (see e.g., Ref. [1] and references therein). Only a few papers are devoted to the study of its influence to nonhydrogen spectral lines where the effect is generally smaller and easier to calculate. A method for calculating ion-dynamic effects to the nonhydrogenic atom lines are developed by Griem [2] and Barnard, Cooper, and Smith [3]. A summary of these theoretical approaches is included in an experimental work on helium [4] while a recent paper by Kobilarov, Konjević, and Popović [5] is devoted to the investigation of the influence of ion dynamics to the helium lines. Furthermore, it has been shown [5] that inclusion of the ion-dynamic effects in the comparison between experimental data and results of the Stark broadening theories for He I lines could explain systematic discrepancies detected at lower electron densities and in this way opens new possibilities for high precision electron-density-plasma diagnostics.

The aim of this work is to extend the study of the influence of ion dynamics to the width and shift of isolated spectral lines of elements heavier than helium. Carbon and oxygen are selected for a number of reasons. First, in comparison with helium plasma there is a considerable difference in the mass of the ion perturber, second, gaseous compounds for these elements are available so their introduction in the plasma can be easily and precisely controlled and finally, other high quality experimental data are available for comparison (see, e.g., Ref. [6] and references therein).

II. THEORY

The details of theoretical approaches used for evaluation of plasma-broadened line shapes and shifts are given elsewhere [2–5]; to facilitate the discussion only final formulas will be given here.

A. Quasistatic treatment of ions [2]

Full half width is

$$w_{\text{stat}}=2w_e(1+gA_N)N_e10^{-16} \tag{1}$$

and shift at the half width

$$d_{\text{stat}1/2}=(d_e\pm3.20A_Ng_1w_e)N_e10^{-16}\ , \tag{2}$$

where $g=1.75(1-0.75R)$, $g_1=g/1.75$, and $A_N=AN_e^{1/4}10^{-4}$. In the above equations w_e and d_e are the electron-impact half-halfwidth and shift in angstrom units, respectively, and A is an ion-broadening parameter. All three quantities are for electron density $N_e=10^{16}$ cm^{-3}. Debye shielding parameter R is given by

$$R=0.090N_e^{1/6}T_e^{-1/2}\ ,$$

where T_e is the electron temperature.

B. Dynamic treatment of ions [3–5]

Full half width [3,4] is

$$w_{\text{dyn}}=2w_e(1+gA_nW_j)N_e10^{-16}\ , \tag{3}$$

where

$$W_j=\begin{cases}1.36B^{-1/3}/g\ , & B<(1.36/g)^3\\ 1\ , & B\geq(1.36/g)^3\end{cases}$$

and

$$B=A_N^{1/3}(0.0806w_e/\lambda^2N_e^{2/3})(\mu/T_g)$$

with atom-ion perturber reduced mass μ in amu and gas temperature T_g.

Shift at the half-width [3–5] is

$$d_{\mathrm{dyn}1/2}=(d_e\pm3.20A_Ng_1D_jw_e)N_e10^{-16}\ , \tag{4}$$

where

$$D_j=\begin{cases}(2.35B^{-1/3}-3A_N^{1/3}R)/g_1\ , & B<1\\ 1\ , & B\geq1\ .\end{cases}$$

There are certain restrictions on the applicability of Eqs. (1)–(4) and they are [2]

$$R\leq0.8\ ,\quad 0.05\leq A_N\leq0.5\ . \tag{5}$$

For the evaluation of the theoretical Stark widths and shifts of C I and O I lines from Eqs. (1)–(4), w_e, d_e, and A are taken from Ref. [2] while electron and ion temperatures for our plasma conditions were taken equal, $T_e=T_i$. For the calculation of atom emitter-ion perturber reduced mass μ plasma composition data were used and the influence of all singly ionized species were taken into account in the following way:

$$1/\mu=1/M_{\mathrm{emitter}}+\frac{\sum_{i=1}^{n}N_i/M_i}{\sum_{i=1}^{n}N_i}\ , \tag{6}$$

where M_i is the mass of the perturbing ion and $\sum_{i=1}^{n}N_i=N_e$. The latter is a reasonable assumption for an atmospheric pressure low current arc plasma where only singly ionized atoms are detected.

III. EXPERIMENTAL APPARATUS AND PROCEDURE

A. Plasma source

For the plasma source we used an atmospheric pressure wall stabilized electric arc. The arc consists of a stack of 7.1 mm thick water-cooled copper plates separated by 0.5 mm thick insulating Teflon gaskets. The diameter of the arc channel is 5 mm and its length is 70 mm. Argon gas with the flow rate of 3 l/min is introduced into the arc from both ends in such a way that both tungsten electrodes are burning in an inert atmosphere. The gas mixture is introduced in the middle part of the arc, see Fig. 1. The exhaust pipes are placed between the arc center and electrodes, so the gas mixture leaves the arc channel before reaching the near-electrode region. In this way the effect of cold layers, which could influence spectral line shapes, is avoided. For C I and O I spectral lines recording mixtures of H_2:Ar:CO_2 (4:64:32) and H_2:Ar:O_2 (4:66:30) were used, respectively. The flow rate of gas mixtures was approximately 0.03 l/min. The current, in the range from 19 to 30 A, was supplied to the arc by a current-stabilized power supply with stability of 0.3%.

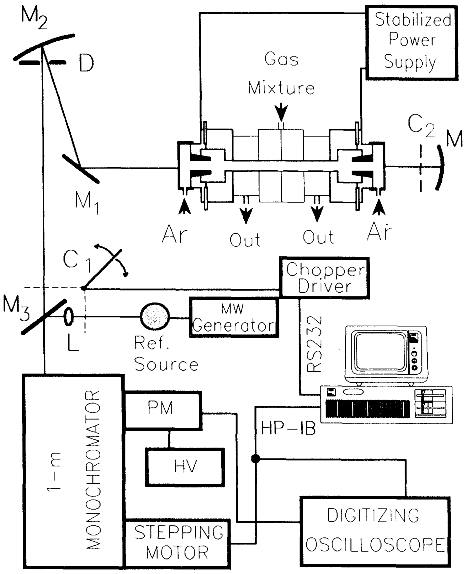

FIG. 1. Schematic diagram of the experimental setup.

B. Line shape and shift recordings

The plasma observations were performed end-on through a 5 mm hole in the electrode. The central part around the arc axis is imaged 1:1 onto the entrance slit of the 1 m monochromator by means of the concave 1 m focal length, focusing mirror M_2, see Fig. 1. A 10 mm diaphragm D placed in front of the focusing mirror ensures that light comes from a narrow cone about the arc axis. The concave mirror M_4 placed two focal lengths from the center of the arc is used along with the light chopper C_2 for the line self-absorption testing (see, e.g., Ref. [7]). A low pressure lamp with a microwave excitation was used as a source of unshifted spectral lines. For C I spectral lines a low pressure neon lamp was used while for the shift of O I 4368.25 Å a low pressure continuous flow oxygen discharge was used. The neon lamp contained a certain amount of impurities in the form of carbon compound so with microwave excitation C I 5052.17 Å is detected (see Fig. 2) and used for the shift measurements of the same line emitted from the arc plasma. For C I 4932.05 Å, the Ne I 4930.944-Å spectral line is used as a reference line. Wavelengths of investigated C I and O I lines are taken from Ref. [8] while the wavelength of the Ne I line not listed in [8] was taken from Ref. [9]. For the shift measurements, the lights from the arc plasma and from the reference source are focused onto the slit of the monochromator, see Fig. 1. The mirror M_3 is partially reflecting so the light from both, arc plasma and reference source have the same optical path from the mirror M_3 to the exit slit of the monochromator. Using the

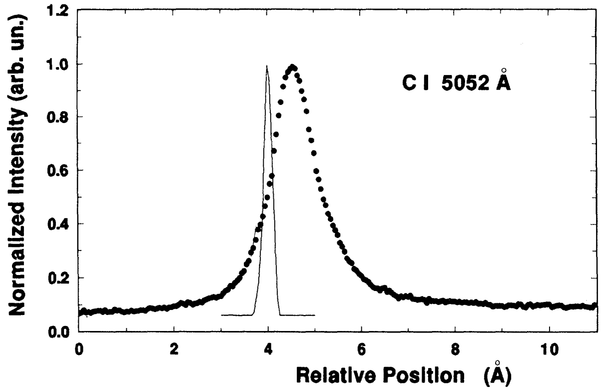

FIG. 2. Simultaneously recorded and normalized to the same peak intensity C I 5052.17-Å line profiles from the arc plasma (broadened profile) and from the reference source.

chopper C_1 light could be obtained from the arc or from a reference source alternatively. Two He-Ne lasers, one positioned behind the arc the other one directed through the exit slit of the monochromator, are used to align the arc and the reference source.

The monochromator with the inverse linear dispersion of 8.33-Å/mm is equipped with the photomultiplier tube and stepping motor (36 000 steps/rev). Signals from the photomultiplier were led to the digitizing oscilloscope working in the averaging mode (32 samples over 200 ms for each sample). The stepping motor (HP-IB interface), chopper (RS-232 interface) and oscilloscope (HP-IB interface) are controlled by the personal computer. The same computer was used for data acquisition.

With the slits of the monochromator not exceeding 15 μm all spectral line profiles recorded from the reference source had a Gaussian form so, in order to determine the center of the unshifted line profile, they were fitted by least square method to the Gaussian. The spectral line profiles from the arc plasma were also treated by the computer program developed for deconvolution of Gaussian and plasma broadened neutral line profiles $j_{A,R}(x)$ [10]. Shift of plasma-broadened lines was always measured at the half width. Before the line shape measurements, the experimental test for the possible distortion of the profile caused by the self absorption was carried out. Under the present experimental conditions all investigated spectral lines were found optically thin. An example of simultaneously recorded spectral line profiles from the arc plasma and from the reference source is given in Fig. 2.

IV. PLASMA DIAGNOSTICS

An electron density in the range $(1.45-3.12)\times 10^{16}$ cm^{-3} at the arc axis was determined from the width of the Balmer H_β line in conjunction with theoretical calculations by Vidal, Cooper, and Smith [11]. Electron temperatures were deduced from the plasma composition data. For this purpose we used the values of measured electron densities and plasma composition data which were evaluated for our experimental conditions using a procedure described by White, Jonson, and Dantzig [12]. Plasma composition data were also used for the estimation of the influence of various ions present in the arc plasma to the width and shift of investigated C I and O I lines, see Eqs. (3)–(5).

V. RESULTS AND DISCUSSION

Experimental results for the shift and width of investigated C I and O I lines are given together with other experimental data in Tables I–III. Experimental results with an estimated uncertainty ±30% or better (marked by code letter B in Refs. [6,7] and [13]) are taken for the comparison only. Estimated uncertainty of reported electron densities and temperatures do not exceed ±9% and ±3%, respectively. Stark widths w_m and shifts d_m, in Tables I–III are determined with an accuracy ±4% and ±3%, respectively. In the case of the C I 4932.05-Å line shift measurements are performed in respect to the spectral line of another element—neon—so the estimated uncertainty is in the range of ±4%. In two columns, following experimental results in Tables I and II ratios of measured and theoretical widths and shifts calculated using the quasistatic w_m/w_{stat} and d_m/d_{stat} [Eqs. (1) and (2)] and dynamic ion approximation, w_m/w_{dyn}, and d_m/d_{dyn}, [Eqs. (3) and (4)] are given. Parameters W_j and D_j [see Eqs. (3) and (4)] are given in these tables also. Since in the case of O I 4368.25-Å line ion-broadening parameter A_N over the whole electron density range for all experiments in Table III is smaller than 0.05 [see condition (5)] ion-broadening contribution is excluded from theoretical data. So in this table

$$w_{\mathrm{stat}}=2w_eN_e10^{-16}\ , \quad d_{\mathrm{stat}}=d_eN_e10^{-16}\ , \tag{7}$$

see Eqs. (1) and (2). For the same reason w_{dyn} and d_{dyn} were not calculated as well.

Here one should notice that correction of the theoretical half-widths in cases when $A_N<0.05$ can be evaluated by adding a "quadrupole" ion impact width as suggested by Griem [Eq. (218b) in Ref. [2]]. Since, in this way, evaluated correction for the ion widths is very large (about 25% of the electron impact widths), while typical ion-broadening contribution for isolated nonhydrogenic atom lines does not exceed 10–15% at the most, we have neglected the ion contribution to the widths for this particular line. Furthermore, for all investigated lines the influence of the Debye shielding effect on the linewidths and line shifts was found negligible.

Although reported C I and O I Stark widths and shifts in Tables I–III are high accuracy data, the uncertainty in electron density determination introduces relatively large errors in w_m/w_{theor} and d_m/d_{theor} so the estimated uncertainties of these ratios in Tables I–III range between 13% and 16%. Here only experimental errors are taken into account. The ratios of the experimental widths and shifts to the corresponding theoretical values are used for both, as a measure of the agreement with the theory and for the intercomparison of the experimental results measured in plasmas with different electron densities and

TABLE I. Experimental widths w_m and shifts d_m for C I 5052.17-Å line. Ratios of measured and calculated widths using static- $w_{\rm stat}$ and $d_{\rm stat}$ and dynamic-ion approximation $w_{\rm dyn}$ and $d_{\rm dyn}$ are given. Data for theoretical calculations are taken from Griem [2]. Corresponding ion broadening parameters W_j and D_j evaluated from Eqs. (3) and (4), respectively, are given also.

N_e (10^{16} cm^{-3})	T (K)	w_m (Å)	$w_m/w_{\rm stat}$	$w_m/w_{\rm dyn}$	W_j	d_m (Å)	$d_m/d_{\rm stat}$	$d_m/d_{\rm dyn}$	D_j	Ref.
1.42	9 320					0.29	1.10	0.95	2.19	This
1.85	9 670	0.67	0.99	0.94	1.65	0.37	1.07	0.94	2.04	work
2.15	9 870	0.79	1.01	0.96	1.60	0.43	1.06	0.93	1.95	
2.20	9 890	0.82	1.01	0.97	1.60	0.43	1.03	0.91	1.94	
2.20	9 890	0.80	0.99	0.95	1.60	0.43	1.04	0.92	1.94	
2.55	10 110	0.93	0.99	0.95	1.55	0.49	1.02	0.91	1.85	
2.85	10 280	1.06	0.99	0.95	1.51	0.55	1.01	0.91	1.78	
2.90	10 310	1.06	0.98	0.94	1.51	0.56	1.01	0.91	1.77	
0.91	9 000	0.36	1.14	1.07	1.88	0.10	0.57	0.48	2.46	[14]
1.65	10 000	0.57	0.94	0.90	1.70	0.27	0.87	0.76	2.10	
3.40	10 800	1.14	0.88	0.85	1.47	0.54	0.83	0.75	1.70	
5.96	11 600	1.94	0.84	0.81	1.33	1.01	0.87	0.81	1.42	
7.93	11 600	3.08	1.00	0.97	1.28					[15]

TABLE II. Same as for Table I but for the C I 4932.05-Å line.

N_e (10^{16} cm^{-3})	T (K)	w_m (Å)	$w_m/w_{\rm stat}$	$w_m/w_{\rm dyn}$	W_j	d_m (Å)	$d_m/d_{\rm stat}$	$d_m/d_{\rm dyn}$	D_j	Ref.
2.20	9 890	1.05	0.81	0.79	1.31	0.59	0.82	0.76	1.46	This
2.85	10 280	1.35	0.80	0.78	1.25	0.72	0.77	0.73	1.34	work
0.91	9 000	0.42	0.82	0.78	1.54	0.21	0.73	0.65	1.92	[14]
1.65	10 000	0.83	0.86	0.83	1.37	0.45	0.84	0.77	1.61	
3.40	10 800	1.40	0.68	0.67	1.21	0.83	0.74	0.74	1	
5.96	11 600	2.34	0.64	0.63	1.10	1.56	0.78	0.78	1	
7.93	11 600	3.90	0.79	0.79	1.06					[15]

TABLE III. Experimental widths w_m and shifts d_m for O I 4368.25-Å line. Ratios of measured and calculated widths $w_{\rm stat}$ and shifts $d_{\rm stat}$ [see Eq. (7)] are given. Data for theoretical calculations are taken from Griem [2].

N_e (10^{16} cm^{-3})	T (K)	w_m (Å)	$w_m/w_{\rm stat}$	d_m (Å)	$d_m/d_{\rm stat}$	Ref.
2.40	10 600	0.51	1.29	0.10	1.17	This
3.12	10 980	0.67	1.28	0.13	1.17	work
5.70	12 080	1.08	1.10	0.20	0.98	[16]
1.91	10 100	0.39	1.27			[14]
2.99	10 900	0.58	1.15			
4.94	11 900	1.07	1.24			
7.13	12 700	1.57	1.25			
3.97	11 580	0.78	1.14	0.12	0.85	[17]
7.84	12 500	1.44	1.04	0.27	0.97	
4.41	13 570	0.82	1.02			

electron temperatures.

Comparison between three sets of the experimental results for the width of the C I 5052.17-Å line in Table I (this experiment and data from [14] and [15]) show, within the uncertainty of the experimental results, agreement with the semiclassical theory [2]. However the agreement of our results with [15] is much better and discrepancy of w_m/w_{stat} does not exceed $\pm 2\%$. The results from [14] show large variations of w_m/w_{stat} which are even larger for the shifts, see d_m/d_{stat} in Table I. It is very difficult to trace the cause of the large variations of w_m/w_{stat} and d_m/d_{stat} in [14]. The flow of the test gas through the end region of the arc (plasma observations were performed end-on in [14]) may cause distortions of the line profiles and influences line shift measurements.

In the case of the C I 4932-Å line, see Table II, similar conclusions as for the C I 5052-Å line may be drawn. Again our results agree better with [15] and large variations of w_m/w_{stat} in [14] exist. This time, however, our shifts results, d_m/d_{stat}, agree better with [14].

Our experimental results for the O I 4368-Å line are in agreement within the uncertainty of the experimental measurements) with three other experiments [14,16,17] and with the theory [2]. For the evaluation of w_m/w_{stat} and d_m/d_{stat} for this line contribution of the ion broadening is not taken into account and this may influence the results of the comparison but this influence should not exceed a few percent. One can draw similar conclusions for the shift results in Table III.

Comparison of the ratios of the experimental Stark widths and shifts with theoretical values evaluated using quasistatic and ion-dynamic approximation in Tables I and II differ only a few percent. The only exception is C I 5052-Å line where the use of ion dynamics in evaluation of the shifts results in $\simeq 10\%$ larger theoretical values. While the average ratios for the static w_m/w_{stat} and dynamic widths w_m/w_{dyn} are 0.99 and 0.95, respectively, standard deviation is 0.011 in both cases. In case of the shifts the average ratios are $d_m/d_{\text{stat}} = 1.04$ and $d_m/d_{\text{dyn}} = 0.92$ with the standard deviation of 0.032 and 0.016, respectively. It is also important to notice that d_m/d_{stat} gradually changes from 1.01 to 1.10 and the agreement with quasistatic theory becomes worse at the lower electron densities (larger D_j). So in spite of the fact that the average agreement of the experiment with quasistatic results is better the comparison with the theoretical results which include dynamic treatment of ions shows more consistency.

In order to support the above conclusions we performed a search through the literature (see, e.g., Refs. [6,7] and [13]) and some illustrative examples for Ar I [18] and N I [19] spectral lines are discovered, see Table IV. Again experimental shift ratios d_m/d_{dyn} measured at different electron densities and temperature show more consistency while the ratios with quasistatic theoretical results d_m/d_{stat} show the tendency of the gradual increase with the decrease of the electron density. This is in agreement with ion-dynamic theory [2,3] which predicts at lower electron densities larger effects of the ion motion to the linewidths and in particular to the line shifts. So, in the estimation of the influence of ion dynamics to the Stark width and shift, it is essential to measure line parameters at low and high electron densities. Consistency of the comparison with the theory in the whole region of the plasma parameters is a more important indication of the validity of the theory than the absolute agreement with the theoretical results which depend, for example, upon the completeness and accuracy of the set of perturbing levels taken in the calculations of the linewidths and, in particular, for line shifts.

VI. CONCLUSIONS

In this paper we present results of the measurement of Stark widths and shifts of C I 5052.17- and 4932.05-Å and O I 4368.25-Å lines in the plasma of an atmospheric pressure wall stabilized electric arc. The experimental results are compared with the data from semiclassical calculations by Griem [2] using quasistatic [2] and ion-dynamic [3] approximations. It seems that comparison of the experimental results for the shift of the C I 5052.17-Å line shows slightly better consistency if ion-dynamic contribution to the Stark shift [3] is taken into account, see ratios d_m/d_{dyn} in Table I. Some other experimental results performed independently, see Table I, support our results for the shift of the C I 5052.17-Å line. Therefore we may conclude that, within the limits of the uncertainty of our

TABLE IV. Same as for Table I but for the Ar I 4272.17-Å (Ref. [18]) and N I 4935.12-Å line (Ref. [19]).

Spectral line	N_e (10^{16} cm^{-3})	T (K)	w_m (Å)	d_m (Å)	w_m/w_{stat}	w_m/w_{dyn}	W_j	d_m/d_{stat}	d_m/d_{dyn}	D_j	Ref.
Ar I	1.20	9 750	0.20	0.12	0.78	0.75	1.56	0.97	0.86	1.97	[18]
4272 Å	2.47	10 560	0.44	0.24	0.81	0.79	1.37	0.92	0.85	1.61	
	4.70	11 430	0.80	0.44	0.76	0.75	1.23	0.88	0.88	1	
	7.30	12 150	1.31	0.65	0.78	0.77	1.13	0.82	0.82	1	
	9.40	12 700	1.69	0.89	0.75	0.74	1.07	0.85	0.85	1	
N I	5.9	11 700	1.18	−0.20	0.88	0.86	1.63	0.78	0.63	2.00	[19]
4935 Å	16.4	14 640	3.50	−0.46	0.85	0.84	1.32	0.64	0.64	1	

experimental results and in the range of plasma parameters covered in this experiment, there are some indications that the Barnard, Cooper, and Smith theory [3] correctly describes the dynamic contribution of the ions to the Stark shift.

ACKNOWLEDGMENT

The authors gratefully acknowledge Dr. Ivanka Antunović Holclajtner for the evaluation of plasma composition data.

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