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Vladimir Milosavljevic
Technological University Dublin, vladimir.milosavljevic@tudublin.ie

Vida Zigman
Nova Gorica Polytechnic, Nova Gorica, Slovenia

Stevan Djenize
University of Belgrade,Serbia

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Stark width and shift of the neutral argon 425.9 nm spectral line

V. Milosavljević[a,*], V. Žigman[b], S. Djeniže[a]

[a]Plasma Physics, Faculty of Physics, University of Belgrade, Studentski Trg 14-16, P.O. Box 368, Belgrade 11000, Serbia and Montenegro
[b]Nova Gorica Polytechnic, P.O. Box 301, Nova Gorica, Slovenia

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Abstract

The Stark parameters, the width (W) and the shift (d), of the neutral argon (Ar I) 425.9 nm spectral line have been studied in a linear, low-pressure, optically thin pulsed arc discharge. The line shapes are measured in three different plasmas at about 16,000 K electron temperature (T) and about 7.0×10^{22} m^{-3} electron density (N). The separate electron and ion contributions to the total Stark width (W_t), i.e. W_e and W_i, as well as to the total Stark shift (d_t), i.e. d_e and d_i, have also been obtained and represent new experimental data in this field.

On the basis of the observed asymmetry of the Stark broadened line profile we have deduced the ion broadening parameters, which describe the influence of the ion static (A) and the ion-dynamical effect on the width (D) and on the shift (E) of the line shape. Stronger influence of the ion contribution on the 425.9 nm Ar I line shape than is the one predicted by current theory has been evidenced.

On the basis of the accurately recorded 425.9 nm Ar I line shape (in the 4s′ –5p′ transition), the basic plasma parameters, i.e. electron temperature (T) and electron density (N) have been recovered. This has been achieved by applying the recently developed line deconvolution procedure. The plasma parameters (T and N) have also been measured using independent diagnostics techniques.

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Keywords: Line shapes; Widths and shifts; Neutrals in plasmas; Plasma diagnostic techniques and instrumentation

1. Introduction

In thermal plasmas at local thermodynamic equilibrium (LTE), and for temperatures of about 1–2 eV (electron density about 1×10^{23} m^{-3}), the dominant mechanism for Stark broadening of lines of isolated neutral atoms is caused by collision processes with electrons and ions. Stark broadening in plasmas is important to theoretical understanding as well as for experimental methods, e.g. as a diagnostic tool. Plasma broadened and shifted spectral line profiles have been used for a number of years as a basis of an important non-interfering plasma diagnostic technique. Numerous theoretical and experimental efforts have been made to find solid and reliable basis for this application. This technique became, in some cases, the most sensitive and often the only possible plasma diagnostic tool [1].

Classical emission spectroscopy on laboratory plasmas has been mainly applied to optically thin plasma for which absorption of the emitted radiation is negligible. Investigation of line profiles of atomic argon in medium dense plasmas, at about 1×10^{23} m^{-3}, are of particular interest for verifying predictions of theoretical approximations. Argon is the preferred constituent of laboratory plasmas [3–9], and the Ar I spectral line shapes represent important sources of information about the physical conditions in the place of birth of the radiation [2]. The importance of Stark broadening is investigated in many papers, i.e. recently in Refs. [10,11]. On this way, a significant number of experimental studies [12–23] has been dedicated to the investigation of the Ar I total Stark full-width at half intensity maximum (FWHM), W_t of the 425.9 nm spectral line (4s′ –5p′ transition). A number of papers also deals

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* Corresponding author. Tel.: +38 111 630152; fax: +38 111 3282619.
E-mail addresses: vladimir@ff.bg.ac.yu, vlada67@yahoo.com (V. Milosavljević).

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with the total Stark shift (d_t) [15–19,24,25]. However, to our knowledge, only one study [26] presents theoretical W_e, d_e and A values in the 4s′–5p′ transition.

In this work we applied the line deconvolution procedure [27] to precisely recorded 425.9 nm Ar I line profiles. The basic plasma parameters, i.e. electron temperature (T^D) and electron density (N^D) have been obtained, using the line deconvolution procedure, for three different plasmas created in a linear, low-pressure, pulsed arc discharge in helium–argon and hydrogen–argon mixtures. To our knowledge, our results for the T and N have been the first published data obtained directly from the 425.9 nm Ar I line profile, using a deconvolution procedure. The same parameters have also been measured (T^{exp} and N^{exp}) using independent, well-known, experimental diagnostical techniques. Excellent agreement has been found within the two sets of the obtained parameters (T^D and T^{exp}; and N^D and N^{exp}). This recommends the applied deconvolution procedure for plasma diagnostic purposes, especially in circumstances where direct measurements of T and N are not possible. The method is suitable for optically thin plasmas. For optically thick plasmas the situation is more complicated because of very weak asymmetry of the spectral line profile caused by self-absorption.

In this paper we are presenting the measured Stark width and shift of the 425.936 nm Ar I spectral line (in 4s′–5p′ transition, multiplet) at about 16,000 K electron temperature and at about 7.0×10^{22} m^{-3} electron density. The used T values are typical for many cosmic light sources and laboratory plasmas. On the basis of the observed Ar I line profile asymmetry, the characteristics of the ion contribution to the total Stark FWHM (W_t), as expressed by the the ion-static parameter (A) and the ion-dynamical parameter (D) have also been obtained. Analogously, the total Stark shift (d_t), has been obtained as well, in function of A and the corresponding ion-dynamical parameter E. As an optically thin plasma source we have used a linear, low-pressure, pulsed arc operated in three different discharge conditions. Our W_t, W_e, W_i, d_t, d_e, d_i and A values have been compared to all available theoretical and experimental Stark broadening parameters.

We would like to point out that the existing experimental Ar I Stark width data in Refs. [28–30] and references therein refer to the sum of the electron (W_e) and ion (W_i) contributions to the total Stark width (W_t) and to the sum of the electron (d_e) and ion (d_i) contributions to the total Stark shift (d_t) without the possibility of estimating the contribution of electrons and ions separately. The W_e, W_i, d_e and d_i Ar I values presented here are, to the knowledge of the authors, the first data in the field, with the separate ion and electron contributions evaluated from the measured total Stark width and shift by using the line deconvolution procedure described in Ref. [27]. This method has been recently successfully applied [31–37], to line profiles of He I, Ne I, Ar I and Kr I.

2. Theoretical background and deconvolution procedure

The total line Stark FWHM (W_t) and shift (d_t) with the corresponding electron W_e and d_e and ion W_i and d_i contributions are respectively given by:

$$W_t = W_e + W_i \quad \text{and} \quad d_t = d_e + d_i \tag{1}$$

For a non-hydrogenic, isolated neutral atom line the ion broadening is not negligible and the line profiles are described by an asymmetric K function (see Eq. (7)). The W_t and d_t may be calculated from the equations [26,38]:

$$W_t \approx W_e[1 + 1.75AD(1 - 0.75R)]$$

and

$$d_t \approx W_e[d_e/W_e \pm 2AE(1 - 0.75R)] \tag{2}$$

where

$$R = \sqrt[6]{\frac{36\pi e^6 N}{(kT)^3}}, \tag{3}$$

is the so-called Debye shielding parameter, i.e. the ratio of the mean ion separation to the Debye radius, where k is the Boltzmann constant and N and T represent the electron density and temperature, respectively. A is the quasi-static ion broadening parameter (see Eq. (224) in Ref. [26]), D is a coefficient of the ion-dynamical contribution to the width and E is a coefficient of the ion-dynamical contribution to the shift, with the established criterion:

$$D = \frac{1.36}{1.75(1 - 0.75R)} B^{-1/3} \quad \text{for} \quad B < \left(\frac{1.36}{1.75(1 - 0.75R)}\right)^3;$$

or

$$D = 1 \quad \text{for} \quad B \geq \left(\frac{1.36}{1.75(1 - 0.75R)}\right)^3, \tag{4}$$

and

$$E = \frac{2.35B^{-1/3} - 3A^{1/3}R}{2(1 - 0.75R)} \quad \text{for} \quad B < 1;$$

or

$$E = 1 \quad \text{for} \quad B \geq 1, \tag{5}$$

where

$$B = A^{1/3}\frac{4.03 \times 10^{-7} W_e[\text{nm}]}{(\lambda[\text{nm}])^2}\left(N\left[\text{m}^{-3}\right]\right)^{2/3}\sqrt{\frac{\mu}{T_g[\text{K}]}} < 1; \tag{6}$$

is the factor with atom-ion perturber reduced mass μ (in amu) and gas temperature T_g. When D=1 and E=1 the influence of the ion-dynamic effect on both the width and the shift is negligible and the line shape is treated using the

Table 1
Different discharge conditions: C—bank capacity (in μF), U—bank voltage (in kV), H—plasma length (in cm), Φ—tube diameter (in mm), P—filling pressure (in Pa)

Working gases	Exp.	C	U	H	Φ	P	N^{exp}	N^{D}	T^{exp}	T^{D}
72% Ar +28% He	*a*	14	1.5	7.2	5	133	6.7±7%	6.5±12%	15.6±11%	15.4±12%
97% Ar +3% H_2	*b*	14	1.5	7.2	5	67	7.0±7%	6.8±12%	16.0±11%	15.8±12%
97% Ar +3% H_2	*c*	14	1.5	7.2	5	133	7.1±7%	7.0±12%	16.2±11%	16.2±12%

N^{exp} (in 10^{22} m^{-3}) and T^{exp} (in 10^3 K) denote experimental electron density and temperature, respectively, obtained at a moment when the line profiles were analyzed. N^{D} (in 10^{22} m^{-3}) and T^{D} (in 10^3 K) represent averaged electron density and averaged electron temperature obtained by using the line deconvolution procedure [27].

quasi-static ion approximation, described by Ref. [27] and references therein:

$$K(\lambda) = K_{\mathrm{o}} + K_{\max} \int_{-\infty}^{\infty} \exp(-t^2) \times \left[\int_0^{\infty} \frac{H_{\mathrm{R}}(\beta)}{1 + \left(2\frac{\lambda - \lambda_{\mathrm{o}} - \frac{W_{\mathrm{G}}}{2\sqrt{\ln 2}} t}{W_{\mathrm{e}}} - \alpha\beta^2 \right)^2} d\beta \right] d\beta \qquad (7)$$

Here K_{o} is the baseline (offset) and $K_{\max}$ is the maximum intensity (for $\lambda=\lambda_{\mathrm{o}}$) [27]. $H_{\mathrm{R}}(\beta)$ is the electric microfield strength distribution function of normalized field strength $\beta=F/F_{\mathrm{o}}$, where F_{o} is the Holtsmark field strength. A ($\alpha=A^{4/3}$) is the quasi-static ion broadening parameter and represents the measure of the relative importance of ion and electron broadenings. R is the Debye shielding parameter (see Eq. (3)) and W_{e} is the electron width (FWHM) in the $j_{\mathrm{A,R}}$ plasma broadened spectral line profile [26]. The Gaussian FWHM width W_{G} is given by Eq. (8) (i.e. the Eq. (2.3) in Ref. [27]).

$$W_{\mathrm{G}} = 2\sqrt{\frac{2\ln 2 kT}{m}} \frac{\lambda_{\mathrm{o}}}{c}. \qquad (8)$$

Here T is the emitter equivalent kinetic temperature, m is its mass, and k and c are the Boltzmann constant and velocity of the light, respectively.

For the purpose of the deconvolution iteration process we need to know the value of K (Eq. (7)) as a function of λ for every group of parameters ($K_{\max}$, λ_{o}, W_{e}, W_{G}, R, A). The used numerical procedure for solution of Eq. (7) is described in earlier publications [27,37,39].

From Eqs. (1–8) it is possible to obtain the plasma parameters (N and T) and the line broadening characteristics (W_{t}, W_{e}, W_{i}, d_{t}, d_{e}, d_{i}, A, D and E). One can see that the ion contribution, expressed in terms of the A, D and E parameters directly determines the ion width (W_{i}) and shift (d_{i}) component in the total Stark width and shift (Eqs. (1) and (2)).

The deconvolution method allows direct determination of all six parameters by fitting the theoretical K-profile (Eq. (7)) to experimental data but requires a sufficient number of experimental points per line, and small statistical errors. The upper limits of numerical conditionality of this method are a minimum of 20 experimental points per line (within the range $-3/2W_{\mathrm{e}}+\lambda_{\mathrm{o}}<\lambda<+3/2W_{\mathrm{e}}+\lambda_{\mathrm{o}}$, and maximal statistical indeterminacy in intensity of 5% at every experimental point. Poor experimental measurements weaken the conditionality of the system of equations, and lead to non-applicability of this method. This has been concluded by testing the sensitivity of the algorithm by generating random statistical noise with Gaussian distribution in every point convolved by theoretical profiles. The fitting procedure with the K-convolution integral has also been tested using another set of experimental data (see Refs. [31–37]). The K convolution integral is used for the analysis of our new data for many spectral lines of neutral rare gases. By comparing the different spectral lines obtained under the same plasma conditions, we tested the physical stability of the deconvolution procedure. The obtained parameters, which are tied to plasma conditions, such as T and N, are independent from the analyzed lines. The values of temperature calculated from a single spectral line and the ones obtained by Boltzmann and Saha equations are in very good agreement, within ±7%. Even better agreement, within ±5%, is achieved between the electron density values as obtained from a single spectral line analysis and as measured by interferometry (Table 1).

Taking into account the uncertainties of the line profile measurements and the above mentioned, we estimate errors ±12% for the W_{e}, d_e, W_{i} and d_{i}, ±15% for the A parameter and ±20% for D and E.

3. Experiment

The modified version of the linear low pressure pulsed arc [40–44] has been used as a plasma source. Pulsed discharge was performed in a quartz discharge tube. The working gases were helium–argon (28% He+72% Ar) and hydrogen–argon (3% H_2+97% Ar) mixtures. The used tube geometry and corresponding discharge conditions are presented in Table 1.

The spectroscopic observation of spectral lines has been made end-on along the axis of the discharge tube.

The line profiles were recorded by a step-by-step technique using a photomultiplier (EMI 9789 QB and EMI 9659B) and a grating spectrograph (Zeiss PGS-2, reciprocal linear dispersion 0.73 nm/mm in the first order) system. The instrumental FWHM of 8 pm was obtained by using narrow spectral lines emitted by the hollow cathode discharge. The spectrograph exit slit (10 μm) with the calibrated photomultipliers was micrometrically traversed along the spectral plane in small wavelength steps (7.3 pm). The averaged photomultiplier signal (five shots in each position) was digitized using an oscilloscope, interfaced to a computer.

Plasma reproducibility was monitored by the Ar I and Ar II line radiation and, also, by the discharge current using a Rogowski coil signal (it was found to be within ±5%).

The applied deconvolution procedure is extensively described in Refs. [27,39] It includes a new advanced numerical procedure for deconvolution of theoretical asymmetric convolution integral of a Gaussian and a plasma broadened spectral line profile $j_{A,R}(\lambda)$ for spectral lines. This method gives complete information on the plasma parameters from a single recorded spectral line. The method determines all broadening (W_t, W_e, W_i, d_t, d_e, d_i, A, D and E) and plasma parameters (N and T) self-consistently and directly from the shape of spectral lines without any assumptions or prior knowledge, making it useful in astrophysics. All one needs to know is the instrumental width of the spectrometer when the spectrometer instrumental width is insignificant compared with the width of any component into which the line profile is separated. Otherwise, a deconvolution for the true instrument function should be calculated as well. The measured profiles are the results of convolution with the Lorentzian Stark and Gaussian profiles caused by Doppler and instrumental broadening [26]. Van der Waals and resonance broadenings [26] were estimated to be smaller by more than an order of magnitude in

Table 2
The 425.936 nm Ar I line broadening characteristics

T^{exp}	N^{exp}	W_t^{exp}	W_e^{exp}	W_i^{exp}	A^{exp}	D^{exp}	d_t^{exp}	d_e^{exp}	d_i^{exp}	E^{exp}	Ref.
15.6	6.7	158.4	130.1	28.3	0.190	1.25	113.8	90.3	23.5	1.00	Tw
16.0	7.0	167.0	136.9	30.1	0.191	1.54	109.4	85.3	24.1	1.55	Tw
16.2	7.1	163.8	134.0	29.8	0.193	1.55	113.4	86.2	27.2	1.49	Tw
11.9	6.21	144			0.114						[12]
11.9	6.23	133			0.122						[13]
11.1	0.4	2.85									[14]
9.75–12.7	1.2–9.4	23–197					14–105				[15]
11.4	4.6	120					56				[16]
14.0	1.0	18.7					5				[17]
11.6–10.5	10	244									[18]
11.6–10.5	4.0–9.0						100*				[18]
11.07; 11.04	3.50; 3.40	71; 70					38; 37				[19]
10.98; 10.89	3.30; 3.10	66; 62					36; 34				[19]
10.69; 10.54	2.70; 2.40	57; 49					30; 25				[19]
10.31; 10.12	2.00; 1.70	42; 35					23; 19				[19]
9.93; 9.68	1.40; 1.05	30; 24					16; 12				[19]
9.47; 9.26	0.90; 0.70	19; 14					9; 8				[19]
9.13; 8.90	0.60; 0.47	12; 10					7; 6				[19]
10.55–12.5	2.6–9.2	76.4–198.5									[20]
9.28	0.74				0.121						[21]
9.40	0.83				0.126						[21]
9.52	0.98				0.130						[21]
9.72	1.20				0.137						[21]
9.90	1.40				0.141						[21]
10.05	1.60				0.147						[21]
10.25	1.90				0.151						[21]
10.4	2.15				0.155						[21]
10.55	2.46				0.159						[21]
10.7	2.70				0.161						[21]
10.73	2.82				0.162						[21]
10.76	2.90				0.163						[21]
13.0; 16.2	3.8; 2.6	97; 68									[22]
15.0	1.4	31									[22]
9.8	1.4	31									[23]
10.8	3.1	70									[23]
11.5	5.1	114									[23]
12.6	9.9	200									[23]
14.0	2.6						23				[24]
13.5	12.8						167				[25]

Measured: W_t^{exp}, W_e^{exp}, W_i^{exp}, d_t^{exp}, d_e^{exp} and d_i^{exp} in pm within 12% accuracy at measured electron temperature (T^{exp} in 10^3 K) and electron density (N^{exp} in 10^{22} $\mathrm{m}^{\text{ã}\,3}$). Under Ref. are sources of experimental data: with are denoted (Tw) present data.

* Data taken at electron density 1×10^{23} m^{-3}.

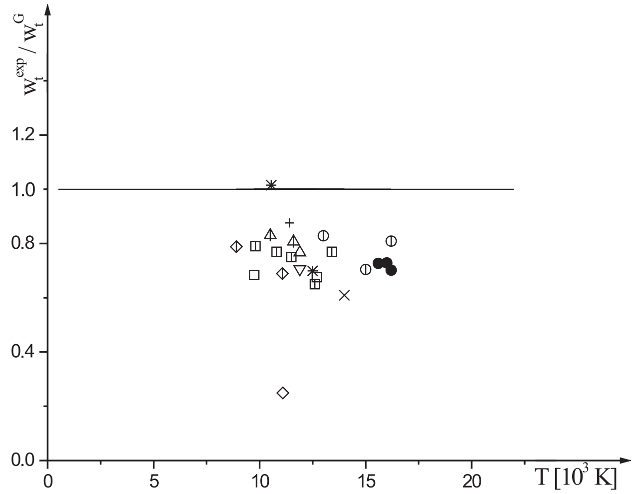

Fig. 1. Ratios of the experimental total Stark FWHM (W_{t}^{exp}) to the theoretical (W_{t}^{G}) predictions [26] vs. electron temperature for λ=425.936 nm. ●, our experimental results and those of other authors: △, Ref. [12]; ▽, Ref. [13]; ◇, Ref. [14]; □, Ref. [15]; +, Ref. [16]; ×, Ref. [17]; ⟁, Ref. [18]; ◈, Ref. [19]; *, Ref. [20]; ⌽, Ref. [22] and ◫, Ref. [23].

comparison to Stark, Doppler and instrumental broadenings. The deconvolution procedure was computed using the least Chi-square function [27].

To check existence of the self-absorption, relative line intensity (I) ratio has been controlled during the plasma decay. Absence of the self-absorption would cause constant relative line intensity ratio [45]. This is fulfilled in the case of the $I_{425.9}/I_{419.1}$ ratio within the 4s′ –5p′ Ar I transition. Therefore, influence of the self-absorption on the 425.9 nm Ar I line intensity can be neglected.

The plasma parameters (N and T) were determined independently using standard diagnostics methods. Thus, the electron temperature was determined from the ratios of the relative line intensities of seven Ar I spectral lines (415.859, 416.418, 419.103, 419.832, 420.067, 425.936 and 426.627 nm) to the five Ar II spectral lines (335.093, 420.197, 426.653, 487.986, 488.903 nm) with an estimated error of ±11%, assuming the existence of LTE [26]. The necessary atomic data have been taken from Ref. [29]. The electron density decay was measured using a well-known single wavelength He–Ne laser interferometer technique for the 632.8 nm transition with an estimated error of ±7%.

4. Results and discussion

The measured N^{exp} and T^{exp} are presented in Table 1 together with the N^{D} and T^{D} values obtained using the line profile deconvolution procedure for the single Ar I line. One can conclude that the agreement among T^{exp} and T^{D} values is excellent (within 3% on average in the three plasmas investigated). This fact confirms the homogeneity of the investigated plasmas in the linear part of our light source (see Fig. 1 in Ref. [41]). For the electron density the situation is similar. The agreement among the two sets of values for the electron density (N^{exp} and N^{D}) is within the experimental accuracy of 7% and within the uncertainties (12%) of the results obtained by deconvolution procedure. The plasma broadening parameters (W_{t}^{exp}, W_{e}^{exp}, W_{i}^{exp}, A^{exp}, D^{exp}) obtained using our deconvolution procedure of the recorded line profiles at measured N^{exp} and T^{exp} values are presented in Table 2 together with those of other authors.

In order to facilitate the comparison among the measured total (electron+ion) FWHM W_{t}^{exp} values and the well-known theoretical one W_{t}^{G}, due to Griem [26] (Eq. (226) from Ref. [26]), the dependence of the ratio $W_{t}^{\mathrm{exp}}/W_{t}^{G}$ on the electron temperature is presented graphically in Fig. 1.

Analogous to Fig. 1 is Fig. 2, where the ratio of the measured to theoretical total (electron+ion) shift values $d_{t}^{\mathrm{exp}}/d_{t}^{G}$ is presented in function of temperature.

Our broadening parameter (W_{t}^{exp}) and Stark shift (d_{t}^{exp}) represent the second set of measured values at electron temperatures higher than 14,000 K. The comparison of our W_{t}^{exp} (d_{t}^{exp}), W_{e}^{exp} (d_{e}^{exp}) and A^{exp} values with unique theoretical data [26] was performed. For the 425.936 nm Ar I spectral line, the theoretical W_{t}^{G} values due to Griem′ s [26] are about 1.3 as high as the experimental W_{t}^{exp} values (see Fig. 1). Similarly, the theoretical d_{t}^{G} values are about 1.4 as high as the experimental d_{t}^{exp} ones (see Fig. 2).

When the comparison of the electron contribution, in particular, is concerned, the W_{e}^{G} values are about 1.5 as high as our W_{e}^{exp} values, while the d_{e}^{G} values are found to be about 1.44 as high as our d_{e}^{exp} values. We are inclined to believe that the discrepancy between measured and theoretical values indicates that the atomic data used for calculations [26] were not reliable enough. Our ion broadening parameters (A^{exp}) over value the theoretical (A^{G}) values by about 80% and are multiplied 1.2–1.5 times with the ion-dynamical coefficient (D^{exp}), depending on the

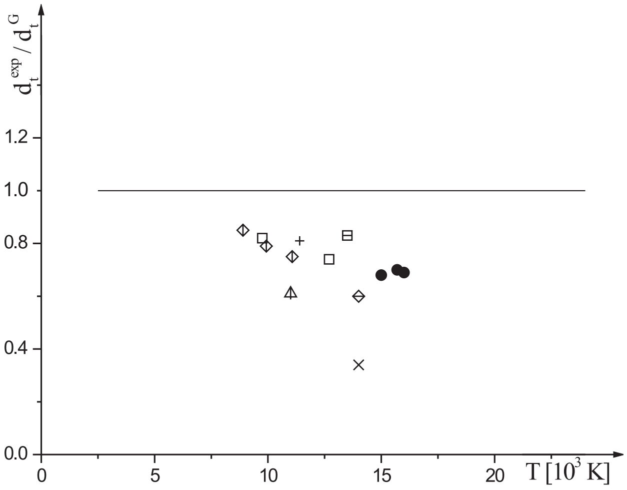

Fig. 2. Ratios of the experimental total shift (d_{t}^{exp}) to the theoretical (d_{t}^{G}) predictions [26] vs. electron temperature for λ=425.936 nm. ●, our experimental results and those of other authors: □, Ref. [15]; +, Ref. [16]; ×, Ref. [17]; ⟁, Ref. [18]; ◈, Ref. [19]; ⬘, Ref. [24] and ⊟, Ref. [25].

discharge conditions. Generally, the electron contribution to the total Stark width found experimentally is about 85% (on average), while the electron contribution to the total Stark shift found experimentally is about 75% at about 16,000 K electron temperature. It turns out that A^{exp} values obtained by Refs. [12,13,21] for the 425.936 nm line agree well with ours (see Table 2). Direct comparison of our $W_{\mathrm{t}}^{\mathrm{exp}}$ ($d_{\mathrm{t}}^{\mathrm{exp}}$) values with other experimental data is impossible, because of the different plasma conditions in the experiments. But, taking into account the approximative normalization factor W_{t}/N (d_{t}/N) we have found a tolerable scatter for the mentioned transitions among W_{t}/N (d_{t}/N) experimental values within 15–38% (20%). The only exceptions are the very low W_{t} (and d_{t}) values from Refs. [14] and [17], respectively. Namely, the Ref. [14] W_{t} values, when compared with ours, as well as with others authors are about four times smaller. Similarly, the d_{t} values due to Ref. [17] are about 2.5 times smaller than ours and the values of other authors.

5. Conclusion

It has been shown that the line deconvolution procedure described by Ref. [27], applied to Ar I line profiles, gives convenient plasma parameters (N and T) at about 16,000 K electron temperature and 7×10^{22} m^{-3} electron density. We recommend this method for plasma diagnostic purposes for the case of optically thin plasmas. We have found clear influence of the quasi-static ion and ion-dynamical effects on the investigated spectral line shapes. They play a much more important role than the semi classical theory predicts. The observed ion-dynamical effect augments the ion contribution to the line Stark width and shift by up to a factor of 1.5, at the plasma conditions studied presently. This reveals the evident contribution, amounting to 20%, of the argon ions to the total line width and shift. This findings are of importance in the use of Ar I line either for astrophysical and laboratory plasma modeling or for diagnostics.

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