Stark Width and Shift of the Neutral Argon 425.9 nm Spectral Line

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

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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 \times 10^{22} \text{ 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 \text{ V} – 5p \text{ V} 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 \(1 \times 10^{23} \text{ m}^{-3}\)), 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 \(1 \times 10^{23} \text{ m}^{-3}\), 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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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
\( (T^{\text{exp}} \) and \( N^{\text{exp}}) \) using independent, well-known,
experimental diagnostic techniques. Excellent agreement has
been found within the two sets of the obtained parameters
\( (T^D \) and \( T^{\text{exp}} \), and \( N^D \) and \( N^{\text{exp}}) \). This recommends the
applied deconvolution procedure for plasma diagnostic
purposes, especially in circumstances where direct meas-
urements 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 \( 7.0 \times 10^{22} \) 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 \( W_t, W_e, W_i, d_e, d_i \) and \( A \) values have been compared
to all available theoretical and experimental Stark broad-
ening parameters.

We would like to point out that the existing experi-
mental 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 estimat-
ing the contribution of electrons and ions sepa-
rately. 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

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

where

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

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

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

is the factor with atom-ion perturber reduced mass \( \mu \) (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
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)

<table>
<thead>
<tr>
<th>Working gases</th>
<th>Exp.</th>
<th>C</th>
<th>U</th>
<th>H</th>
<th>Φ</th>
<th>P</th>
<th>N_{exp}</th>
<th>N^{O}</th>
<th>T_{exp}</th>
<th>T^{O}</th>
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<td>a</td>
<td>14</td>
<td>1.5</td>
<td>7.2</td>
<td>5</td>
<td>133</td>
<td>6.7±7%</td>
<td>6.5±12%</td>
<td>15.6±11%</td>
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<tr>
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<td>67</td>
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<td>16.0±11%</td>
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<tr>
<td>72% Ar +3% H₂</td>
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<td>14</td>
<td>1.5</td>
<td>7.2</td>
<td>5</td>
<td>133</td>
<td>7.1±7%</td>
<td>7.0±12%</td>
<td>16.2±11%</td>
<td>16.2±12%</td>
</tr>
</tbody>
</table>

\(N_{\text{exp}}\) (in 10^22 m\(^{-3}\)) and \(T_{\text{exp}}\) (in 10^4 K) denote experimental electron density and temperature, respectively, obtained at a moment when the line profiles were analyzed. \(N^O\) (in 10^22 m\(^{-3}\)) and \(T^O\) (in 10^4 K) represent averaged electron density and averaged electron temperature obtained by using the line deconvolution procedure [27].

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 \(-3/2W_e+\lambda_o<\lambda<+3/2W_e+\lambda_o\)), 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% H₂+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 \( j_{\lambda, R}(\lambda) \) 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.

Table 2
The 425.936 nm Ar I line broadening characteristics

<table>
<thead>
<tr>
<th>( T_{\text{exp}} )</th>
<th>( N_{\text{exp}} )</th>
<th>( W_{t, \text{exp}} )</th>
<th>( W_{e, \text{exp}} )</th>
<th>( W_{i, \text{exp}} )</th>
<th>( A_{\text{exp}} )</th>
<th>( D_{\text{exp}} )</th>
<th>( d_{t, \text{exp}} )</th>
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Measured: \( W_{t, \text{exp}}, W_{e, \text{exp}}, W_{i, \text{exp}}, d_{t, \text{exp}}, d_{e, \text{exp}} \) and \( d_{i, \text{exp}} \) in pm within 12% accuracy at measured electron temperature (\( T_{\text{exp}} \) in 10^3 K) and electron density (\( N_{\text{exp}} \) in 10^2 m^-3). Under Ref. are sources of experimental data: with are denoted (Tw) present data.

* Data taken at electron density 1×10^23 m^-3.
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, 415.859, 416.418, 419.103, 419.832, 420.067, 425.936 nm) and 426.627 nm) to the five Ar II spectral lines (335.093, 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, 415.859, 416.418, 419.103, 419.832, 420.067, 425.936 nm) with an estimated error of \( \pm 11\% \), 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 \( \pm 7\% \).

4. Results and discussion

The measured \( N_{\text{exp}} \) and \( T_{\text{exp}} \) 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 \( T_{\text{exp}} \) 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 (\( N_{\text{exp}} \) and \( N^G \)) is within the experimental accuracy of 7\% and within the uncertainties (12\%) of the results obtained by deconvolution procedure.

The plasma broadening parameters (\( W_{\text{exp}}^i \), \( W_{\text{exp}}^e \), \( W_{\text{exp}}^A \), \( A_{\text{exp}} \), \( D_{\text{exp}} \)) obtained using our deconvolution procedure of the recorded line profiles at measured \( N_{\text{exp}} \) and \( T_{\text{exp}} \) 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 \( W_{\text{exp}}^i \) values and the well-known theoretical one \( W_{\text{G}}^i \), due to Griem [26] (Eq. (226) from Ref. [26]), the dependence of the ratio \( W_{\text{exp}}^i/W_{\text{G}}^i \) 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 \( d_{\text{exp}}/d_{\text{G}} \) is presented in function of temperature.

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

When the comparison of the electron contribution, in particular, is concerned, the \( W_{\text{G}}^i \) values are about 1.5 as high as our \( W_{\text{exp}}^e \) values, while the \( d_{\text{G}} \) values are found to be about 1.44 as high as our \( d_{\text{exp}} \) 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 (\( A_{\text{exp}} \)) over value the theoretical (\( A_{\text{G}} \)) values by about 80\% and are multiplied 1.2–1.5 times with the ion-dynamical coefficient (\( D_{\text{exp}} \)), depending on the
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 $W_{\exp}$ values obtained by Refs. [12,13,21] for the 425.936 nm line agree well with ours (see Table 2). Direct comparison of our $W_{\exp}$ ($d_{\exp}$) values with other experimental data is impossible, because of the different plasma conditions in the experiments. But, taking into account the approximative normalization factor $W_{i}/N$ ($d_{i}/N$) we have found a tolerable scatter for the mentioned transitions among $W_{i}/N$ ($d_{i}/N$) experimental values within 15–38% (20%). The only exceptions are the very low $W_{i}$ and $d_{i}$ values from Refs. [14] and [17], respectively. Namely, the Ref. [14] $W_{i}$ values, when compared with ours, as well as with others authors are about four times smaller. Similarly, the $d_{i}$ values 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 $7 \times 10^{22} \text{ m}^{-3}$ 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.

Acknowledgments

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References


