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An interferometric and spectroscopic argon arc plasma diagnostic

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Abstract. In the spectral diagnostics of an arc plasma, when the halfwidth of the Balmer H\textsubscript{\alpha} line is measured, there is a certain amount of free choice in the selection of equations giving \(N_e = F(\Delta \lambda_{1/2}(H\beta))\) and \(\Delta E_i = G(N_e, T)\), the lowering of the ionisation energy. In order to clear up this problem, the argon arc plasma refraction, calculated on the grounds of end-on spectroscopic diagnostics, is compared with results of interferometric measurements. The influence of atoms in excited states on plasma refraction is taken into account. Results show that, for plasma temperature \(T > 11\,000\) K, interferometrically measured refraction is smaller than calculated values. The experiment was performed at four laser wavelengths: 457.5, 580.3, 581.9 and 632.8 nm. The arc current was varied over a range from 10 to 100 A.

1. Introduction

For a wall-stabilised argon arc plasma diagnostic, two methods are most often used. First, the spectroscopic method is based on measurement of the Stark-broadened hydrogen H\textsubscript{\beta} spectral line halfwidth. In this case a small amount of H\textsubscript{\alpha} is added to the argon plasma. The second method is based on measurement of plasma refraction.

When the halfwidth of H\textsubscript{\beta} is measured, results of calculated plasma parameters and composition (e.g. plasma temperature \((T)\), free electron density \((N_e)\), densities of atoms \((N_a)\) and ions \((N_i)\)) depend a little on the set of equations used to describe the physical state of the plasma. The most important is the dependence giving \(N_e\) as a function of the measured halfwidth \(\Delta \lambda_{1/2}(H\beta)\)

\[
N_e = F(\Delta \lambda_{1/2}(H\beta)). \quad (1)
\]

In most cases this dependence is used as an analytical expression fitted to values obtained from tables published by Vidal et al (1973) (vcs). From these tables \(N_e\) as a function of the reduced halfwidth \(\Delta \alpha_{1/2}(H\beta)\) may be interpolated. The definition of the reduced halfwidth in terms of the measured halfwidth in the wavelength scale \(\Delta \lambda_{1/2}(H\beta)\) is

\[
\Delta \alpha_{1/2}(H\beta) = \Delta \lambda_{1/2}(H\beta)/F_0
\]

where \(F_0 = 1.254 \times 10^{-9} N_e^{-2/3}\) is Holtsmark’s normal field strength. Depending on author and applied method of calculation, equation (1) gives slightly different results. Differences may reach 4\%. One may also use experimental points (Wiese et al 1972, Baessler and Kock 1980, Helbig and Nick 1981) for fitting equation (1). In this case
results may differ up to 10% depending on the free-electron density range (figure 6 in Helbig and Nick 1981).

There is no final answer to the question of which expression, for lowering of the ionisation energy $\Delta E_1$, better describes physical reality in the arc plasma. Calculated values of $\Delta E_1$, predicted by different formulae, differ a lot; however, the influence of these values on results of plasma diagnostics is rather small.

As may be seen, in the case of spectral diagnostics of the plasma, there exists a certain amount of free choice in selection of equation (1) and formula for $\Delta E_1$. As a consequence, the plasma parameters determined depend on the selection of applied equations.

Spectroscopic methods are not precise enough to select unequivocally equations for plasma diagnostics. A better chance to solve this problem may be via application of laser interferometry to arc plasma diagnostics. The reason for this is that the specific refractivity for a free electron ($K_e$) is much larger than for an atom ($K_a$) or ion ($K_i$) in the ground state, e.g. for argon $K_e/K_a=17$ for $\lambda=632$ nm. For this reason, interferometry may be sensitive enough to monitor changes of the free-electron density when the plasma temperature varies.

An open question, in an interferometric investigation of the plasma, is the influence of atoms in excited states on the total plasma refraction. According to rough estimations (e.g. Griem 1964, Baessler and Kock 1980), in an arc plasma, the refraction of excited atoms equals about 1% of this value for free electrons. Calculations of the specific refractivity of excited atoms, $K^*$, does not give consistent results (Hug et al 1967a, b, Glasser et al 1981, Helbig and Nick 1985). Experimental results (Hug et al 1967a, b, Baum et al 1975, Musiol et al 1981) indicate that the contribution of atoms in excited states to the total plasma refraction is small, but does not give definitive conclusions.

The aim of our investigation was to find out which combination of expressions

\begin{align}
N_e &= F(\Delta \lambda_{1/2} (H_B)) \\
\Delta E_1 &= G(T, N_e) \\
K^* &= H(T)
\end{align}

(2) gives values of the plasma refraction, calculated on the grounds of spectral diagnostic, in better agreement with refraction of the argon plasma measured by means of the Mach-Zehnder interferometer, for selected laser wavelengths. For each expression (2) two limits were selected.

For (2a), as upper limit (larger values), the straight-line fit to experimental values measured by Helbig and Nick (1981) was used (HNexp in tables 1 and 2). As the lower limit an analytical function fitted to values of vcs by Jones et al (1983) and Musiol et al (1982) was applied (DWJ in tables 1 and 2). The latter case gives smaller values of $N_e$, which are in better agreement with experimental data of Wiese et al (1972) than with results of Helbig and Nick (1981). The difference between these two limits is $\leq 6\%$. Other fits of vcs data, e.g. Czernichowski and Chapelle (1983) and Helbig and Nick (1981), give intermediate values.

For (2b), as upper and lower limits, formulae proposed by Unsold (1948) and Griem (1964) were used respectively (U and G in tables 1 and 2). Other formulae, e.g. Rother (1958), give intermediate values.

For (2c), two limits were taken as $K^*=K_a$ and $K^*=K_e+K_i$. (A and E+I in tables 1 and 2). If all kinds of possible resonant interactions between excited states and excited states and continuum are negligible, these limits give maximal and minimal
values of the specific refraction for an excited atom. Our calculations of $K^*$, described in § 5, consistent with results of Helbig and Nick (1985), give values of $K^*$ within these limits.

This gives altogether four combinations for calculation of the plasma temperature, electron density and plasma composition, and eight combinations for calculations of the plasma refraction from spectroscopic measurements. An example of the influence of above limits on the final results of the plasma diagnostic is shown in table 1. For $\lambda_L = 632.8$ nm, the mean values, of four $T_e$, four $N_e$ and eight $(n_p - 1)$ calculated values, are shown in the last row. In columns, values (in per cent) of $\Delta X = (X_m - X)/X_m$ are shown; $X$ denotes $T_e$, $N_e$ or $(n_p - 1)$ and $X_m$ is the mean value.

2. Description of experiment

The experimental set-up is shown in figure 1. In two arms of a Mach–Zehnder interferometer two cascade arcs were placed. Interference of the laser beams, with maximum 0.4 mm diameter on the axis of arc channels, was observed. The laser beams were focused onto the centres of the arcs. Cascades consisted of water-cooled copper or brass plates, which were 5 mm thick and isolated by 1 mm spacers. The arc channel was 4 mm in diameter. Arcs were operated at atmospheric pressure. So-called three-chamber arcs (e.g. Musiol et al 1982) differ only in the length of the arc column and

![Figure 1. Schematic diagram of the experimental set-up.](image-url)
were connected electrically in series, so the current was the same in two arcs. Also the gas flow and electrode regions were identical.

When the arc current varies, on the output of the interferometer one may observe the modulation of the laser light intensity caused by changes of the refraction in the uniform plasma column of the length \( \Delta L = L_L - L_S \); \( L_L \) and \( L_S \) denote the length of the long and short arc column respectively. Interferometry permits measurement of only the difference in optical paths in two arms, so these parts of the long and short arcs, which are identical, have no influence on the interferometric picture.

Independent experiments were performed for four wavelengths \( \lambda_L \): \( \{3\} \) for 457.5 nm, \( \{5\} \) for 580.3 nm, \( \{2\} \) for 581.9 nm and \( \{3\} \) for 632.8 nm; \( \{x\} \) denotes the number of independent experiments. He-Ne and pulsed dye lasers were used. \( \Delta L \) was varied in experiments from 6.46 to 14.6 cm.

Always, the part of the plasma column observed in the spectroscopic measurements was the same as the one investigated interferometrically. For each independent experiment the whole optical system was aligned and the arc position remained unaltered for all spectroscopic and interferometric measurements. Spectral line profiles were registered numerically, at 256 points, on the computer with the sampling oscilloscope card. The optical system was aligned by directing a laser beam at the exit slit of the monochromator and retracing the entire optical path. This laser beam was coaxial with the second laser beam used in interferometric measurements. For the calibration of the spectral sensitivity of the optical system a tungsten strip lamp was utilised.

3. Interferometric measurements

The procedure for each experiment was the following:

(i) For given \( \Delta L \) and \( \lambda_L \), the arc current was continuously varied in time \( t \), from \( I_{\text{max}} \) to \( I_{\text{min}} \). Modulated laser light intensity \( J(\lambda_L, t) \) and arc current \( I(t) \) were registered numerically at 256 points on two channels of the computer having a sampling oscilloscope card with an adequate timebase, mostly 20 s. Such an interferogram was registered 15 to 20 times.

(ii) In the channel registered the arc current \( I(t) \), points of integer values of current, expressed in amperes, were interpolated. In the channel registered \( J(\lambda_L, t) \), positions of maxima and minima of fringes were found. This permits us to calculate the relative fringe shift \( (dk) \) caused by variation of the arc current \( \Delta I \). Starting from the minimal integer value of the current, for every interferogram (say \( j \thinspace \text{th} \)), the relative shift of a fringe \( dk_j(s) \) per \( \Delta I = 1 \text{ A} \) and per cm of \( \Delta L \) was calculated \((s = I_{\text{min}} \text{ to } I_{\text{max}})\).

(iii) The relative fringe shift \( \Delta k_j(I) \), with respect to \( I = 35 \text{ A} \), as the function of the arc current was evaluated. Then, \( \Delta k(I) \), the mean value of \( \Delta k_j(I) \), and its standard deviation \( \sigma(I) \) were calculated; \( J = 1 \) to \( N \), \( N \) denotes the number of registered interferograms.

(iv) For given \( \lambda_L \), \( \Delta K_m(I) \), the weighted mean value of \( \Delta k(I) \) from independent experiments with different \( I_{\text{min}}, I_{\text{max}} \) and \( \Delta L \), was calculated. Each \( \Delta k(I) \) value was weighted by the inverse of its own variance \([1/\sigma(I)]^2\). Figure 2 shows values of \( \Delta k(I) \) and its standard deviation as a function of the arc current for \( \lambda_L = 632.8 \text{ nm} \) and 580.3 nm. For 580.3 nm the results of five independent experiments with different \( \Delta L \), \( I_{\text{min}} \) and \( I_{\text{max}} \), are superimposed. For 632.8 nm, results of three experiments are shown. As may be seen, measured values of \( \Delta k(I) \) overlap well within error bands and are independent of \( \Delta L \) and the range of the arc current variation.
Figure 2. The relative fringe shift as a function of the arc current, per cm of the plasma column. $\Delta k$ is measured with respect to the arc current $I = 35$ A. In (a) results of three independent experiments are superposed for $\lambda_L = 632.8$ nm. In (b) results of five experiments are shown for $\lambda_L = 580.3$ nm.

4. Spectroscopic diagnostic

The end-on spectroscopic diagnostic was made in the following way.

(i) In accordance with results published by Nick et al (1984), it was assumed that the plasma was in PLTE state for $N_e < 6 \times 10^{16}$ cm$^{-3}$ and in LTE for $N_e > 6 \times 10^{16}$ cm$^{-3}$. The underpopulation factor $b$ and the ratio $T_e/T_B$ were taken from Nick et al (1984). These measurements were performed for a very similar arc plasma.

(ii) For selected arc currents, the halfwidths of the 430 nm Ar I line as the function of $N_e$ were measured. For this purpose 1-2 vol.% of hydrogen was introduced into the middle part of the arc, then $H_B$ and 430 nm Ar I line profiles were registered. The continuous radiation under $H_B$ profile was subtracted as described by Helbig and Nick (1981). Free-electron density was calculated from equation (1). An iterative procedure was applied for calculation of the plasma temperature and composition. The dependence

$$N_e = g(\Delta \lambda_{1/2}(430))$$  \hspace{1cm} (3)

was fitted to experimental results. Two such independent calibrations, for eight and 12 selected values of the arc current, were performed.

(iii) For pure argon plasma, the profile of the 430 nm Ar I line was measured and, from equation (3), $N_e$ was calculated. Then, the plasma temperature and concentrations of plasma components were evaluated.

(iv) According to remarks in § 1, calculations (ii) and (iii) were performed for four combinations of equations (2a) and (2b). Each time, the partition function for argon was calculated as a function of $\Delta E_1$ and the plasma temperature. Remarks published by Halenka and Grabowski (1977) were taken into account.

(v) For each of four combinations of the plasma diagnostics, the total plasma refraction was calculated for two limits of $K^*, K^* = K_a$ and $K^* = K_e + K_i$.

It has been proved already in several experiments (e.g. Baessler and Kock 1980, Helbig and Nick 1981) that, for a small concentration of hydrogen (<2 vol.%) in argon plasma, $H_B$ may be treated as optically thin. Also for the 430 nm Ar I line reabsorption may be neglected (Nick 1982, Nick et al 1984, Tonjec et al 1972). The Ar I line is emitted also from the electrode regions. It affects (a little) the total line intensity measured end-on. But on the registered halfwidth of the 430 nm Ar I line it has practically no influence, as we know from our own experience (Czernichowski
and Chapelle 1983) and from other publications (e.g. Tonjec et al 1972). In our case, the reabsorption (if any) and the influence of the electrode regions (if any) are similar during measurements for Ar+ H and pure Ar plasmas and have negligible influence on the line profile and results of the plasma diagnostics.

The free-electron density $N_e$, measured in this experiment, was in the range $0.35 \leq N_e \leq 14.6 \times 10^{16} \text{ cm}^{-3}$.

5. Results

It is commonly accepted how to calculate $K_a$ and $K_e$ (e.g. Alpher and White 1965, Baum et al 1975, Griem 1964). The estimation for $K_i = 0.6K_a$ may be applied, since ions have a very small influence on the total plasma refraction.

5.1. Refraction of atoms in excited states

The refraction of atoms in excited states may be expressed as

$$(n - 1)^* = 2\pi \sum_{Hj} N_{Hj} \sum_{j \neq H} f_{Hj}/(\omega_{Hj}^2 - \omega^2). \quad (4)$$

The first sum is taken over all excited states. The second sum is taken over all possible transitions from and to state $H$, continuous states included. Here $f_{Hj}$ denotes the oscillator strengths for the transition between levels $H$ and $J$, and frequencies of the atomic transition and applied laser light. For about 280 transitions, for which energy levels may be found in Moore's tables (Moore 1949-58), calculations of $f$-values were made by use of the method elaborated by Oumarou (1986) and Oumarou et al (1988). All possible optical transitions for these levels were taken into account. The contribution of other possible transitions was calculated with the method proposed by Glasser et al (1981). Calculations were made for four wavelengths applied in this experiment and in the plasma temperature range from 8000 to 14 000 K. Results of our calculations show that, in our experimental conditions, the contribution of atoms in excited states to the total plasma refraction is smaller than 1%. Thus, the specific refractivity of an excited argon atom is contained in the range of $K^*$ used in our experiment.

5.2. Interferometric and spectroscopic results

According to remarks above, for the temperature $T_a < 11 000 \text{ K}$, the plasma refraction may be calculated correctly because the main role is played by atoms in the ground state and other plasma components give only a small contribution to the total plasma refraction $(n_p - 1)$. Therefore, in the figure showing $(n_p - 1) = f(T_a)$, for low temperatures, one may superimpose experimentally measured relative values of the plasma refraction on theoretically calculated points. Starting from these points, it is interesting to compare calculated and measured plasma refractions in the region of a higher plasma temperature, where free electrons play the dominant role and the number of atoms in excited states grows significantly.

In our experiment, in order to compare calculated and measured plasma refractions, the following procedure was applied.
From measured fringe shift \( \Delta K_m(I) \), relative changes of plasma refraction \( (n_{P(1N)}(I) - 1) \) were determined. A dependence \( T_a = f(I) \) was fitted to experimental points. It permits us to replace the equation

\[
n_{P(1N)}(I) - 1 = f(I)
\]

by

\[
n_{P(1N)}(T_a) - 1 = f(T_a)
\]

where \( T_a \) denotes the temperature of neutral atoms. For given \( \lambda_L \), calculated and interferometrically measured data were placed on the graph showing

\[
n_p(T_a, \lambda_L) - 1 = f(T_a).
\]

In the temperature range \( 10,000 < T_a < 11,200 \) K, where the applied diagnostic method gives good results, for two points, the mean weighted value of displacement \( \sigma = (n_{P(SP)} - n_{P(1N)}) \), between spectroscopic and interferometric data, was calculated. (Each value was weighted by the inverse of the square of its own error.) Then, interferometric points were shifted by \(-\delta\). Application of this procedure gives the results shown in figure 3.

![Figure 3. Comparison of the argon arc plasma refractions, measured interferometrically and calculated on the grounds of spectroscopic diagnostic.](image)

In order to describe a qualitative agreement between spectroscopic and interferometric data (shifted by \(-\delta\)), in the whole measured temperature range, the following expression was evaluated

\[
\Gamma(\lambda_L) = \sum_{j=1}^{M} [(n_{P(SP)}(T_a)_j - 1) - (n_{P(1N)}(T_a)_j - 1)]^2.
\]

\( M \) denotes the number of common points, for spectroscopic and interferometric data. For each \( \lambda_L \), the mean value, \( \Gamma_m(\lambda_L) \), of eight \( \Gamma(\lambda_L) \) values was calculated. Table 2
Table 2. Values of $\Gamma(\lambda)/\Gamma_m(\lambda)$. $\Gamma_m(\lambda)$ is the mean of eight $\Gamma(\lambda)$ values obtained for combinations of equations (2a)-(2c). The first three columns give the upper and lower limits applied for equations (2). The final column shows the sum of $\Gamma/\Gamma_m$ values for four wavelengths.

<table>
<thead>
<tr>
<th>$\lambda$ (nm)</th>
<th>$N_e = f(H_\beta)$</th>
<th>$\Delta E_i$</th>
<th>$N^*$</th>
<th>6328</th>
<th>5919</th>
<th>5803</th>
<th>4575</th>
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shows values of $\Gamma(\lambda_L)/\Gamma_m(\lambda_L)$. Smaller values of $\Gamma/\Gamma_m$ denote better agreement of spectroscopic and interferometric data.

5.3. Errors

The main source of possible systematic errors is the difference of arc window temperatures $\Delta T$. Quartz windows, 1.2 mm thick, absorb a small part of the arc radiation and warm up the surrounding gas. Variation of the optical path caused by changes of window thicknesses may be neglected. The difference in the temperature of windows is caused by the different lengths of arc channels. Windows of longer arc are warmer. Correction for this effect was made experimentally. The window temperature was measured as a function of the arc current. In our experimental conditions, for arc with $L = 14.6$ cm, $\Delta T$ was 11 °C when the arc current was changed from 35 to 90 A. For the same gas and cooling water flows, but without plasma, windows were warmed up by touching with 4 mm diameter warm metal cylinders. Cylinders were quickly removed and (after a short time of stabilisation) the interferogram was registered together with changes of window temperature. A Michelson interferometer was used in order to double the effect.

In figures 2 and 3 only statistical errors for interferometric data are shown. These error bands should be enlarged by possible systematic errors, which should not be larger than 3%. Error bands for spectroscopic points contain statistical errors and the contribution from the possible systematic error of $\Delta \alpha_{1/2}(H_\beta)$ taken as 4%.

6. Conclusions

Table 2 and figure 3 yield the following conclusions.

(i) For each $\lambda_L$, the smallest values of $\Gamma/\Gamma_m$ are in the sixth row. It means that the best agreement between spectroscopic and interferometric results exists when the upper limit is taken for equation (2a) and lower limits for (2b) and (2c). In this case, results of spectroscopic and interferometric plasma diagnostics overlap within the error bands; however interferometric data for each $\lambda_L$ are systematically below points calculated on the ground of the spectroscopic diagnostic. Two reasons are possible.
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First, the free-electron density calculated from equation (1) is too small. Knowledge of the calculated dependence \((n_{p,SP} - 1 = f(N_e))\) permits us to estimate how big \(N_e\) should be in order to get a perfect overlap of spectroscopic and interferometric data. Depending on \(\lambda_L\) and \(N_e\) range, estimated values of \(N_e\) are greater by 5-7%, than obtained from spectroscopic diagnostics (with the exception of the last two points for 4574 Å, for which disagreement is larger by 8 and 16%). It means that values of \(\Delta \alpha_{1/2}(H_B)\) applied in our calculations should be smaller by 3-5%.

Secondly, excited states give negative contribution to plasma refraction higher than expected. Of course, these two effects may occur together. This problem may be solved in a similar experiment, with two laser beams used at the same time. Then \(N_e\) may be obtained with the two-lambda technique and \(n_{PL}(I)\) is measured simultaneously for the same lambda.

(ii) The expression for lowering of the ionisation energy has a small, but not negligible, influence on the calculated plasma refraction. In our case, the influence of \(\Delta E_I\) is much smaller than the effect caused by differences in equation (1). Nevertheless, it may be possible to measure this influence, in a similar experiment, when problems described in (i) are solved.

Results of our experiment may be compared with previous ones. Baum et al (1975) measured a radial distribution of refraction for an argon arc. Their results for \(\lambda = 632.8\) nm are close to our interferometric data.

In the previous experiment (Musiol et al 1981), \((n_{PL} - 1)\) was measured with an interferometer of Ashby and Jephcott type. Interferometric results of this experiment agree within error bands with new measurements.

In all these experiments interferometric data overlap within error bands with points calculated from spectroscopic plasma diagnostics; however interferometric data lie a bit below calculated points.

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