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A program for the evaluation of electron number density from experimental hydrogen balmer beta line profiles☆

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Abstract

A program for the determination of plasma electron number density, $10^{20} \leq (N_e) \leq 10^{23}$ m^{-3}, from the comparison of experimental and theoretical hydrogen Balmer beta (H_β) line profiles is described in detail. Three theoretical data sets (one set is calculated within the framework of this paper) are included with the program and may be selected as a user's choice. Apart from N_e determination from the comparison of the whole experimental and theoretical profiles, this program offers a fast estimation of N_e from the halfwidth of the experimental line shape. If necessary, certain parts of the experimental profile may be neglected in the procedure of comparison with theory. This possibility enables the use of noisy line shape recordings for N_e determination. The H_β asymmetry study may be carried out by generating the difference between experimental and best-fitted theoretical line profiles. © 2002 Elsevier Science B.V. All rights reserved.

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1. Introduction

The electron number density, N_e, is one of the most significant parameters of fundamental importance for the characterization of spectrochemical plasma sources, like various types of arcs, Inductively Coupled Plasmas (ICPs), Microwave Induced Plasmas (MIPs), etc. Various methods for N_e determinations are well described in a number of textbooks and papers [1–3]. The determination of N_e from the profile of hydrogen Balmer lines and in particular from the H_β (486.13 nm) line is a well-established and widely used plasma diagnostic technique ([1–3] and references therein). To achieve the present state of accuracy, considerable experimental and theoretical efforts were involved [1,4] and, as a result, it was established that electron density in the range 10^{22}–10^{23} m^{-3} may be determined from the experimental H_β line widths, in conjunction with Vidal, Cooper and Smith (VCS) tables [5], with an accuracy of 4–7% [6,7]. The overall agreement of the experimental H_β profile with VCS [5] is good, except for the central part, where the theoretical profile always has a larger dip, whose magnitude depends upon both the electron number density and the mass of the perturbing ions present in the plasma. This discrepancy is related to the effect of ion-dynamics on the line shape [8]. Namely, the authors of VCS [5] used a quasistatic ion approximation to evaluate the ionic contribution to the line width. Ion dynamics, however, play an important role in the central part of the line profile. The details of various theoretical approaches and, in particular, the influence of ion dynamics on the hydrogen line shape calculations, can be found in [4] and references therein. In order to overcome the problem of the central dip in the H_β line profile, several programs, among those developed for using with VCS data tables [5] for N_e determination [9–13], neglect this part of the profile [12,13].

Recently, two theoretical calculations, one based on Model Microfield Method (MMM) by Stehle [14] and another one using computer simulation (CS) by Gigosos [15], have included ion dynamics in H_β line shape evaluations. This enabled the correct use of the whole H_β line profile, which is of importance for correct and reliable plasma diagnostics.

Numerous well-established methods for N_e diagnostics from H_β line profiles have been developed, generally based on two different approaches: (i) the use of approximate experimental formulas relating N_e with the Stark width and (ii) the comparison of measured and tabulated H_β profiles, evaluated for a range of electron number densities N_e, and electron temperatures, T_e. The use of approximate formulas is discussed in detail elsewhere [16]. In principle, this technique is based on the use of well-tested empirical formulas, relating Stark width with the electron number density. A similar method uses the dependence of the theoretical line width on N_e for N_e determination [17,18]. The advantages of these methods are simplicity and fast data reduction.

The comparison of measured and theoretical line shapes requires the fitting of the overall experimental profile with the theoretical one. This method is much more laborious than the previous one, but undoubtedly more correct and accurate, if the theoretical data describes well the overall line shape. Furthermore, in case of noisy spectra, the use of the whole profile instead of only its half-width is an advantage.

During the procedure of N_e determination from the comparison of experimental and theoretical line profiles, one meets two technical difficulties. One is related to the optimum fitting of experimental data and the other one is related to the comparison of the latter with theoretical data, which is nowadays available in the form of tables of line profiles computed for different N_e and T_e values. Thus, to use these data tables for comparison, one has to generate profiles and then apply a numerical procedure to determine intermediate theoretical profiles. Finally, when the accuracy of calculated N_e values have to be determined, one should consider, in addition to the quality of the experimental data, the results of experimental tests of the theoretical data used for comparison. As it was already pointed out, VCS tables have been carefully tested in the range 10^{22} $m^{-3} < N_e < 10^{23}$ m^{-3} [6], and the very same experimental data will be used here to test our program for N_e determination using VCS, MMM and CS. At lower electron densities, 10^{20} $m^{-3}<N_e<10^{22}$ m^{-3}, the results of two precision experiments [19,20] will be used here for theoretical data testing.

In this paper, a program for the determination of electron number density in the range $10^{20}<N_e<10^{23}$ m^{-3} from experimental H_β line shape recordings is presented. This range is typical of the majority of analytical plasma sources (typically $\sim 10^{21}$ in ICPs and 10^{20} m^{-3} in MIPs). For N_e lower than 10^{20} $m^{-3,}$ there is a lack of theoretical calculations due to the appearance of H_β fine structure [1]. For $N_e>10^{23}$ m^{-3}, asymmetries due to quadratic Stark effects and quadrupolar effects appear. None of the theoretical results used in this work have considered those effects. In sparks and laser-produced plasmas, N_e may exceed 10^{23} m^{-3} and, in these cases, the use of approximate formulas is simpler and more appropriate.

This program comprises three sets of theoretical data: tables for VCS and MMM, as well as new computer simulation (CS), results. The numerical procedures are developed for both experimental and theoretical data fittings, and attention is paid to N_e determination in the case of noisy line spectra recordings. Finally, the results of program testing and comparison with experimental data are presented.

2. Theoretical data

A computer simulation method [15] is used to evaluate H_β line profiles for different plasma compositions (various emitter-ion perturber reduced masses, μ) for electron number density values in the range $10^{20}<N_e<10^{23}$ m^{-3}. Calculated profiles may easily be used with non-equilibrium plasmas, with different electron and heavy particle temperatures.

The simulation technique used in the calculation of the line shapes considers a model of homogeneous and isotropic plasma, in which ions and electrons, separately, are in thermal equilibrium, so that their velocities are distributed according to the Maxwell law considering temperatures T_e and T_i that may be different. The simulation generates line shapes from the integration of the differential equation that describes the temporal evolution of an emitter submitted to the electric microfield created by surrounding ions and electrons.

The numerical process that allows us to determine the electric microfield sequence that alters the emitter is characterized by two parameters. The first parameter, the value of $\rho=r_0/r_D$, where: $r_0=(3/(4\pi N_e))^{1/3}$ is the mean distance between perturbers of the same kind and $r_D=(\varepsilon_0\ k\ T_e/(q_e^2\ N_e))^{1/2}$ is the Debye length, where ε_0 and k are the dielectric and Boltzmann constants respectively, while q_e is the electron charge. The second parameter is the quantity $(T_e/\mu_r)^{1/2}$, where μ_r is the relative reduced mass of the emitter-ion-perturber pair defined by $\mu_r=\mu\ T_e/T_i$ and μ is the real reduced mass. The first parameter, ρ, characterizes the magnitude of the electric field screening and, consequently, marks the statistical distribution of the electric field modulus. The second parameter establishes the mobility of the ionic perturbers in relation to the emitter. If ions have the same temperature as electrons, i.e. a one-temperature plasma, the corresponding value of μ_r is equal to the real reduced mass μ.

Finally, once the electric microfield sequence has been obtained, the electron number density N_e is fixed, the evolution equation of the emitter atom is integrated and the autocorrelation function of the emitter dipolar moment is obtained. This sequence of calculations is repeated a large number of times, so that representative sampling of the electric fields experienced by emitters in a plasma is taken into account. The Fourier transform of the average autocorrelation function gives the spectral profile. A detailed description of the simulation technique and of its applicability domains is given in [15]. The resulting H_β line profiles are tabulated in the electron number density range $10^{20}<N_e<10^{23}$ m^{-3} and presented as $S(\Delta\lambda)=f(\Delta\lambda)$ for various ρ, μ and N_e values. In the present computer simulations, as in the other two calculations [5,14], the fine structure splitting of hydrogen energy levels is neglected. Furthermore, it should be noted that the MMM data tables [14] contained in this program have been calculated for an emitter-perturber reduced mass $\mu=0.5$ (pure hydrogen plasma) and for $T_e=T_{i.}$ Therefore, one should not use these data with other plasmas.

3. Program concept and fit methodology

Three user options, CREATE, APPROXIMATE Ne and FIT, and are available within this program.

The option CREATE generates an output file of the H_β line profile, normalized according to intensity or area, from various data tables: VCS [5], MMM [14] and CS (computer simulations).

The option APPROXIMATE N_e allows one to evaluate N_e from the experimental H_β line width, using an empirical formula derived from experimental results [6] and published in [21]. This N_e determination may be performed with or without approximate deconvolution [21], i.e. separation of Stark broadening from Doppler and instrumental broadening.

The option FIT performs the calculation of N_e from the comparison of experimental profiles with various data tables, using an experimentally determined electron temperature and a reduced mass, which may be estimated from plasma composition. This option also allows the determination of N_e from the comparison of the part of the experimental H_β profile with the theoretical one, using variable statistical weights. This method may be used to decrease the influence of line background and/or noise. Furthermore, by generating in an output ASCII file, the difference between the theoretical and experimental profiles, the discrepancy between theory and experiment may easily be analyzed and asymmetries studied.

3.1. Generation of the intermediate I_{theory} ($\Delta\lambda$, μ, ρ, N_e)-CREATE option

The three included files VCS.inf, MMM.inf and CS.inf contain tabulated hydrogen H_β Stark profiles in the form of S (α) tables, where: $S(\alpha)=F_0\ I(\Delta\lambda)$, $\alpha=\Delta\lambda/F_0$ and where F_0 is the Holstmark field. In this way, a consistent presentation of all theoretical profiles is provided, in which all profiles are normalized according to area.

Therefore, we define S (α) data for:

Various μ, ρ, N_e values in the form of a matrix S (α, μ, ρ, log N_e), in the CS.inf file, where

$\alpha=\{0,3.98\text{e-}3,6.31\text{e-}3,\ldots,2.5,\}$
$\mu=\{0.5,0.8,0.9,1,1.5,\ldots,10\}$,
$\log\ N_e=\{20,20.33,20.66,\ldots,23\}$ and
$\rho=\{0.1,0.15,0.2,\ldots,0.6\}$, defined by the expression

$$N_e^{1/6}/T^{1/2}=(4\pi/3)^{1/3}\ (\varepsilon_0 k/q^2)^{1/2}\rho \tag{1}$$

Various T_e and N_e values in the form of a matrix S (α, T_e, log N_e), for the same α, where

$T_e=\{2.5,\ 5,\ 10$ and 20 kK$\}$,
$\log\ N_e=\{19,19.5,20,\ldots,23\}$ in the VCS.inf file,
and$T_e=\{2.5,5,10,20,40$ and 80 kK$\}$,
$\mu=0.5$ and the same log N_e values in the MMM.inf file.

The intermediate Stark profiles are obtained using:

- cubic spline interpolation for ρ (see Eq. (1)) as suggested by [10];
- linear interpolation for the real reduced mass, μ, in the case of CS tables; and
- cubic spline interpolation for log N_e as suggested by [10].

It is important to remember that, in the case of the generation of theoretical H_β Stark profiles from CS tables for non-equilibrium plasma conditions, i.e. $T_e \neq T_i$, the influence of ion dynamics is treated by introducing the relative reduced mass $\mu_r=\mu$ (T_e/T_i) on the basis of the ion velocity proportionality $\upsilon_i \sim (T_i/\mu)^{1/2}=(T_e/\mu_r)^{1/2}$ The MMM data tables used in this paper may only be applied to the generation of H_β Stark profiles for hydrogen plasmas in complete thermal equilibrium, i.e. $T_e=T_i$.

The resulting Stark profile for plasma parameters T_e, T_i, μ and N_e is then convoluted with Doppler and instrumental broadening:

$$I(\Delta\lambda) \propto \exp(-a\Delta\lambda^2/w_{DI}^2) \tag{2a}$$

$$W_{DI}=(W_D^2+W_I^2)^{0.5} \tag{2b}$$

$$W_D=3.58\cdot 10^{-7}\ \lambda_0\ \left(\frac{T_i}{M_i}\right)^{0.5} \tag{2c}$$

where: W_I and W_D are the instrumental and Doppler broadening halfwidth (HWHM), a is a constant and M_i is the ion mass. For the determination of various temperatures see [1–3,22]. The convolution of generated Stark profiles I (T_e, μ, N_e) with Doppler and instrumental broadening is performed using well-known routines for FFT [23,24]. Finally, the profile is exported to a file in the form of line intensity as a function of wavelength (in nm units) and is calculated from

$$I(\Delta\lambda)=S(\alpha)/F_0;\ \Delta\lambda=\alpha\ F_0$$

3.2. Estimation of the N_e–APPROXIMATE option

Whenever fast preliminary N_e data are needed from the experimental H_β profiles, a well-tested approximate formula [21,25] is used:

$$N_e\ [\mathrm{cm}^{-3}]=10^{16}\cdot\left(\frac{W_S}{4.7333}\right)^{1.49} \quad (3)$$

where W_S is the H_β Stark halfwidth at half maximum (in 0.1 nm units).

In the presence of Doppler and instrumental broadening, the deconvolution of experimental profiles is performed using the following approximate formula [21]

$$W_S=(W_m^{1.4}-W_{DI}^{1.4})^{1/1.4} \quad (4)$$

where: W_{DI} is given by Eq. (2b) and W_m is the measured H_β halfwidth at half maximum (in 0.1 nm units).

W_m is determined from experimental data in the following way:

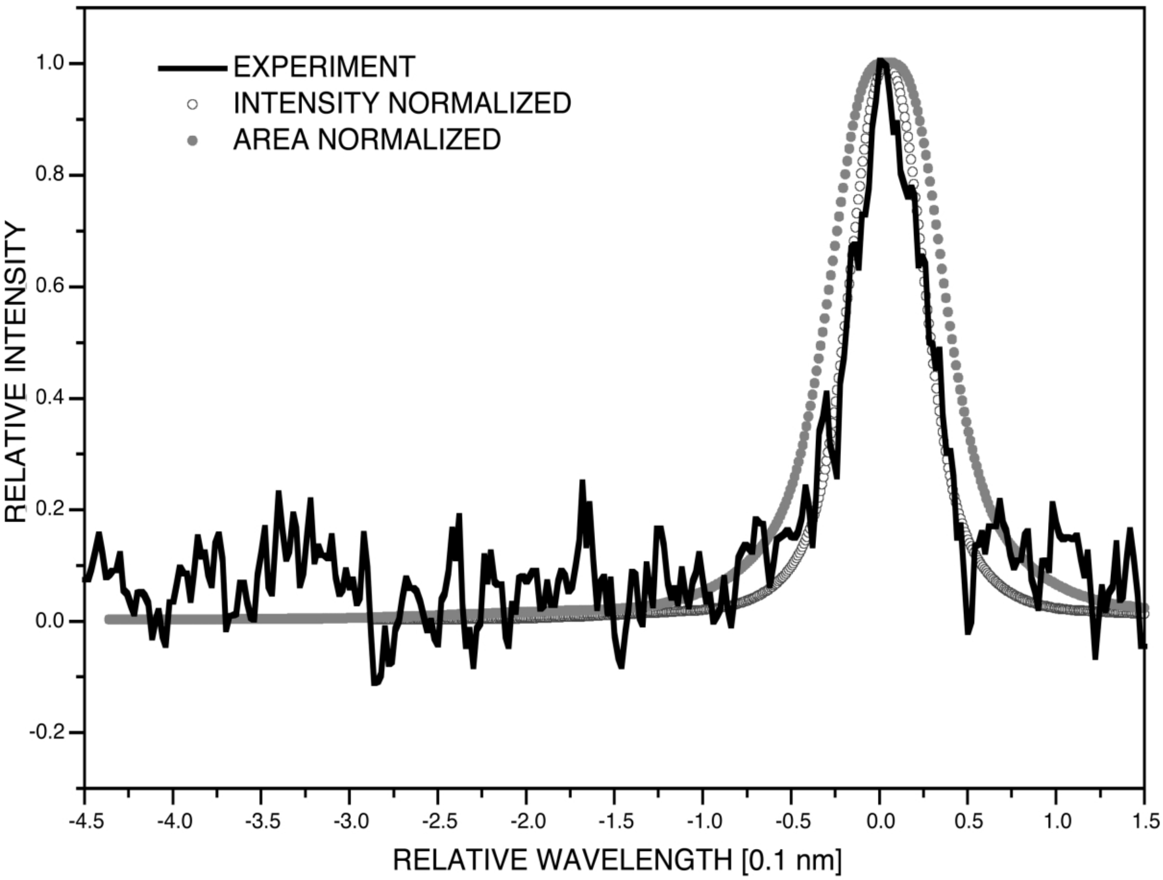

Fig. 1. Comparison of fitted profiles after normalization to: ● the area or to ○ the maximum intensity with weights $I_1=30$; $s_1=0$; $I_2=90$ $s_2=1$;$s_3=0$ set before least-square fitting.

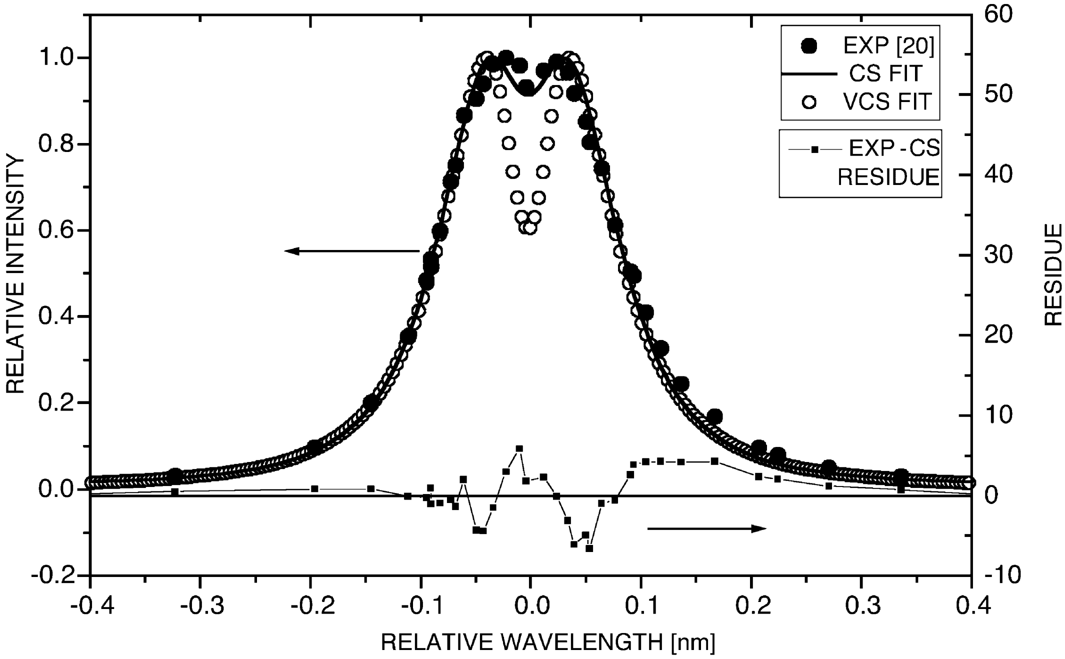

Fig. 2. Comparison of experimental D_β line profile [20] with the best fitted theoretical profiles according to VCS and CS methods. For experimental conditions see Table 1.

a. determination of intensity half maximum values, y;
b. determination of a data points pair, which satisfies $y(\mathrm{i})<y<y(\mathrm{i}+1)$;
c. determination of x (wavelength) for value y by linear interpolation between points $x(\mathrm{i})$, $y(\mathrm{i})$ and $x(\mathrm{i}+1)$, $y(\mathrm{i}+1)$;
d. procedure is repeated for the opposite side of the profile; and
e. W_{m} is determined as a mean arithmetic value.

3.3. Determination of N_e from the comparison of experimental and theoretical data—FIT option

First, the center of the experimental line profile is determined and the wavelength scale is shifted to center, which is taken as the zero point. Depending upon the user's choice, this is followed by line normalization according to intensity or area.

In the second step, the halfwidth of the experimental profile is determined and an approximate N_{e} is evaluated from Eqs. (2a), (2b), (2c), (3) and (4). This value is used as a starting N_{e} in the fitting procedure.

Then the minimum of the following function is determined:

$$\mathrm{chi}^2(N)=\sum[I_{\mathrm{exp}}(\Delta\lambda_{\mathrm{i}}) - I_{\mathrm{theory}}(\Delta\lambda_{\mathrm{i}},\mu,\rho;N_{\mathrm{e}})]^2 sig(i) \quad (5)$$

where N_e is a free parameter, while sig (i) is the statistical weight defined as

$$sig(i)=\begin{cases} s_1 \text{ for } I_{\mathrm{exp}}(\Delta\lambda_{\mathrm{i}})<I_1 \\ s_2 \text{ for } I_1<I_{\mathrm{exp}}(\Delta\lambda_{\mathrm{i}})<I_2 \\ s_3 \text{ for } I_2<I_{\mathrm{exp}}(\Delta\lambda_{\mathrm{i}}) \end{cases} \quad (6)$$

In the above expression, s_1, s_2, s_3, I_1, I_2 are the input parameters: percentage of amplitude I and statistical weights s, which can range between zero and one.

In this way $sig(i)$ allows the exclusion of certain parts of an experimental profile from the comparison with theory. This is particularly important for

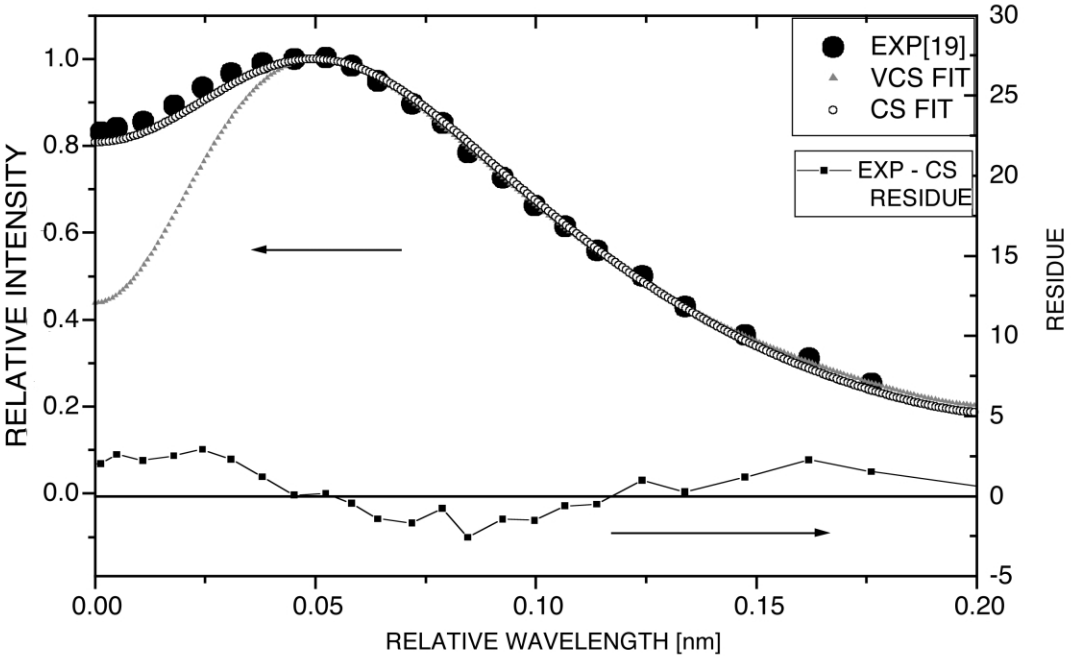

Fig. 3. Comparison of experimental H_β line profile [19] with the best fitted theoretical profiles according to VCS and CS methods. For experimental conditions see Table 1.

noisy line shape recordings, where background area is significant (may be as large as 20%) and area normalization thus becomes useless, as illustrated on Fig. 1 (on which a noisy H_β line profile is presented). In such cases, the actual least square fitting procedure should be preceded by line normalization, followed by the use of zero statistical weights ($s_1=0$) for data points with an intensity smaller than I_1 (30% for the example in Fig. 1) and larger than I_2 (90% in the same example) and unit statistical weight ($s_2=1$) for all data points with intensities between 30% and 90%. In this way, the best fit will be obtained without resorting to smoothing.

As illustrated in Fig. 1, the use of maximum intensity normalization and least square statistical weights allows the use of noisy line shape recordings for plasma diagnostic purposes. Furthermore, by introducing $s_1=1$ and $s_2=s_3=0$ this procedure may be used for the fitting of line wings only.

4. Program testing

Before the results of program testing are shown and discussed it is important to mention the following.

- The fitting of experimental data with MMM and CS data shown in Figs. 2 and 4 was performed with normalization, according to area and without using statistical weights.
- As a consequence of the way experimental data were presented in [19], the fitting of experimental data shown in Fig. 3 was performed with normalization according to the maximum intensity value.
- Due to previously discussed problems related to the central region of profiles, VCS data fitting was performed with intensity normalization and statistical weight $s=1$ for data points having intensities smaller than 60% of I_{max}, i.e. $I_1=60$, $s_1=1$, $I_2=80$, $s_2=0$, $s_3=0$ (see Eq. (6)).

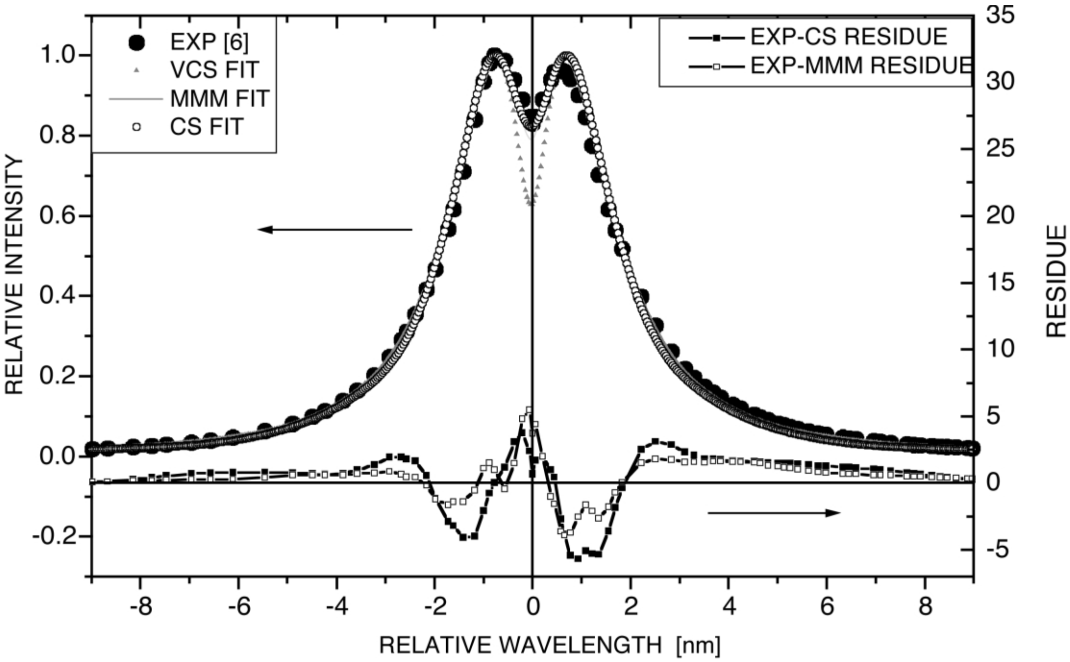

Fig. 4. Comparison of experimental H_β line profile [6,26] with the best fitted theoretical profiles according to VCS, MMM and CS methods. For experimental conditions see Table 1.

– Finally, it should be stressed that the process of digitizing of experimental data from original figures introduces a certain error in the comparison of theory and experiment.

Three previously published experiments were selected for program testing and comparison with experimental data [6,19,20]. They have in common that a wall-stabilized arc was used as the plasma

Table 1
Experimental conditions and results of comparison

Figure	2	3	4
Reference	[20]	[19]	[6]
Reduced mass μ	1.3	0.8	0.5
Electron temperature [K]	13500	17000	13400
Ion temperature [K]	8100	4500	13400
Instrumental FWHM [nm]	[a] 0	0.005	0.03
Experimental N_e [m^{-3}]	8.3 E+20	1.49 E+21	8.3 E+22
Approximate N_e Eqs. (2–4) [m^{-3}]	8.01 E+20	1.36 1E+21	7.85 E+22
VCS FIT N_e [m^{-3}]	8.77 E+20	1.41 E+21	8.17 E+22
MMM FIT N_e [m^{-3}]			7.66 E+22
CS FIT N_e [m^{-3}]	7.98 E+20	1.18 E+21	7.41 E+22
Experimental dip [%]	6	18	17.1
VCS FIT-dip [%]	40.7	56	37
MMM FIT-dip [%]			20.9
CS FIT-dip [%]	9.7	19	19.3

[a] Not explicitly stated.

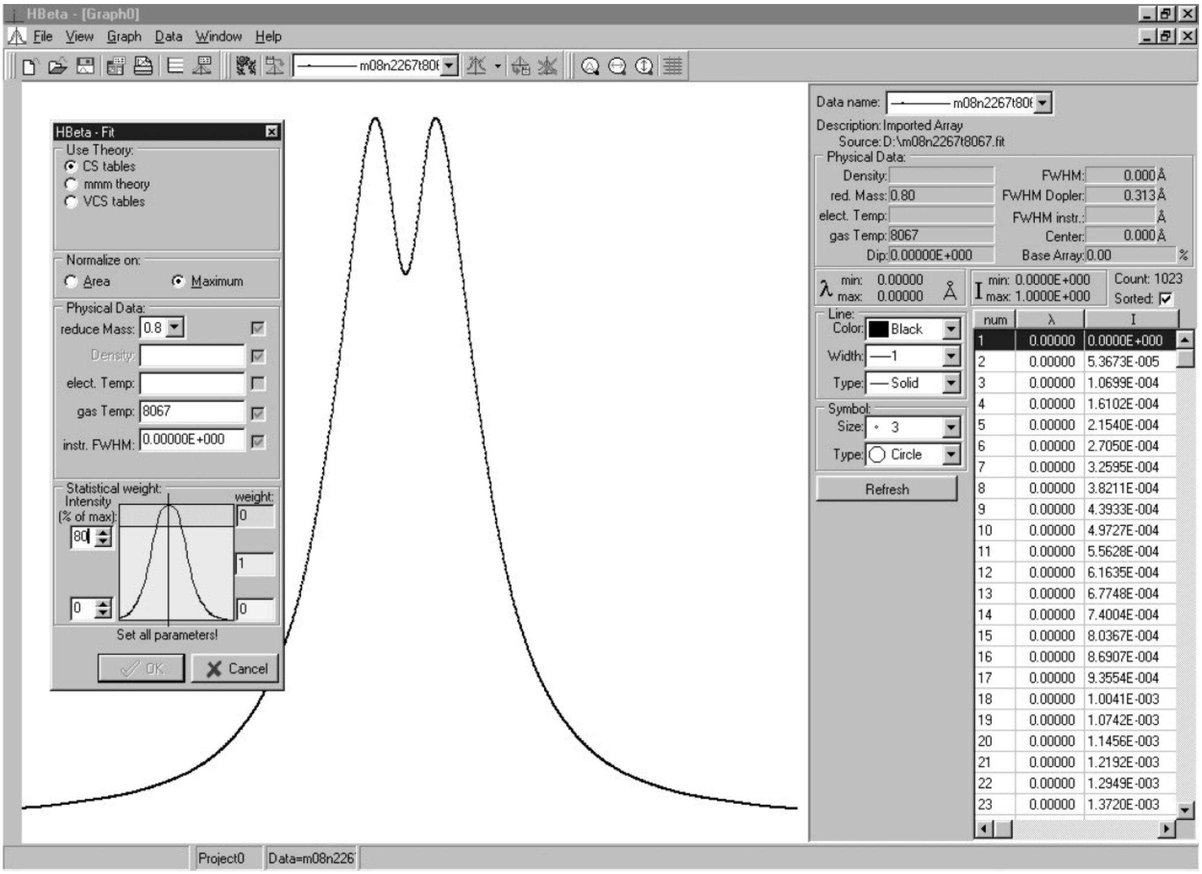

Fig. 5. Illustration of the Windows version main screen.

source. In [6], N_e was determined using the absolute line intensity. In [20], from which the D_β profile was used for program testing, the electron density was measured using two-wavelength laser interferometry, with a 7% accuracy. Finally, in [19], N_e was determined, like in the present case, from the comparison of the H_β profile with VCS tables; consequently this was used to test the influence of the digitalization procedure on the calculated results, and possibly also of the use of different numerical fitting procedures.

The comparison of experimental profiles [6,19,20], with their VCS, MMM and CS best fits, is illustrated in Figs. 2–4. All necessary experimental data required for fitting experimental profiles with theoretical data are given in Table 1, together with the results. From this comparison, the following conclusions may be drawn.

- The CS method provides the best fit to the overall line profile, as shown, in particular, by the good agreement between experimental and calculated dip values (Table 1). This conclusion extends to the MMM method insofar as the latter has only been tested with data from ref. [6].
- The approximate formulas (Eqs. (2–4)) agree, within the estimated experimental error, with experimental results for N_e.
- The electron density values, calculated using the VCS data, agree within 6% with experiments in [6] and [20]. Here, one should bear in mind

that the central part of the profile was neglected in this comparison.

- Since the authors of [19] determined N_e using the VCS data tables, the agreement between their N_e values and those obtained in the present work, using the same tables, was expected. The difference of 6% is most likely due to the digitalization of the H_β profile from the printed figure.
- Electron density values obtained with the CS method are systematically smaller than experimental values, as is the case for the MMM result for [6], albeit to a lesser extent.
- All three theoretical methods (VCS, MMM and CS) predict fully symmetrical profiles, while asymmetry may exist in experimental line profiles. For example, the difference between the experimental and CS-fitted $D_{\beta 0}$ profiles (Fig. 2) is much larger on the red wing than on the blue wing, where the agreement is very good. In the case of the H_β profile determined at higher N_e (Fig. 4), the fitting residues for the MMM and CS methods are more symmetric, but nevertheless show a higher discrepancy on the red wing, for both sets of theoretical data. It should be stressed that this discrepancy cannot be explained by a trivial asymmetry caused by the transformation of angular frequency to wavelength units.

As a final conclusion, it is apparent from the above observations that N_e determination from experimentally measured line profiles will undoubtedly benefit from both theoretical improvements and high precision experimental testing.

5. Conclusions

The main application of this program is the evaluation of electron number density in the range $10^{20} < N_e < 10^{23}$ m^{-3} from experimental H_β line profiles. The program may also be used to generate theoretical line profiles, for comparison with experimental profiles. The main properties of this program are as follows.

- Three sets of theoretical data tables, VCS [5], MMM [14] and CS (Computer Simulation) calculated here after [15] may be used to determine plasma electron number density. The MMM data tables were calculated only for pure hydrogen plasmas (reduced mass emitter-ion perturber $\mu = 0.5$) in thermal equilibrium, $T_e = T_{i,g}$. The CS method takes into account ion dynamics (it can be used for different μ values) and may be applied to non-thermal plasmas, $T_e \neq T_{i,g}$. The VCS data was calculated assuming the quasistatic ion approximation $\mu = \infty$, and may also be applied to non-thermal plasmas, $T_e \neq T_{i,g}$.
- It is very simple to extend or include new data tables in addition to the present ones.
- Using simple approximate formulas, fast and accurate determination of N_e from the halfwidth of the experimental H_β line profile can be performed.
- The program allows the user to select among the VCS, MMM and CS methods as well as the type of normalization (unit area or intensity maximum) used in the fitting procedure. Furthermore, by using statistical weights certain parts of the experimental profile may be excluded from the calculations.
- The difference between experimental and theoretical profiles is provided in an output file, which facilitates the analysis of H_β line asymmetry, as well as providing a good indicator of the quality of the fitting procedure.

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We wish to thank C. Stehlé for supplying us with MMM data tables and V. Helbig for the approximate relation $N_e = f$ (HWHM). We thank also W.L. Wiese for supplying us with the experimental H_β profile presented in Fig. 3.

Appendix A:

This program is offered in the DOS version only. A Windows version is available upon request from: *zikic@gibagroup.com.* The possibilities and advantages of the Windows version are described below.

The Windows version of the program is an object-oriented windows 32-bit application for Win 95 and Win NT system platforms. It consists of two parts, the first one being the user-friendly graphical interface, and the second one the fitting routines collected in a FORTRAN dynamic link library. The graphical interface is written in Borland Delphi, and all relevant mathematical routines are in the MS FORTRAN Power Station. This concept provides easy migration to other platforms like Unix or Mac, or machine independent ones like Java.

The Windows version of the program enables:

- graphic presentation and printing of experimental and fitted curves;
- user-friendly interface with drop-down menus, toolbar icons and additional windows for entering and viewing physical parameters (N_e, T_e, T_i, μ, FWHM, approximate N_e, center position, Doppler FWHM and dip);
- viewing of data tables (file name, file location, number of points, wavelength range and all x,y data values);
- manipulation of data (smooth, normalize, scale, shift, editing/deleting and truncate); and
- the ability to work simultaneously with several projects, each having many graphic/data windows.

An illustration of the Windows version main screen is presented in Fig. 5.

Appendix B: Copyright

Appendix C: Disclaimer

Neither the authors nor the Publisher warrant that the data files are free from defects, or that the documentation is accurate. Neither the authors nor the Publisher are liable for any damage of whatever kind sustained through downloading and/or using the data files. By downloading and/or using the data files the reader of *Spectrochimica Acta Electronica*, acting as a user of an electronic publication therein, agrees to the above terms and conditions.

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