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Role of Penetrating Ions in the Method of Measuring the Electron Density by the Asymmetry of Hydrogenic Spectral Lines in Plasmas

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ABSTRACT: A new diagnostic method for measuring the electron density N_e using the asymmetry of hydrogenic spectral lines in dense plasmas was previously proposed and implemented in several experiments. The method has the following advantages compared to the method of deducing N_e from the experimental widths of spectral lines. First, the latter, traditional method requires measuring widths of at least two spectral lines (because the widths are affected not only by the Stark broadening, but also by competing broadening mechanisms, such as, e.g., the Doppler broadening.), while for the new diagnostic method it is sufficient to obtain the experimental profile of just one spectral line. Second, the traditional method based on the experimental widths would be difficult to implement if the center of the spectral lines is optically thick. In distinction, the new diagnostic method can still be used even if the spectral line is optically thick in its central part. In the theory underlying this new diagnostic method, the contribution of plasma ions to the spectral line asymmetry was calculated only for configurations where the perturbing ions are outside the bound electron cloud of the radiating atom/ion (non-penetrating configurations). In the present paper we take into the contribution to the spectral line asymmetry from *penetrating configurations* where the perturbing ion is inside the bound electron cloud of the radiating atom/ion. We show that in high density plasmas, the allowance for penetrating ions can result in significant corrections to the electron density deduced from the spectral line asymmetry.

Key words: spectroscopic diagnostics of plasmas, electron density measurements, asymmetry of spectral lines, penetrating configurations

1. INTRODUCTION

The asymmetry of hydrogenic spectral lines in dense plasmas is primarily caused by the nonuniformity of the ion microfield, as noted by Sholin and his co-workers in papers [1-3] (for the latest advances in the theory of the asymmetry we refer to papers [4, 5] and references therein). Typically the blue maximum of the spectral line is higher than the red maximum, and the positions of the intensity maxima are asymmetrical with respect to the unperturbed line center.

A new diagnostic method for measuring the electron density using the asymmetry of hydrogenic spectral lines in dense plasmas was proposed and implemented in paper [6]. In that paper, in particular, from the experimental asymmetry of the C VI Lyman-delta line emitted by a vacuum spark discharge, the electron density was deduced to be $N_e = 3\text{x}10^{20}$ cm^{-3}. This value of N_e was in a good agreement with the electron density determined from the experimental widths of C VI Lyman-beta and Lyman-delta lines.

Later this diagnostic method was employed also in the experiment presented in paper [7]. In that laser-induced breakdown spectroscopy experiment, the electron density $N_e \sim 3\text{x}10^{17}$ cm^{-3} was determined from the experimental asymmetry of the H I Balmer-beta (H-beta) line.

This new diagnostic method has the following advantages compared to the method of deducing N_e from the experimental widths of spectral lines. First, the latter, traditional method requires measuring widths of at least two

spectral lines – because the widths are affected not only by the Stark broadening, but also by competing broadening mechanisms, such as, e.g., the Doppler broadening. In distinction, for using the new diagnostic method it is sufficient to obtain the experimental profile of just one spectral line – because the Doppler broadening does not cause the asymmetry.

Second, the traditional method based on the experimental widths would be difficult to implement if the center of the spectral lines is optically thick. In distinction, the new diagnostic method can still be used even if the spectral line is optically thick in its central part. This is because the overwhelming contribution to the asymmetry originates from the wings of the spectral line, the wings being usually optically thin. More details can be found in Sect. 1.6 of book [8] */.

In the theory underlying this new diagnostic method, the contribution of plasma ions to the spectral line asymmetry was calculated only for configurations where the perturbing ions are outside the "atomic sphere", i.e., outside the bound electron cloud of the radiating atom/ion (non-penetrating configurations). In the present paper we take into the contribution to the spectral line asymmetry from *penetrating configurations*, i.e., from the configurations where the perturbing ion is inside the bound electron cloud of the radiating atom/ion (hereafter, radiator). We show that in high density plasmas, the allowance for penetrating ions can result in significant corrections to the electron density deduced from the spectral line asymmetry.

2. ALLOWANCE FOR PENETRATING IONS

Let us first present a brief overview of the underlying theory for non-penetrating configurations. The dipole interaction of the radiator with perturbing ions outside the bound electron cloud splits the spectral line into Stark components symmetrically with respect to the unperturbed frequency or wavelength – in terms of both positions and intensities of the Stark components. The quadrupole interactions of the radiator with perturbing ions outside the bound electron cloud causes the asymmetry of the Stark splitting – in terms of both positions and intensities of the Stark components.

However, in paper [9] it was shown that the quadrupole interaction, despite casing the asymmetric splitting of the spectral line into Stark components, does not shift the center of gravity of the line profile. Therefore, in the new diagnostic method presented in paper [6], first the center of gravity of the experimental profile was determined and then it was taken as the reference point. Then with respect to this point, the integrated intensities of the blue (I_B) and red (I_R) wings of the experimental profile were found. After that, the experimental degree of asymmetry, defined as

$$\rho_{quad} = \frac{I_B - I_R}{0.5[I_B + I_R]}, \tag{1}$$

was determined and then compared with the corresponding theoretical value given below.

The theoretical intensities of the blue and red wings, resulting from dipole and quadrupole interactions of the radiator with perturbing ions outside the bound electron cloud, can be expressed as follows (see paper [6]):

$$I_B = \sum_{k>0} I_k^{(0)} \left(1 + \frac{Z_p a_o}{Z_r^2 R_o} \epsilon_k^{(1)} \langle R_0 / R \rangle \right), \tag{2}$$

and

$$I_R = \sum_{k<0} I_k^{(0)} \left(1 + \frac{Z_p a_o}{Z_r^2 R_o} \epsilon_k^{(1)} \langle R_0 / R \rangle \right), \tag{3}$$

*/ We note that Ref. [40] from Ch. 1 of [8] on the paper referred here as [6] has typographic errors. The correct one is our Ref. [6] here.

where Z_p is the charge of perturbing ions, Z_r is the nuclear charge of the radiator, a_o is the Bohr radius, and $R_o = [(4\pi/3)N_p]^{-1/3}$ is the mean interionic distance, $N_p = N_e / Z_p$ being the perturbing ion density. Here $I_k^{(0)}$ and $\epsilon_k^{(1)}$ are the unperturbed intensity and the quadrupole correction to the intensity, respectively, the subscript k being the label of Stark components of the spectral line: $k > 0$ and $k < 0$ correspond to the blue-shifted and red-shifted components, respectively (the values of $I_k^{(0)}$ and $\epsilon_k^{(1)}$ for several Lyman and Balmer lines were tabulated in paper [2]). The quantity $<R_0/R>$ is the scaled inverse distance between the perturbing ion and the radiator averaged over the distribution of such distances.

Finally, the theoretical degree of asymmetry was presented in paper [6] in the form:

$$\rho_{quad} = 0.46204 \left(\frac{N_e[cm^{-3}]}{10^{21}} \right)^{\frac{1}{3}} \frac{Z_p^{\frac{2}{3}}}{Z_r^2} \sum_{k>0} I_k^{(0)} \epsilon_k^{(1)}, \tag{4}$$

Then the electron density N_e was determined in paper [6] by substituting the experimental degree of asymmetry into the left side of Eq. (4).

In the present paper we seek to add the contribution of penetrating ions to the spectral line asymmetry – in order to refine this diagnostic method. To get the message across in a simple form, we limit ourselves below to the practically important case $Z_p = Z_r = Z$. The energy shifts due to penetrating ions can be calculated by the perturbation theory in the basis of the spherical wave functions of the so-called "united atom" of the nuclear charge $2Z$.

The perturbed energy shifts (counted from the unperturbed energies) for the orbital quantum number $l > 0$ are given by (see, e.g., Eqs. (6) and (7) from paper [10] or Eqs. (5.11), (5.12) from book [11]):

$$E_{nlm} = -\frac{8[1(1+1) - 3m^2]\, Z^4 R^2 e^2}{a_o^3 n^3 1(l+1)(2l-1)(2l+1)(2l+3)}. \tag{5}$$

For the case of $l = 0$, the calculated energy shift is:

$$E_{n00} = \frac{8 Z^4 R^2 e^2}{3\, a_o^3 n^3}. \tag{6}$$

We note that Eq. (6) can be also obtained from Eq. (5), first by setting $m = 0$, and then by cancelling out $l(l + 1)$ in the numerator and denominator, and by setting $l = 0$. (This was mentioned in book [11], but in Eq. (5.11) from [11] corresponding to our Eq. (6), there was a typographic error in the sign.)

The frequency change of an individual Stark component is thus given by

$$\Delta\omega_k = -\frac{Z^2 e^2 \Delta_k^1}{2\hbar a_o^3} R^2, \tag{7}$$

where

$$\Delta_k^1 = 16 Z^2 \left[\frac{l(1+1) - 3m^2}{n^3 l(l+1)(2l-1)(2l+1)(2l+3)} - \frac{l'(l'+1) - 3m'^2}{n'^3 l'(l'+1)(2l'-1)(2l'+1)(2l'+3)} \right]. \tag{8}$$

For the specific case where either $l = 0$ or $l' = 0$, Eq. (8) reduces to

$$\Delta_k^1 = \begin{cases} 16Z^2\left[\dfrac{1}{3n^3} - \dfrac{l'(l'+1)-3m'^2}{n'^3 l'(l'+1)(2l'-1)(2l'+1)(2l'+3)}\right], & l=0; l'\neq 0 \\ 16Z^2\left[\dfrac{l(l+1)-3m^2}{n^3 l(l+1)(2l-1)(2l+1)(2l+3)} - \dfrac{1}{3n'^3}\right], & l'=0; l\neq 0 \end{cases}. \tag{9}$$

Then the quasistatic profile of each Stark component can be represented in the form:

$$S_k(\Delta\lambda) = \int_0^{u_{\max}} W(u)\left[I_k^{(0)} + I_k^{(1)}\right]\delta\left(\Delta\lambda - \frac{Z^2 e^2 \Delta_k^1 R_o^2}{2\hbar a_o}u\right)du. \tag{10}$$

Here, $u \equiv R^2$, and the probability of finding the perturbing ion a distance u away from the radiating atom is taken to be the binary distribution. For simplifying the integration, we use the expansion of the distribution in powers u/R_0^2 and keep the terms up to $\sim u^2$:

$$W(u)du = \frac{3}{2}\frac{\sqrt{u}}{R_o^3}\exp\left(-\frac{\sqrt{u}^3}{R_o^3}\right)du \approx \frac{3}{2}\frac{\sqrt{u}}{R_o^3} - \frac{3}{2}\frac{u^2}{R_o^6}. \tag{11}$$

For the case of a hydrogenic radiator under the presence of a penetrating ion, the relative intensities of each line component can be best calculated analytically using a robust perturbation theory developed by Oks and Uzer [12]. A more detailed explanation of this procedure is outlined in Appendix A. The relative intensities of each component can be written as

$$I_k = \Delta_{Ik}^0 + Z^2 \Delta_{Ik}^1 u^2, \tag{12}$$

where Δ_{Ik}^0 and Δ_{Ik}^1 are tabulated in Appendix B for each component of the spectral line Balmer-alpha, considered here as an example.

The upper limit u_{max} of the integration in Eq. (10) should be the smallest of the following two "candidates". One candidate for u_{max} is the root mean square size of the bound electron cloud, which depends on the sublevel in consideration:

$$r_{rms} = \sqrt{\frac{n^2}{2Z^2}[5n^2 + 1 - 3l(l+1)]}. \tag{13}$$

The other candidate for u_{max} is defined by the limit of the applicability of the perturbation theory. Of course, this would ensure that formally calculated corrections to the energy and intensities of the spectral line would remain relatively small.

The allowance for penetrating ions shifts the center of gravity of the spectral line, as shown in paper [13]. This is the only contribution to the shift of the center of gravity since the dipole and quadrupole interactions of the radiator with perturbing ions outside the bound electron cloud do not shift the center of gravity, as shown in paper [9] and mentioned above. For the He II Balmer-alpha line, which we use as an example, the center of gravity shift due to penetrating ions was calculated analytically in paper [13] to be

$$\Delta\lambda_{PI}(mA) = 17\, N_e(cm^{-3})/10^{17}. \tag{14}$$

The shift of the center of gravity by this amount serves as the reference point for calculating the integrated intensities of the blue and red wings with the allowance for penetrating ions.

After carrying out the integration in Eq. (10), the profile reduces to

$$S_k(\Delta\lambda) = \frac{1}{|\Delta_k^1|Z^4} I_k\left[\frac{2\pi c|\Delta\lambda|}{\lambda_o^2\Delta_k^1 Z^4}\right]\left(\frac{3\left(\frac{2\pi c|\Delta\lambda|}{\lambda_o^2\Delta_k^1 Z^4}\right)^{\frac{1}{2}}}{2R_o^3} - \frac{3\left(\frac{2\pi c|\Delta\lambda|}{\lambda_o^2\Delta_k^1 Z^4}\right)^{2}}{2R_o^6}\right)\Theta\left[\frac{u_{\max}\lambda_o^2\Delta_k^1 Z^4}{2\pi c} - |\Delta\lambda|\right], \tag{15}$$

where $\Theta[...]$ is the Heaviside step function. Thus, for the contributions of the penetrating ions to the red and blue parts of the integrated profile, we obtain

$$I_{PI,B} = \sum_{k<0}\int_{-\Delta\lambda_{\max}}^{\Delta\lambda_{PI}} S_k(\Delta\lambda)\, d\Delta\lambda \tag{16}$$

and

$$I_{PI,R} = \sum_{k>0}\int_{-\Delta\lambda_{PI}}^{\Delta\lambda_{\max}} S_k(\Delta\lambda)\, d\Delta\lambda, \tag{17}$$

respectively. Here

$$\Delta\lambda_{\max} = u_{\max}\lambda_o^2\Delta_k^1 Z^4/(2\pi c), \tag{18}$$

which is obtained by equating to zero the argument of the Heaviside step function. Additionally, what is meant in Eqs. (16) and (17) by $k < 0$ (or $k > 0$) is the inclusion of only those components which involve corrections to the energy which are positive (or negative), implying a blue-shifted (or red-shifted) component of the spectral line.

By combining the above result with the contribution of the quadrupole interaction (the interaction of the radiator with perturbing ions outside the bound electron cloud) to the integrated intensities of the blue and read parts of the profile, we obtain our final result for the degree of asymmetry

$$\rho_{act} = \frac{I_B + I_{PI,B} - I_R - I_{PI,R}}{0.5[I_B + I_{PI,B} + I_R + I_{PI,R}]}, \tag{19}$$

where subscript *act* stands for *actual* – in distinction to ρ_{quad}.

The combination of Eqs. (4) and (19) connects the degree of asymmetry with the electron density N_e and thus allows a more accurate determination of the electron density from the experimental asymmetry. We illustrate this below by the example of the He II Balmer-alpha line.

Table 1 presents the following quantities for the He II Balmer-alpha line at five different values of the actual electron density:

- the theoretical degree of asymmetry ρ_{act} calculated with the allowance for penetrating ions,
- the theoretical degree of asymmetry ρ_{quad} calculated without the allowance for penetrating ions,
- the electron density $N_{e,quad}$ that would be deduced from the experimental degree of asymmetry while disregarding the contribution of the penetrating ions,

– the relative error $|N_{e,quad} - N_{e,act}| / N_{e,act}$ in determining the electron density from the experimental degree of asymmetry while disregarding the contribution of the penetrating ions.

$N_{e,act}/(10^{18}cm^{-3})$	2	4	6	8	10
ρ_{act}	0.09254	0.11420	0.12837	0.13892	0.14727
ρ_{quad}	0.09550	0.12033	0.13774	0.15160	0.16331
$N_{e,quad}/(10^{18}cm^{-3})$	1.81936	3.41983	4.85657	6.15540	7.33363
$\lvert N_{e,quad} - N_{e,act} \rvert / N_{e,act}$	0.09032	0.14504	0.19057	0.23057	0.26663

Table 1 the relative error in determining the electron density N_e from the experimental asymmetry degree while disregarding the contribution of the penetrating ions for the He II Balmer-alpha line. The physical quantities in Table 1 are explained in the text directly above Table 1.

It is seen that in high density plasmas, the allowance for penetrating ions can indeed result in significant corrections to the electron density deduced from the spectral line asymmetry.

3. CONCLUSIONS

For improving the diagnostic method for measuring the electron density using the asymmetry of spectral lines in dense plasmas, we took into consideration the contribution to the spectral line asymmetry from *penetrating configurations*, i.e., from the configurations where the perturbing ion is inside the bound electron cloud of the radiating atom/ion. After performing the corresponding analytical calculations, we demonstrated that in high density plasmas the allowance for penetrating ions can result in significant corrections to the electron density deduced from the spectral line asymmetry.

We note that the electron densities $N_e \sim (10^{18} - 10^{19})$ cm^{-3}, which we used in the illustrative example of the He II Balmer-alpha line, are achievable in plasma spectroscopy. Examples are experiment [14] with a hydrogen plasma and experiment [15] with a helium plasma.

APPENDIX A. DETAILS OF CALCULATING PERTURBED MATRIX ELEMENTS

The redistribution of intensities of Stark components, along with wavelength shifts due to the presence of perturbing ions, play a crucial role in determining the degree of asymmetry of the spectral line. These values have been tabulated according to the robust perturbation theory developed by Oks and Uzer [12] based on using the super-generalized Runge-Lenz vector derived by Kryukov and Oks [16]. Since the unperturbed system has an additional constant of the motion (namely the Runge-Lenz vector), then the task of calculating the corrections to the state is simplified. The reason for this beneficial result is the correction to the Runge-Lenz vector is non-degenerate with respect to the same states which are degenerate in the correction to the Hamiltonian. The mixing of the states is elucidated by the Runge-Lenz vector correction under the influence of the perturbing ion. Here are some details, with formulas being presented in atomic units.

According to paper [16], for the problem of an electron in the field of two Coulomb centers of charges Z_1 and Z_2, the additional conserved quantity is the following projection of the super-generalized Runge-Lenz vector on the internuclear axis

$$A_z = \mathbf{p} \times \mathbf{L} \cdot \mathbf{e}_z - \frac{L^2}{R} - Z_1 \frac{z}{r} - Z_2 \frac{R - z}{|\mathbf{R} - \mathbf{r}|} + Z_2, \tag{A.1}$$

where **p**, **L**, and **r** are the linear momentum, the angular momentum, and the radius-vector of the electron, respectively; **R** is the vector directed from charge Z_1 to charge Z_2. For the case where $R << r$, the unperturbed part A_{z0} of the operator A_z can be chosen as

$$A_{z0} = -\frac{L^2}{R}, \tag{A.2}$$

corresponding to the unperturbed Hamiltonian of the so-called "united atom" of the nuclear charge $Z_1 + Z_2$:

$$H_0 = \frac{p^2}{2} - \frac{Z_1 + Z_2}{r}, \tag{A.3}$$

Operators H_0 and A_{z0} have common eigenfunctions (the spherical eigenfunctions of the Coulomb problem). The spectrum of eigenvalues of the operator H_0 is degenerate. Therefore, calculating corrections to the eigenfunctions of the operator H_0 using the standard perturbation theory would require going to the 2nd order of the *degenerate* perturbation theory, thus involving generally infinite summations (see, e.g., the textbook [17]).

In distinction, the spectrum of eigenvalues of the operator A_{z0} is nondegenerate (the eigenvalues being $-l(l+1)/R$). Therefore, the corrections to the eigenfunctions can be easily calculated in the 1st order of the standard *nondegenerate* perturbation theory. The coefficients of the corresponding linear combinations of the unperturbed eigenfunctions are

$$< nl'm \mid A_z - A_{z0} \mid nlm >= \frac{l'(l'+1) - l(l+1)}{R} \tag{A.4}$$

and do not involve infinite summations. This example is another illustration of the advantages of the robust perturbation theory developed in paper [12] over the standard perturbation theory.

In this way, we obtained the following expression for the 1st order corrections to the eigenfunctions for the specific case of $Z_1 = Z_2 = Z$:

$$\Psi^{(1)}_{nlm} = \frac{5\left[\frac{(l_>^2 - m^2)(n^2 - l_>^2)}{(2l_> + 1)(2l_> - 1)}\right]^{\frac{1}{2}}}{n\,[l(l+1) - l'(l'-1)]} Z\,R\,\Psi^{(0)}_{nl'm'}, \tag{A.5}$$

where $l_>$ denotes the greater value between l and l'. The selection rules are $l' = l \pm 1$ and $m' = m$.

We note that in the opposite case, where $R >> r$, the unperturbed part $A_{z1,0}$ of the operator A_z can be chosen in the usual way

$$A_{z1,0} = z\,p^2 - p_z(\mathbf{rp}) - Z_1\frac{z}{r}, \tag{A.6}$$

where the notation (**rp**) stands for the scalar product (also known as the dot-product) of the operators **r** and **p**. The corresponding unperturbed Hamiltonian is

$$H_{1,0} = \frac{p^2}{2} - \frac{Z_1}{r}, \tag{A.7}$$

The operator $A_{z1,0}$ has a nondegenerate spectrum of eigenvalues equal to q / n, where $q = (n_1 - n_2)$ is the difference of the parabolic quantum numbers. Therefore, the first nonvanishing corrections to the common eigenfunctions of the operators $H_{1,0}$ and $A_{z1,0}$ can be easily calculated in the 1st order of the standard *nondegenerate* perturbation theory. The coefficients of the corresponding linear combinations of the unperturbed eigenfunctions

are

$$< nl'm \mid L^2 \mid nlm >= \frac{\frac{q'}{n} - \frac{q}{n}}{R}, \tag{A.8}$$

where $|q' - q| = 2$, as follows from the selection rules.

In distinction, for obtaining the same corrections to the eigenfunctions using the operator $H_{1,0}$, whose spectrum of eigenvalues is degenerate, it would require going to the 2nd order of the *degenerate* perturbation theory and dealing with its complications, as Sholin did in his paper [2].

APPENDIX B. TABLE OF INTENSITIES AND ENERGY LEVEL CORRECTIONS FOR THE HE II BALMER-ALPHA LINE

The perturbed intensity and frequency corrections for He II Balmer-aslpha line are presented below. The quantum numbers of the upper and lower sublevels are in the spherical quantization.

Upper sublevel	*Lower sublevel*	Δ^0_{lk}	Δ^1_{lk}	Δ^1_k
322	211	$\frac{768}{4715}$	0	$\frac{173}{5670}$
321	211	$\frac{384}{4715}$	$-\frac{32}{14145}$	$\frac{197}{5670}$
321	210	$\frac{384}{4715}$	$-\frac{64}{2829}$	$-\frac{37}{567}$
321	200	0	$\frac{2792}{127305}$	$\frac{949}{2835}$
320	211	$\frac{128}{4715}$	$-\frac{128}{127305}$	$\frac{41}{1134}$
320	210	$\frac{512}{4715}$	$-\frac{3968}{127305}$	$-\frac{181}{2835}$
320	200	0	$\frac{11168}{381915}$	$\frac{953}{2835}$
311	211	0	$\frac{32}{14145}$	$\frac{19}{810}$
311	210	0	$\frac{232}{14145}$	$-\frac{31}{405}$
311	200	$\frac{160}{2829}$	$-\frac{400}{25461}$	$\frac{131}{405}$
310	211	0	$\frac{8}{3105}$	$\frac{43}{810}$
310	210	0	$\frac{2512}{127305}$	$-\frac{19}{405}$

310	200	$\frac{160}{2829}$	$-\frac{280}{8487}$	$\frac{143}{405}$
300	211	$\frac{5}{943}$	$-\frac{40}{25461}$	$-\frac{53}{810}$
300	210	$\frac{5}{943}$	$-\frac{295}{101844}$	$-\frac{67}{405}$
300	200	0	$\frac{5525}{305532}$	$\frac{19}{81}$

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