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An approach to a non-LTE Saha equation based on the Druyvesteyn energy distribution function: a comparison between the electron temperature obtained from OES and the Langmuir probe analysis

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Abstract
Plasma diagnostics using the optical emission spectroscopy (OES) technique is often based on the assumption of Maxwellian distribution functions. Whenever the equilibrium condition is not fulfilled, the electron energy distribution function (EEDF) is no longer Maxwellian and there appear discrepancies between the electron temperature obtained via the spectral line ratio (e.g. Saha equation) and the effective temperature obtained from the EEDF, measured by a Langmuir probe. In this work we derive a modified version of the Saha equation by assuming a Druyvesteyn energy distribution function for the electrons. We apply the modified Saha equation to a low pressure argon plasma produced in an inductive RF discharge to obtain the electron temperature. We show that the modified version introduces substantial corrections in the measured values of the electron temperature given by OES, which approach those given by the second derivative method of Langmuir probe analysis.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Optical emission spectroscopy (OES) is a powerful *in situ* tool, for non-intrusive plasma diagnostic, and is receiving growing interest in a wide range of applications where plasma sources are employed. There are two important configurations often used for material processing: (a) Laser ablation, when a high-energy laser pulse is strongly focused onto a target to generate a plasma plume that expands freely in vacuum, or into an inert or reactive gas atmosphere, and subsequently reaches a substrate to form a thin film on it. (b) Plasma sputtering, when a dc or ac discharge is used to generate a plasma from a buffer gas (usually an inert gas) that erodes a target. Hence, the species (atoms, molecules, electrons, ions, clusters, etc.) removed in this interaction are eventually deposited onto a substrate to form the film. In both cases the plasma parameters, such as electron temperature, density, electron and ion energy distribution functions (EEDF and IEDF) and metastable population, can play an important role in the control of the deposition conditions [1].

Interpretation of the optical spectrum from the plasma is often based on equilibrium relations. Basically one needs to infer from the emitted spectrum the densities in the various quantum states of atoms and ions and the electron density. If

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some of the radiation is reabsorbed in the plasma or reflected at its boundary, one must solve radiative transfer equations. In the absence of the self-absorption term in the rate equations, the analysis becomes considerably simplified. This is often the case for the optical emission from laboratory plasmas with low to moderate temperatures, except in the case of resonant transitions. In the case of local thermodynamic equilibrium (LTE) [2], the density of specific quantum states can be determined for a system that has the same total mass density, temperature and chemical composition as the actual system. The relevant temperature corresponds to the distribution function of the species dominating the reaction rates. In that case electrons and ions will both have nearly Maxwellian velocity distributions, even if the two kinetic temperatures may be quite different. From the Saha equation, a relation between the total densities of two subsequent ionization stages can be derived. When the equilibrium condition is not fulfilled, the ion and electron velocity distributions are no longer Maxwellian and a modified version of the Saha equation has to be implemented.

The Saha equations can be solved either by a kinetics theory method [3] or from the thermodynamics for equilibrium systems [4, 5]. When equilibrium thermodynamics is used, two different results are obtained for a two temperature plasma. Morro and Romero [4] proposed a modified Saha equation that depends on the electron and the ion temperatures [6, 7]. Later, Van de Sanden *et al* [5] and Chen and Han [8] showed that the modified Saha equation obtained by Morro is not valid for a two temperature plasma, because its derivation is based on equilibrium assumptions (from the minimization of the Gibbs free energy). On the other hand, the equation obtained by Chen for a two temperature plasma has the same mathematical form of the standard Saha equation, only changing T by T_e.

In this work we derive a modified version of the Saha equation from the kinetic solution by assuming a Druyvesteyn velocity distribution function for the electron and the ion species. We apply the modified Saha equation to a low pressure argon plasma produced by an inductive RF discharge to obtain electron temperatures, which are consistent with the result obtained with an electrostatic Langmuir probe. The obtained temperatures are almost an order of magnitude higher than those predicted by the standard Saha equation.

2. Experimental apparatus

The experimental apparatus consists basically of an inductively coupled plasma produced by a three loop antenna placed inside a cylindrical stainless steel (316L) chamber. The RF power supply is based on a push–pull oscillator designed with a variable output power ranging from 10 to 500 W, operating at 13.56 MHz. The chamber is pumped to a base pressure of 10^{-7} mbar; during operation it is filled with argon and the working pressure is kept constant (5×10^{-2}). The chamber has a quartz window for optical emission spectroscopy (OES) and the target holder and the Langmuir probe are placed on two separated retractile manipulators.

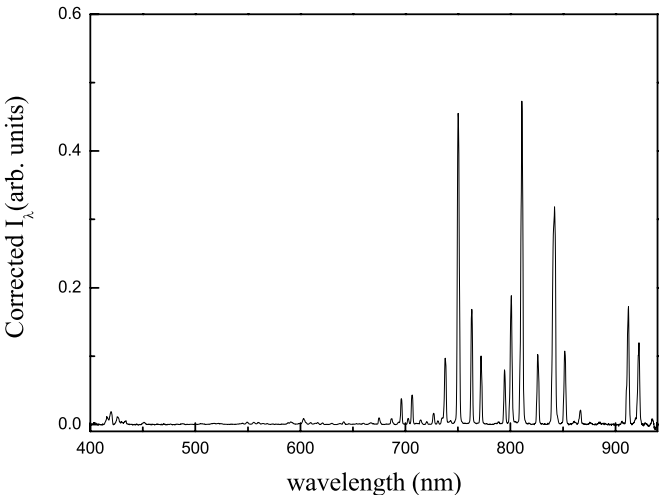

Figure 1. Calibrated optical emission spectrum measured for a pressure of 5×10^{-2} mbar and 120 W of RF power.

2.1. Optical emission spectroscopy

For OES measurements, an optical system consisting of a set of collimating lenses is used to focus the plasma light onto the entrance of an optical fibre coupled to a Czerny–Turner spectrometer (HR4000 model from Ocean Optics). The spectrometer is equipped with a holographic grating (Composite™) of 300 lines mm^{-1} with a linear CCD array of 3468 pixels, yielding a resolution of 0.5 nm in the spectral range from 200 to 1100 nm. Emission intensities are corrected according to the wavelength dependence of the spectral sensitivity of the CCD and of the grating transmission efficiency. Our measured spectra were further corrected with a NIST-traceable calibrated tungsten halogen light source (300–1050 nm) from Ocean Optics (model LS-1-CAL). A typical calibrated optical emission spectrum is shown in figure 1 for a pressure of 5×10^{-2} mbar and 120 W of RF power.

2.2. Electrostatic probe measurement

A single Langmuir probe was constructed with a tungsten tip of 0.5 mm diameter brazed to a glass tube head. A low band pass filter is placed inside the tube and close to the probe tip to reduce RF distortion. The glass head, glued to a stainless steel tube, is inserted along the axis of the vacuum chamber and can be rotated and displaced to allow a radial sweep. In the measurements, the probe was biased from −70 to 70 V and the voltage applied together with the current output were simultaneously measured by an ADC (USB6008, National Instruments). In figure 2 we have a typical I–V curve for a pressure of 5×10^{-2} mbar and 120 W of RF power.

The EEDF $F_e(E)$ was obtained following the standard second derivative analysis of the I–V curve [9],

$$I_e = \frac{1}{4}\left(\frac{2e^3}{m_e}\right)^{1/2} A \int_V^\infty E^{1/2} F_e(E) \left(1 - \frac{V}{E}\right) \mathrm{d}E, \quad (1)$$

where A is the area of the collecting probe surface, m_e and e are the electron mass and charge, respectively, $V = \phi_p - V_b$ is the

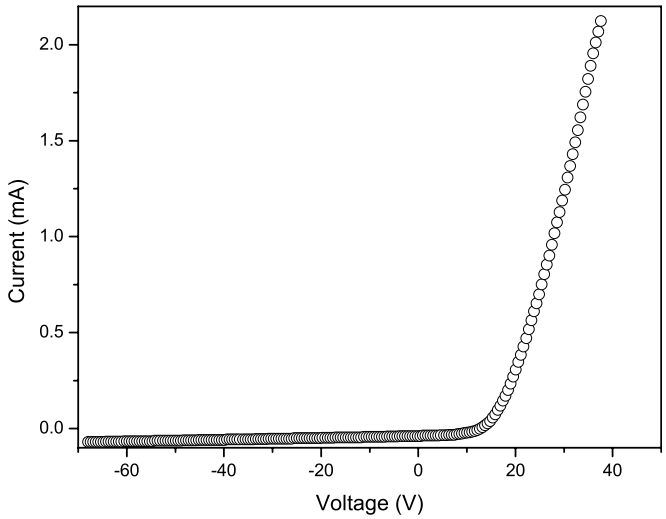

Figure 2. Measured I–V curve for a pressure of 5×10^{-2} mbar and 120 W of RF power.

difference between the plasma potential ϕ_p and the potential applied to the probe V_b, $E = \frac{1}{2}mv^2/e$ is the kinetic energy of the particle, given in electronvolts, and $F_e(E)$ is the EEDF.

Therefore differentiation of equation (1) twice with respect to V yields

$$F_e(V) = \frac{2m}{e^2 A}\left(\frac{2eV}{m}\right)^{\frac{1}{2}} \frac{d^2 I_e}{dV^2}. \tag{2}$$

Once the EEDF is obtained, the number density n_e can be promptly calculated:

$$n_e = \int_0^\infty F_e(V)\, dV \tag{3}$$

and also the effective temperature, given in electronvolts, by

$$T_{eff} = \frac{2}{3n_e}\int_0^\infty V F_e(V)\, dV. \tag{4}$$

3. Theory

3.1. Druyvesteyn distribution

For plasmas that are not in local thermodynamic equilibrium (non-LTE), a Maxwellian distribution function cannot be assumed for the energy of electron. Instead, it is necessary to calculate it from the Boltzmann equation,

$$\frac{\partial}{\partial t} f_e(\vec{r}, \vec{v}, t) + \vec{v}\cdot\vec{\nabla}_r f_e(\vec{r}, \vec{v}, t) + \vec{a}\cdot\vec{\nabla}_v f_e(\vec{r}, \vec{v}, t) = \left[\frac{d}{dt} f_e(\vec{r}, \vec{v}, t)\right]_{col}, \tag{5}$$

where $\vec{a}$ is the acceleration vector of electrons due to external forces, $f_e(\vec{r}, \vec{v}, t)$ is the electron velocity distribution function and $[\frac{d}{dt} f_e(\vec{r}, \vec{v}, t)]_{col}$ is the collision term.

The solution of equation (5) is not easy to obtain because the collisional effect, in the collisional integral on the right-hand side, has to be determined self-consistently for all the particle species. This leads to a set of coupled non-linear integro-differential equations in seven dimensions $(x, y, z, v_x, v_y, v_z, t)$, whose solution is intractable. However, in order to obtain results using this equation, several approximations can be used involving linearization and approximation of zero order. Druyvesteyn [9] solved the Boltzmann equation for a plasma permeated by an electric field, and got the following energy distribution function, assuming isotropy, $f_e(\vec{r}, \vec{v}) \equiv f_e(v)$:

$$F_D(E) = 1.04 n W_{av}^{-\frac{3}{2}} E^{\frac{1}{2}} \exp\left[-\frac{0.55E^2}{W_{av}^2}\right], \tag{6}$$

where n is the number density of particles, E is the energy, W_{av} is the average energy and $F_D(E)\, dE = 4\pi v^2 f_D(v)\, dv$.

3.2. Modified Saha equation

In non-LTE plasmas, the number of particles in a state n, with energy between E_n and $E_n + dE$, is given by

$$dN_n = N\frac{f(E_n)\, dE}{\int f(E)\, dE}. \tag{7}$$

Using a similar relation for particles in a state m, with energy between E_m and $E_m + dE$, and integrating the ratio of these two equations one gets

$$\frac{N_m}{N_n} = \frac{f(E_m)}{f(E_n)}. \tag{8}$$

From the Maxwell–Boltzmann distribution for a system in thermodynamic equilibrium, one obtains

$$\frac{N_m}{N_n} = \frac{g_m}{g_n}\exp\left[-\frac{E_m - E_n}{k_B T}\right], \tag{9}$$

where g_m and g_n are the statistical weights (degeneracy) of states m and n, respectively. Nevertheless, there are cases where the electron energy cannot be correctly described by a Maxwellian distribution, and for conditions as such the plasma is better characterized by a Druyvesteyn function [10]:

$$f_D(E) = C\exp\left[-\frac{E^2}{E_{av}^2}\right]. \tag{10}$$

Therefore a generalized Boltzmann law allows to write

$$\frac{N_m}{N_n} = \frac{g_m}{g_n}\exp\left[-\frac{\left(E_m^2 - E_n^2\right)}{E_{av}^2}\right]. \tag{11}$$

Using this equation for the particular case involving the ground state ($E_0 = 0$) and the first ionization potential χ_I,

$$\frac{dN_0^+}{N_0} = \frac{dg}{g_0}\exp\left[-\frac{\left(\chi_I + \frac{1}{2}m_e v^2\right)^2}{E_{av}^2}\right], \tag{12}$$

where dN_0^+ is the differential number of ions in the ground state with free electrons in the velocity interval between v and $v + dv$, $dg = g_0^+ \cdot dg_e$ being $dg_e = 2\frac{d^3x\, d^3p}{h^3}$ the degeneracy of

free electrons with $\mathrm{d}^3x = 1/n_\mathrm{e}$ and h is the Planck constant. The degeneracy of ions is given by g_0^+.

Considering that for a collisional plasma the relation between the characteristic times for electron temperature *isotropization* and energy relaxation is given approximately by $2\sqrt{2}/3(T_\perp/T_\parallel)$ [11] and provided that the plasma is not magnetized, we can safely assume an isotropic velocity distribution. In this case,

$$\mathrm{d}^3p = 4\pi m_\mathrm{e}^3 v^2\,\mathrm{d}v, \tag{13}$$

and after substitution in equation (12) we find an equation that depends on the velocity of free electrons, given by

$$\frac{\mathrm{d}N_0^+}{N_0} = \frac{8\pi m_\mathrm{e}^3}{n_\mathrm{e}h^3}\frac{g_0^+}{g_0}v^2\exp\left[-\frac{\left(\chi_I + \frac{1}{2}m_\mathrm{e}v^2\right)^2}{E_\mathrm{av}^2}\right]\mathrm{d}v. \tag{14}$$

Integrating this equation we obtain

$$\frac{N_0^+}{N_0} = \frac{8\pi m_\mathrm{e}^3}{n_\mathrm{e}h^3}\frac{g_0^+}{g_0}I, \tag{15}$$

where

$$I = \int_0^\infty v^2\exp\left[-\frac{\left(\chi_I + \frac{1}{2}m_\mathrm{e}v^2\right)^2}{E_\mathrm{av}^2}\right]\mathrm{d}v. \tag{16}$$

This integral is solved defining $t = v^2$, hence,

$$I = \frac{1}{2}\exp\left[-\frac{\chi_I^2}{E_\mathrm{av}^2}\right]\int_0^\infty \sqrt{t}\exp[-\alpha t - \beta t^2]\,\mathrm{d}t \tag{17}$$

with

$$\alpha = \frac{m_\mathrm{e}\chi_I}{E_\mathrm{av}^2} \tag{18}$$

and

$$\beta = \frac{m_\mathrm{e}^2}{4E_\mathrm{av}^2}. \tag{19}$$

Following the discussion presented in [12], we have

$$\int_0^\infty x^{\nu-1}\exp[-\gamma x - \beta x^2]\,\mathrm{d}x = (2\beta)^{-\frac{\nu}{2}}\Gamma(\nu) \times \exp\left[\frac{\gamma^2}{8\beta}\right]D_{-\nu}\left(\frac{\gamma}{\sqrt{2\beta}}\right), \tag{20}$$

where $D_{-\nu}(\frac{\gamma}{\sqrt{2\beta}})$ is the parabolic cylinder function D that can be related to a parabolic cylinder function U using the relation

$$D_{-a-\frac{1}{2}}(x) = U_a(x). \tag{21}$$

Taking $\nu = \frac{3}{2}$, $\beta = \frac{m_\mathrm{e}^2}{4E_\mathrm{av}^2}$, $a = 1$ and $\gamma = \alpha = \frac{m_\mathrm{e}\chi_I}{E_\mathrm{av}^2}$, we can rewrite the integral as

$$I = \frac{\sqrt{\pi}}{4}\left[\frac{2E_\mathrm{av}^2}{m_\mathrm{e}^2}\right]^{\frac{3}{4}}\exp\left[-\frac{\chi_I^2}{2E_\mathrm{av}^2}\right]U_1\left(\sqrt{2}\frac{\chi_I}{E_\mathrm{av}}\right). \tag{22}$$

Therefore equation (15) becomes

$$\frac{N_0^+}{N_0} = \frac{2\pi^{\frac{3}{2}}m_\mathrm{e}^3}{n_\mathrm{e}h^3}\frac{g_0^+}{g_0}\left[\frac{2E_\mathrm{av}^2}{m_\mathrm{e}^2}\right]^{\frac{3}{4}}\exp\left[-\frac{\chi_I^2}{2E_\mathrm{av}^2}\right]U_1\left(\sqrt{2}\frac{\chi_I}{E_\mathrm{av}}\right). \tag{23}$$

Using equation (11), the relation for the ratio between neutral species in the ground state ($E_0 = 0$) and a given excited state (E_m) gives

$$\frac{N_0}{N_m} = \frac{g_0}{g_m}\exp\left[\frac{E_m^2}{E_\mathrm{av}^2}\right]. \tag{24}$$

Now using equation (11) for two excited states E_k and E_l of the ionic specie, we have

$$\frac{N_k^+}{N_l^+} = \frac{g_k^+}{g_l^+}\exp\left[-\frac{(E_k+\chi_I)^2 - (E_l+\chi_I)^2}{E_\mathrm{av}^2}\right] \tag{25}$$

taking E_l as the ion ground state, the equation becomes

$$\frac{N_k^+}{N_0^+} = \frac{g_k^+}{g_0^+}\exp\left[-\frac{\left(E_k^2 + 2E_k\chi_I\right)}{E_\mathrm{av}^2}\right]. \tag{26}$$

Multiplying these two equations, we have

$$\frac{N_k^+}{N_m} = \frac{N_0^+}{N_0}\frac{g_k^+}{g_m}\frac{g_0}{g_0^+}\exp\left[-\frac{\left(E_k^2 - E_m^2 + 2E_k\chi_I\right)}{E_\mathrm{av}^2}\right] \tag{27}$$

and finally, after substituting equation (23), the expression for the modified Saha equation becomes

$$\frac{N_k^+}{N_m} = \frac{2\pi^{\frac{3}{2}}m_\mathrm{e}^3}{n_\mathrm{e}h^3}\frac{g_k^+}{g_m}\left[\frac{2E_\mathrm{av}^2}{m_\mathrm{e}^2}\right]^{\frac{3}{4}}U_1\left(\sqrt{2}\frac{\chi_I}{E_\mathrm{av}}\right) \times \exp\left[-\frac{\left(E_k^2 - E_m^2 + 2E_k\chi_I + \frac{1}{2}\chi_I^2\right)}{E_\mathrm{av}^2}\right] \tag{28}$$

which can be compared with the standard Saha equation given below:

$$\frac{N_k^+}{N_m} = 2\frac{g_k^+}{g_m}\frac{1}{n_\mathrm{e}}\left[\frac{m_\mathrm{e}k_\mathrm{B}T}{2\pi\hbar^2}\right]^{\frac{3}{2}}\exp\left[-\frac{(E_k - E_m + \chi_I)}{k_\mathrm{B}T}\right], \tag{29}$$

where E_k and E_m are the energy levels measured in relation to the ground state of the ion and the neutral, respectively.

3.3. Temperature calculation for non-LTE plasma

In plasma spectroscopy theory [2], there is a relation between the number density N_i and the intensity of the transition line I_{ij}

$$I_{ij} = \frac{\hbar\omega_{ij}^3 r_0}{2\pi c}f_{ji}\int_a^b N_i(x)\,\mathrm{d}x, \tag{30}$$

where r_0 is the classical electron radius, c is the speed of light, f_{ji} is the oscillator strength and ω_{ij} is the frequency for a transition from the level i to j.

Assuming that the ionized species makes a transition from the k to the l state and that the neutral species makes a transition from the m to the n state, the relation between the emission

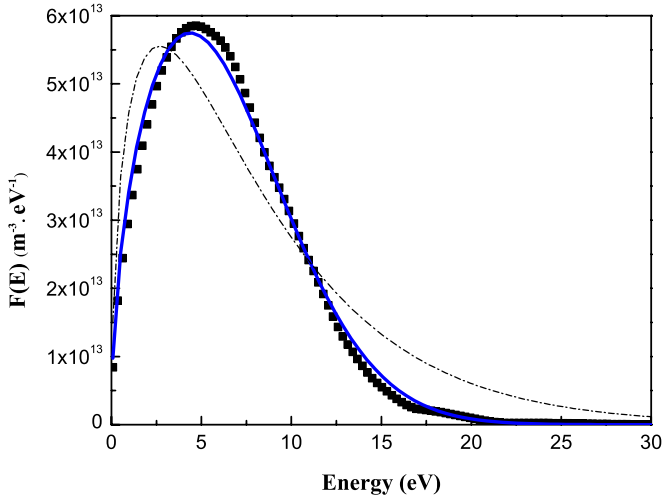

Figure 3. Calculated EEDF from the second derivative of the I–V curve of figure 2 (full squares). From a non-linear fitting procedure (Levenberg–Marquardt method) the best Maxwellian (broken line) and Druyvesteyn (full line) curves that fit to the second derivative data.

lines is given by

$$\frac{I_{kl}}{I_{mn}} = \frac{\lambda_{mn}^3 f_{lk}}{\lambda_{kl}^3 f_{nm}} \frac{N_k^+}{N_m}. \tag{31}$$

From the Druyvesteyn distribution function

$$E_{\mathrm{av}}^2 = \frac{W_{\mathrm{av}}^2}{0.55} \tag{32}$$

and

$$W_{\mathrm{av}} = \frac{3}{2} k_{\mathrm{B}} T. \tag{33}$$

Substituting these three equations in equation (28), we finally have the relation between the electron temperature and the ratio of the transition lines emitted from the plasma:

$$\frac{I_{kl}}{I_{mn}} = \frac{\lambda_{mn}^3 f_{lk} g_k^+}{\lambda_{kl}^3 f_{nm} g_m} \frac{2\pi^{\frac{3}{2}} m_{\mathrm{e}}^3}{n_{\mathrm{e}} h^3} \left[\frac{2.86 k_{\mathrm{B}} T}{m_{\mathrm{e}}}\right]^{\frac{3}{2}} U_1\left(0.699 \frac{\chi_I}{k_{\mathrm{B}} T}\right) \times \exp\left[\frac{E_k^2 - E_m^2 + 2E_k\chi_I + \frac{1}{2}\chi_I^2}{4.09(k_{\mathrm{B}} T)^2}\right]. \tag{34}$$

4. Results

Following the second derivative analysis of the I–V curve shown in figure 2, the EEDF can be calculated; the result is shown in figure 3. Also shown in figure 3 are the best fitted curve of a Maxwellian (broken line) and a Druyvesteyn (full line) function to the EEDF data. It is clear that, for the experimental conditions of this work (working pressure of 5×10^{-2} mbar and RF power of 120 W), the EEDF is better described by a Druyvesteyn function, indicating that the plasma is outside the thermodynamical equilibrium.

Concerning the optical emission spectroscopy (OES) analysis, it is important to point out that the optical emission spectrum from a low pressure argon RF discharge is dominated by Ar I optical transitions [13], in particular from 4p $\rightarrow$ 4s. Some of these transitions were avoided due to the large number of atoms in the $3p^5 4s$ metastable and resonant levels [14]. On the other hand experiments point to a sub-dominant role of metastable excitation of the np^1, np^5 ($J = 0$) levels, and often their emission can be assumed to be completely free of a metastable atom excitation contribution [15]. Therefore, we shall restrict the solution of equation (34) to the set of levels (750.4, 425.9 and 451.1 nm), in particular to the Ar I $3s^2 3p^5 (^2P^o_{1/2}) 4s \rightarrow 3s^2 3p^5 (^2P^o_{1/2}) 4p$ and Ar II $3s^2 3p^4 (^3P) 3d \rightarrow 3s^2 3p^4 (^3P) 4p$ transitions, which correspond to the wavelength of 750.38 nm and 686.12 nm, respectively.

Provided that the line intensities from the selected optical transitions of two subsequent ionization stages are given, equation (34) can be solved graphically [2] by plotting the right-side term as a function of T_{e}. However, the electron density has to be provided by either Langmuir probe measurement (which is the case in this work) or by Stark broadening of a particular optical transition [16]. Hence for each (I_{kl}/I_{mn}) ratio a corresponding T_{e} will satisfy the solution of equation (34) univocally.

The graphical solutions of the standard Saha equation and the modified Saha equation are shown in figures 4(*a*) and (*b*), respectively. The optical constants were obtained from the NIST database [17]. One can readily see that, for the same (I_{kl}/I_{mn}) set of values, the solutions of the two Saha equations lie in two quite different temperature ranges, differing roughly by an order of magnitude.

5. Discussion

The results from Langmuir probe analysis (equation (4)), modified (equation (34)), and the standard Saha equations (equation (29)) are presented in table 1, for a RF power of 120 W and a working pressure of 5×10^{-2} mbar. For the line intensities studied in this work, the ratio of $I_{kl}/I_{mn} = 0.02$ was obtained. From figure 4 the values obtained for the standard and the modified Saha equation were, respectively, 0.72 eV and 5.4 eV.

Although the modified Saha equation, based on a Druyvesteyn EEDF, corrects the electron temperature to approach that given by the second derivative probe method, there still exists a discrepancy of approximately 18% between the two methods. Discrepancies between probe analysis and line ratio methods, such as the standard Saha equation or the Boltzmann plot, are also observed for plasmas where the EEDF is well represented by a Maxwellian distribution [14].

In fact, recently, Kang *et al* [14] developed a method that corrects the temperature obtained from the ratio of two spectral lines from the same ionization stage to the effective electron temperature measured via a single Langmuir probe. In their model a correction factor is assumed, which depends on the gas pressure and is derived from the balance between the measured light intensity and the degree of excitation by collision via the EEDF and electron density obtained from the probe measurement.

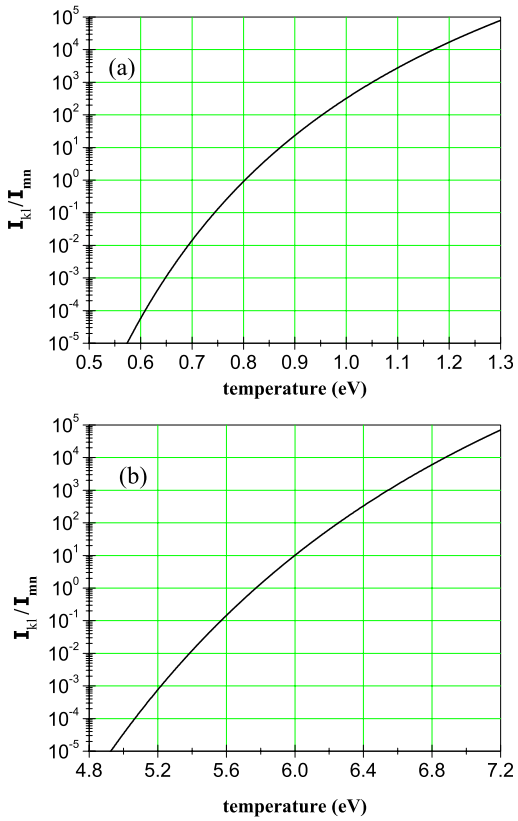

Figure 4. Graphical solution of the standard Saha equation of [2] (*a*) and the modified Saha equation presented in equation (34) (*b*).

Table 1. Electron density calculated from equation (3) and electron temperature from equations (4), (34) and (29).

n_e (m^{-3}) (equation (3))	T_e (eV) (equation (4))	T_e (eV) (equation (34))	T_e (eV) (equation (29))
3.2×10^{15}	6.6	5.4	0.72

Although the method of Kang *et al* [14] is applicable only for lines from the same ionization state, while Saha's equation is valid for lines from subsequent ionization states, it is interesting, for the sake of completeness, to compare the results from the two models. Applying equation (11) of Kang *et al* to the Ar I lines 750.4 and 425.9 nm, and taking the data on the relevant cross sections from [15], we obtain $T_e = 3.5$ eV for a Maxwellian distribution function and $T_e = 6.1$ eV for a Druyvesteyn distribution function, for the conditions given in table 1. Therefore the results are all in good agreement.

6. Conclusions

We have presented a model to modify the Saha equation for the case where the EEDF is better described by a Druyvesteyn than a Maxwellian function. The electron temperature obtained from the modified Saha equation, using the spectral line ratio method, approaches reasonably well the effective temperature measured via Langmuir probe. Our result also is in qualitative agreement with the model proposed by Kang *et al* [14]. However, the method described in this paper is considerably simpler to apply in order to obtain the electron temperature in RF produced plasmas at least in the pressure range around 10^{-2} mbar.

Acknowledgments

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