A novel diagnostic technique for H−(D−) densities in negative hydrogen ion sources

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Abstract. A new diagnostic method for the determination of negative hydrogen ion densities H− (D−), namely the Hα/Hβ line ratio method, is presented and applied to high power (P ≈ 100 kW) negative ion sources operating at low pressure (p < 1 Pa). The basis of the method is the mutual neutralization process which enhances Balmer line radiation selectively. Detailed parameter studies carried out with a collisional radiative model show that the Balmer line ratio Hα/Hβ is very well suited to obtain negative ion densities in low temperature hydrogen plasmas with typical plasma parameters of T_e ≈ 1–5 eV and n_e ≈ 10^{16–5} \times 10^{18} m^{-3}. The Hα/Hβ line ratio method has the great advantage that it can be accomplished easily with the non-invasive and in situ plasma diagnostic method of optical emission spectroscopy. The method is applied to the RF ion sources of IPP Garching, currently developed for the neutral beam injection system of ITER. Line-of-sight averaged negative hydrogen densities in the range of 10^{16–10^{17}} m^{-3} are measured close to the extraction system in hydrogen and deuterium discharge. Absolute values depend on pressure, power and caesium conditioning of the source. Negative ion densities measured with this novel technique in front of the grid correlate very well with the extracted negative ion current densities.

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1. Introduction

Negative hydrogen ion sources have a variety of applications as a primary particle source in high energy physics, in spallation sources, for ion beam deposition studies or for tandem accelerators, and are one of the key elements for the formation of intense beams of energetic neutrals (deuterium) for fusion plasma heating [1]. In general, the negative ions are produced in low-pressure hydrogen plasmas, however, the demands on the source differ substantially. For applications as primary particle source low extracted negative ion currents are sufficient (ranging typically from nA to mA) which is achieved by using small sources with single aperture extraction. The side effect of co-extracted electrons is no problem for the loading of the extraction grids. In fusion research, the neutral beam injection system requires large negative ion beams with high ion currents (tens of amperes) at high energies (hundreds of kV up to 1 MeV), which means that large sources with a multi aperture extraction system are to be used. As a consequence minimizing the amount of co-extracted electrons at simultaneous maximization of negative ions is a crucial point. While negative hydrogen ion sources are routinely used as primary particle source, sources for fusion have to be developed and optimized in all relevant performance parameters for long term, low maintenance operation. This challenging program has to be supported by diagnostic tools with emphasis on diagnostics of negative hydrogen ions. Therefore, focus is laid on negative ion sources for fusion; requirements and their status will be discussed in the following in more detail.

Although development of negative ion sources began in the late 1970s, a strong development program for high power negative ion sources for neutral beam applications in fusion experiments started just a few years ago. It was initiated by the basic necessity of a high energy neutral beam system for heating and current drive for the next generation of fusion devices, namely ITER [2]. At the required energy of 1 MeV (D) the neutralization efficiency for negative ions is still about 60%, whereas for positive ions the neutralization efficiency tends to zero at energies above 200 kV [3]. Further ITER requirements are: the source should deliver a current of 40 A D⁻ at a source pressure of 0.3 Pa at maximum for 3600 s [4]. The pressure limit is determined by the
stripping losses of negative ions in the extraction system which should be kept below 30%. At negative ion current densities of $j_{D^-} = 20 \text{ mA cm}^{-2}$ maximum at 0.3 Pa the effective extraction area will be 2000 cm² (1280 holes), resulting in a large area source of $1.5 \times 0.6 \text{ m}^2$ cross section. The ratio of co-extracted electrons to negative ions is required to be $j_e/j_{D^-} \leq 1$.

Large negative ion sources are in operation at JT60-U [5, 6] and LHD [7, 8], representing roughly a 2/3 and 1/3 ITER size, respectively. These sources are filament driven arc sources, based on the so-called KAMABOKO type [9]. Research and optimization of such a source is also carried out at CEA [10, 11]. The present ITER reference design is based on the filamented arc sources, which however suffer from reduced availability and increased maintenance effort due to the limited lifetime of the filaments. Therefore, as an alternative, RF driven sources are developed within the framework of an European Fusion Development Agreement (EFDA) contract at the Max Planck Institute for Plasma Physics (IPP) [12, 13].

The physics of production, acceleration and neutralization of large negative ion beams for neutral beam injection systems is reviewed in [14]. Negative ion sources operating in low pressure (ITER relevant) rely nowadays on the so-called surface effect, where impinging hydrogen particles (H and/or H⁺) pick up electrons from a surface with a low work function. The latter is achieved by evaporation of caesium which forms a thin layer on the surfaces, preferably close to the extraction system. In comparison to negative ions sources based on volume processes, i.e. the dissociative attachment of the vibrationally excited hydrogen molecules, from caesium seeded discharges an order of magnitude higher current densities are extracted with a simultaneous reduction of co-extracted electrons (factor 10) at the required low pressure of 0.3 Pa [13].

Standard diagnostics for ion sources are electrical measurements of extracted ion and electron currents, and calorimetrically measured ion current densities. However, these parameters depend beside the negative ion density and electron density in the plasma on a variety of external parameters such as the geometry of the grid system, the beam optics and the extraction voltage. On the other hand, optimization of the formation of negative ions—with a binding energy of 0.75 eV only—in caesiated hydrogen sources with a complex plasma chemistry makes plasma diagnostics essential. Electron density and temperature can be obtained with conventional Langmuir probe measurements, whereas simple in situ diagnostic methods for negative ion densities are still highly desirable.

In the last years, two diagnostic methods for negative hydrogen ions have been developed and are used as standard for small laboratory plasmas. One of them is the laser detachment method: an intense laser beam detaches the additional electron, the enhancement in electron density is detected with a Langmuir probe system (for details see [15]). This method allows for spatially and time resolved measurements [15, 16]. Typical ion densities obtained in small laboratory plasmas (typical size: 20 cm in diameter) are in the range of $10^{15} - 10^{16} \text{ m}^{-3}$. Correct measurement of the $\text{H}^-$ density depends mainly on a proper size of the laser volume. The latter is given by the laser energy which must be high enough to detach all the electrons. However, the analysis is more complex when magnetic fields are present. Additional problems may arise in high power, low-pressure sources with extraction (as in the case of neutral beam sources) due to disturbances from the high voltage on the Langmuir probe system. For RF sources, Langmuir probe measurements are more complicated because of the RF noise which have to be compensated for.

A second method is the non-invasive cavity ring down spectroscopy [17] being applied so far to laboratory plasmas. This very sensitive absorption technique (down to absorption coefficients of $10^{-8} \text{ cm}^{-1}$) measures the laser light absorbed in the detachment process. Since the decay time
of the cavity is detected with and without plasma, absolute line-of-sight (LOS) averaged values are directly obtained; complicated calibration procedure can be avoided. However, the optical set-up is very difficult to adjust and the mirrors with a very high reflectivity can be contaminated. The huge advantage of this method for large high power, low pressure (RF) sources with extraction is that the method is based on pure optical measurements.

As an alternative, an easy and simple non-invasive technique for in situ measurements of negative ion densities in high voltage and RF environment was developed at IPP Garching based on optical emission spectroscopy (OES). In the following, it will be shown that the Balmer line ratio $H_\alpha/H_\beta$ is sensitive on the negative ion density. Section 2 describes the diagnostic method, the underlying collisional radiative (CR) model, and discusses the parameter range for application. A formula for a straightforward analysis is presented in subsection 2.4. Section 3 describes the experimental set-up of the IPP RF sources and presents first results of the applications of this method. The correlation of measured negative ion densities in front of the grid with the measured extracted current density of negative ions is discussed in subsection 3.4.

2. Diagnostic method

OES is widely used for plasma diagnostics in many applications. Principles of OES applied to low pressure, low temperature plasmas with molecules are, for example, described in [18, 19]. Applications to low temperature plasmas containing hydrogen and deuterium are given in [20]. The non-invasive technique measures the plasma emission (line radiation) in the visible spectral range. Thus, the experimental set-up is very simple, especially when using fibre optics directly at the experiment. The results are LOS averaged but with a proper arrangement of several LOS some spatial resolution can be obtained. The resolution in time is determined by the data acquisition system. Although data are very easy to obtain the analysis can be quite complex, in particular in molecular plasmas with plasma chemistry, and has to be supported by CR modelling. The diagnostic technique with its various analysis methods was already successfully applied to negative hydrogen ion sources, namely to the RF sources of the IPP, where it is now a standard diagnostic tool, and to the KAMABOKO type filamented arc source at CEA [21]–[23].

The following plasma parameters with spatial and temporal resolution were measured: electron temperature, electron density, gas temperature, atomic to molecular hydrogen density ratio, and other plasma components like impurities, caesium, and tungsten in the case of arc sources, being evaporated from the filaments.

For the measurement of negative hydrogen ion densities with OES a new analysis technique has been developed based on the interpretation of Balmer line emission. Hence, in the following, focus is laid on the population mechanisms to the excited states of atomic hydrogen for the typical plasma parameters of negative ion sources.

2.1. Population of excited states of atomic hydrogen

Line emission $\varepsilon_{pk}$ from atomic hydrogen is correlated with the population density in the electronically excited state (quantum number $p$) of atomic hydrogen $n_H(p)$ by:

$$\varepsilon_{pk} = n_H(p)A_{pk}$$

(1)
with the transition probability $A_{pk}$ from level $p$ to level $k$. The dependence of the population density in the excited state on plasma parameters, such as electron density $n_e$ and electron temperature $T_e$, can be calculated with so-called CR models. The combination of measurement and calculation gives access to the plasma parameters and particle densities.

CR models balance collisional and radiative processes for each electronic state and have to be used for non-equilibrium plasmas, such as low pressure, low temperature hydrogen plasmas [19]. In order to describe ionizing and recombinating plasmas, the electronically excited state of a particle is usually coupled to the ground state of the particle and the next ionization stage. This means, in case of excited atomic hydrogen, $n_{H}(p)$ is coupled to H and H$^+$ particles, with densities $n_H$ and $n_{H^+}$, respectively, via:

$$n_H(p) = n_H n_e R_H(p) + n_{H^+} n_e R_{H^+}(p).$$

The population coefficients $R_H(p)$ and $R_{H^+}(p)$ are calculated by the CR model. Several CR models exist for atomic hydrogen, one of them is the atomic data and analysis structure (ADAS) code [24] which has implemented the latest data for the cross sections and is used in the fusion and astrophysics community as a standard. The model described in [25, 26] is based on the same set of cross sections but has the great advantage of being directly connected with a CR model for molecular hydrogen. Hence, dissociative excitation from molecular hydrogen, $H_2$, and dissociative recombination of molecular ions, $H_2^+$, can be taken into account with this model. The model includes all collisional and radiative excitation and de-excitation processes with the exception of stimulated emission which is of no relevance in low density discharges. While the electronic structure of the states is resolved, the splitting due to different angular momentum is not included explicitly. However, the input data of the model is averaged and summed up respectively in a way that the angular momentum splitting is considered implicitly. This is of particular importance for taking the metastable character of the $p = 2$ level into account. Since the two levels 2s and 2p are coupled by strong coupling reactions it can be assumed that they are in equilibrium [27]. Thus an explicit treatment of the $p = 2$ splitting is not needed. Cross sections for dissociative excitation and dissociative recombination are taken from [28, 29] and from [29, 30], respectively.

Dissociative recombination of vibrationally unexcited $H_3^+$ particles leads to energies which are not sufficient to produce an excited atom, whereas highly rotational and vibrational excited $H_3^+$ can contribute to $H(p)$. However, a complete set of branching ratios for excitation of the different electronically excited states is not available. In order to calculate an upper limit for the influence, the total rate coefficient [31] was used together with a $H_3^+$ density being equal to the electron density. Contributions to the $p = 2$ level are of particular importance. They can lead by internal redistributions to enhanced Balmer line emission. As a result, the $p = 3$ level is enhanced by less than 0.1% in comparison to the effective atomic excitation only. This means dissociative recombination of $H_3^+$ is negligible for Balmer line emission in typical low pressure hydrogen plasmas, which is in accordance with results presented in [32]. Cross sections for heavy particle collisions with $H_3^+$ which can contribute to the Balmer line radiation (e.g. collisions of $H_3^+$ with $H_2$) decrease rapidly with decreasing relative energy of the particles [33]. Since in low pressure plasmas the heavy particles have similar energies, contributions of heavy particle collisions with $H_3^+$ to Balmer line emission are negligible small, even for $H_3^+$ densities being equal to electron densities. $H_3^+$ particles are formed by heavy particle collisions ($H_2^+ + H_2 \rightarrow H + H_3^+$); a process which is of less importance at these low pressures. The absolute value of the $H_3^+$ density is of minor interest since it was verified that the contributions of both dissociative recombination.
and heavy particle collisions are of no importance, even if $H_3^+$ is dominant ion species. Thus, in the following, the contribution of $H_3^+$ to the Balmer line emission is neglected.

Besides the excitation channels from $H$, $H^+$, $H_2$ and $H_2^+$ particles a further excitation channel for atomic hydrogen has to be added, namely the mutual neutralization process

$$H^- + H^+ \rightarrow H(p) + H,$$

being of particular importance in negative ion sources. Taking into account these five excitation channels, which are illustrated in figure 1, the following formula is obtained:

$$n_{H}(p) = n_{H}n_{e}R_{H}(p) + n_{H^-}n_{e}R_{H^-}(p) + n_{H_2}n_{e}R_{H_2}(p) + n_{H_2^+}n_{e}R_{H_2^+}(p) + n_{H^-}n_{H^+}R_{H^-}(p).$$

The five population coefficients ($R_s(p)$, $s$ denotes the particle species) can be calculated as a function of plasma parameters with the above-mentioned CR model for $H_2$ and $H$ [25, 26] which was extended by the mutual neutralization process (3). Cross sections for the latter process are taken from [34]. However, molecular ions may contribute also to the mutual neutralization. According to [35], the same mutual neutralization rate coefficient can be used for all positive ion species of hydrogen. Hence, the $H^+$ density in the last term of equation (4) can be replaced by the electron density.

The population coefficient for the mutual neutralization process, $R_{H^-}(p)$, is almost independent of electron temperature but depends on the ion temperature. This basic dependence originates from the convolution of the cross section for mutual neutralization with the energy of the ion species in the centre of mass description. The corresponding rate coefficient decreases with increasing ion temperature by roughly a factor of two for temperatures higher than $\approx 1000$ K, which is close to typical gas temperatures, and lower than 10 eV [30]. In a first approximation, $H^+$ particles and $H$ particles are assumed to have the same temperature since the energy of the hydrogen particle is preserved in the ionization process (electron impact). Atomic hydrogen particles gain the Franck–Condon energy of 2.24 eV in the dissociation process. Heavy particle collisions with hydrogen molecules decrease the energy. Thus, an ion temperature higher than gas temperature and lower than the Franck–Condon energy can be attributed to the $H^+$ particles.
According to [36], the energy of negative hydrogen ions depends on the formation process: volume processes lead to energies below 1 eV, whereas a few electron volts are achieved by the surface mechanism. Surface produced negative ions are decelerated in the plasma volume by consecutive collisions with background particles. As a reasonable assumption, the temperature of the negative ions of below 5 eV can be used for the calculation of the rate coefficient. In this temperature range the variation of the rate coefficient is only 10% so that the precise ion temperature is of limited interest. The dependence of the rate coefficient on the negative ion temperature is actually so weak that for a fixed temperature of 2 eV of the positive ions a variation of the negative ion temperature from 5 to 0.5 eV changes the rate coefficient by 10% at most.

Individual contributions of the different particle species H, H+, H2, H2+ and H− to the population density of atomic hydrogen in the excited state and their sum are shown in figure 2 for reasonable plasma parameters of negative ion sources as a function of the quantum number of the excited state. It is obvious that the different processes contribute quite similarly to the individual quantum numbers above \( p > 5 \), except for the contribution from H+ recombination which populates preferentially higher quantum numbers and is of minor importance for the population in total. Dissociative recombination of H2+ particles prefers to populate quantum numbers \( p > 3 \) whereas dissociative excitation from H2 particles shows a steady decrease with increasing quantum number. Effective excitation from H particles is almost constant for low quantum numbers and decreases with higher quantum numbers. This process is the dominant process for the parameter range chosen for figure 2. However, one exception can be seen clearly: the mutual neutralization process populates selectively the quantum number \( p = 3 \) with an enhancement of one order of magnitude in comparison to the quantum numbers \( p = 2 \) and \( p = 4 \). For the chosen parameters, this enhancement affects clearly the sum of all processes where the population density shows a peak for \( p = 3 \), resulting in increased Hα line emission. This indicates that Balmer line ratios with Hα can be sensitive to negative ion densities. However, before general conclusions can be drawn the parameter dependence must be critically reviewed.

**Figure 2.** Contributions of different particle species to the population of excited states of atomic hydrogen at the specified temperature and particle densities.
Effective rate coefficient (m³ s⁻¹)

\[ \text{Electron density (m}^{-3}\text{)} \]

**Figure 3.** Effective emission rate coefficients for excitation from the individual particle species at \( T_e = 3 \text{ eV} \) for \( H_\alpha \) (upper part) and \( H_\beta \) (lower part) as a function of electron density.

### 2.2. Parameter dependence

To obtain a better understanding of the contribution of the five excitation channels to the Balmer line radiation the parameter dependence of rate coefficients and line ratios will be investigated first. For this purpose, the contributions from the five excitation channels are considered separately. This makes it, in a next step, possible to evaluate the specific importance of the channels and thus simplify the analysis for specific plasma sources by neglecting channels which are of minor relevance in these sources. Of course, this restricts the applicability of the simplified method to a certain plasma parameter range.

The combination of equations (1) and (4) together with the usage of an effective emission rate coefficient for each of the five particle species (index \( s \)) \( X_{pk}^{\text{eff},s} \):

\[ X_{pk}^{\text{eff},s} = R_s(p)A_{pk} \]  

results in the following equation for line emission; \( H_\alpha \) line emission and thus a transition from level \( p = 3 \) to level \( k = 2 \) is chosen as an example:

\[ H_\alpha \equiv \varepsilon_{32} = n_H n_e \left( X_{H_\alpha}^{\text{eff},H} + \frac{n_{H^+}}{n_H} X_{H_\alpha}^{\text{eff},H^+} + \frac{n_{H_2}}{n_H} X_{H_\alpha}^{\text{eff},H_2} + \frac{n_{H^+}}{n_H} X_{H_\alpha}^{\text{eff},H^+} + \frac{n_{H^-}}{n_H} X_{H_\alpha}^{\text{eff},H^-} \right). \]  

Since particle densities are now related to the atomic hydrogen density the individual contributions to the line emission are better to identify. Figure 3 shows the calculated \( X_{pk}^{\text{eff},s} \).
for $\text{H}_\alpha$ and $\text{H}_\beta$ as a function of electron density for an electron temperature of $T_e = 3$ eV. As can be seen, the dependence on electron density is different for $\text{H}_\alpha$ and $\text{H}_\beta$. The effective emission rate coefficient which describes the contribution of $\text{H}^-$ to $\text{H}_\alpha$ emission, $X_{\text{H}_\alpha}^{\text{eff}, \text{H}^-}$, dominates in the whole electron density range. Regarding the dependence on electron temperature (not shown in figure 3), effective emission rate coefficients for Balmer line excitation from H particles and dissociative excitation of $\text{H}_2$ particles ($X_{\text{H}_\alpha,\beta}^{\text{eff}, \text{H}_2}$ respectively) show the well known steep dependence of electron impact excitation rate coefficients on electron temperature, i.e. a strong increase with electron temperature. Recombination, i.e. excitation processes from $\text{H}^+$ particles, and thus $X_{\text{H}_\alpha,\beta}^{\text{eff}, \text{H}^+}$ decreases with increasing electron temperature. This dependence is less pronounced for the dissociative recombination process (excitation from $\text{H}_2^+$ particles), described by $X_{\text{H}_\alpha,\beta}^{\text{eff}, \text{H}_2^+}$. The effective rate coefficient for mutual neutralization $X_{\text{H}_\alpha,\beta}^{\text{eff}, \text{H}^+}$ is almost independent of electron temperature, since the process itself depends on ion temperature, as discussed before.

In order to eliminate the basic dependence of the line emission on electron density and electron temperature, line ratios can be used. In addition, the selective influence of $\text{H}^-$ on $\text{H}_\alpha$ will be much better pronounced. The line ratios $\text{H}_\alpha/\text{H}_\beta$ and $\text{H}_\beta/\text{H}_\gamma$ are plotted in figure 4 for the same parameter range as in figure 3. As expected, the line ratios show a reduced dependence on electron density with one exception: the excitation via mutual neutralization ($\text{H}^-$) results in a strong dependence on electron density for the line ratio $\text{H}_\alpha/\text{H}_\beta$. Figure 5 shows the dependence on electron temperature which is almost negligible in the temperature range relevant for negative
The dependence of Balmer line emission on plasma parameters offers other diagnostic possibilities for different types of plasmas. In recombining plasmas, where the electron temperature is low ($\lesssim 1$ eV) and the electron density is high ($\gtrsim 10^{19}$ m$^{-3}$) the contribution of H$^+$ to Balmer line emission is by far the most dominant one, in particular for higher quantum numbers as can be deduced from figure 2. This allows a determination of electron temperature and electron density from highly excited Balmer lines, a diagnostic technique which is applied, for example, to the cold plasma edge of fusion experiments [37, 38] and plasma devices working in recombining modes [39]. For an ionizing plasma the line ratio of Balmer lines can be used for diagnostics of electron density [40] in particular when the degree of dissociation is high. This can be seen in figure 4: the line ratios of $H_{\alpha}/H_{\beta}$ and $H_{\beta}/H_{\gamma}$ excited from atomic hydrogen (denoted by H) increase more than a factor of two in the electron density range of $10^{17}$ m$^{-3}$ to $< 10^{19}$ m$^{-3}$. In ionizing low pressure hydrogen plasmas with a low degree of dissociation ($H/H_2$ density ratio $\lesssim 0.1$) contributions from H$_2$ are of relevance as well as contributions from H$^+_2$ ($H/H_2^+$ density ratio $\gtrsim 10^3$) as can be seen in figure 3. For a low degree of ionization, contributions from H$^+_2$ are negligible and the degree of dissociation can be obtained from Balmer line emission [41]: the line ratios ($H_\alpha/H_\beta$ and $H_\beta/H_\gamma$) increase from excitation from H particles to excitation from H$_2$ particles (dissociative excitation) as shown in figure 4. Thus, the variation of line ratios within these two limits is correlated with the density ratio H/H$_2$. However, an additional process,
not yet discussed, may influence the Balmer line emission also, namely the self-absorption of Lyman lines, enhancing population of excited states which enhances Balmer line emission. The enhancement is more pronounced for $H_\alpha$ than for $H_\beta$ etc, and can be in the order of a factor of two, as discussed in more detail in [41].

An advantage of using line ratios for diagnostic purposes is the simplification of the experimental effort. For measurements of line ratios a relative calibration of the spectroscopic set-up, namely the knowledge of the photon sensitivity of the system on wavelength is sufficient. In addition, the complex interpretation of single line analysis can be avoided and line ratios are more suitable for plasma monitoring.

2.3. *Line ratio method for negative ion sources*

The last section dealt with the parameter dependence of the individual contributions to the Balmer line radiation in general. In addition, examples for already established diagnostic techniques for recombining and ionizing plasmas were presented. This section is dedicated to applications of Balmer line radiation for diagnostic techniques in low pressure, high power, hydrogen plasmas developed for negative ion sources. Thus, different excitation channels from the ones that are discussed above will be of importance.

These plasmas are ionizing plasmas with an ionization degree and degree of dissociation of typically $\approx 10^{-2} - 10^{-1}$ and $\geq 0.1$, respectively. As can be seen in figures 2 and 3 contributions of $H^+$, $H_2$ and $H^+_2$ to Balmer line emission can be neglected in these plasmas: recombination is irrelevant at electron temperatures $> 1$ eV and electron densities $< 10^{19}$ m$^{-3}$, dissociative excitation and dissociative recombination are negligible for density ratios $H_2/H$ $> 0.1$ and $H/H < 1000$, respectively. Assuming that $H^+$ is the dominant ion species (density ratio $H^+/H^+_2$ $\geq 10$) this last-mentioned limit is usually achieved. Due to the low pressure ($< 1$ Pa) and thus low absolute atomic hydrogen densities, self-absorption of Lyman lines has not to be taken into account. Hence, the particle species H and $H^-$ determine the line emission of the most intense Balmer lines $H_\alpha$, $H_\beta$ and $H_\gamma$.

For these parameters, Balmer line emission and their line ratio depend on electron density, electron temperature and the density ratio $H^-/H$. Figure 6 shows the line ratios $H_\alpha/H_\beta$ and $H_\beta/H_\gamma$ as a function of the density ratio $H^-/H$ for relevant electron densities and electron temperatures. The line ratio $H_\beta/H_\gamma$ is almost independent on the $H^-/H$ density ratio, but it is rather sensitive on electron density for $n_e > 10^{17}$ m$^{-3}$ (figure 4). A dependence on electron temperature is negligible for low $H^-/H$ density ratios. As expected, the $H_\alpha/H_\beta$ line ratio is very sensitive on the density ratio $H^-/H$ for ratios $H^-/H > 10^{-3}$ and rather insensitive to the electron density for ratios $H^-/H < 10^{-1}$. However, the steep dependence on the density ratio $H^-/H$ vanishes for electron densities higher $10^{19}$ m$^{-3}$ as indicated by the flattened curve for $5 \times 10^{18}$ m$^{-3}$. At low electron temperatures a strong dependence on electron temperature is obtained due to the strong decrease of the effective excitation from atomic hydrogen whereas the contribution from mutual neutralization remains almost constant.

The determination of $H^-$ densities instead of density ratios, requires knowledge of the atomic hydrogen density. A very precise method is two-photon excitation laser-induced fluorescence (LIF) [42], with the drawback of being very complex and expensive. An alternative is the analysis of absolute line emission of $H_\gamma$ [41], where the effective excitation from atomic hydrogen is the dominant excitation path. This is applicable to the negative ion sources under discussion, since contributions of mutual neutralization, i.e. $H^-$ particles, on the population of quantum numbers
p > 4 and thus Hγ, Hδ, etc, are negligible (see figure 2). According to equations (1) and (6) Hγ line emission is given by:

\[ H_\gamma \equiv \varepsilon_{52} = n_H A_{52} = n_H n_e X_{eff,H}^H(T_e, n_e). \] (7)

In order to determine the atomic hydrogen density from measured Hγ line emission, electron temperature and electron density must be known. The effective emission rate coefficient for Hγ, \( X_{eff,H}^H \), is shown in figure 7 for the relevant parameter range.

In conclusion, the combination of the two line ratios Hα/Hβ and Hβ/Hγ is very well suited for the determination of H−/H density ratios: Hα/Hβ is related to the negative ion density whereas Hβ/Hγ monitors changes in plasma parameters such as electron density and electron temperature (figure 6). Analysis of absolute Hγ line emission yields the atomic hydrogen density and thus the absolute particle density of H− in the plasma volume observed by the LOS chosen. Of course, an independent measurement of electron density and electron temperature from other diagnostic techniques, such as Langmuir probes, is favoured; however, in the way suggested, the consistency of one diagnostic technique can be checked.

For deuterium, and thus D− densities, the identical method and effective rate coefficients are applicable since effective excitation of atomic hydrogen does not differ for the isotopes. The same applies to H+ processes and it is assumed that this is applicable also to the mutual neutralization (H− process). An isotope effect exists for the contributions from molecules (H2 and H2+) since the cross sections are different; however, these processes are of no relevance here.
2.4. Formula for a straightforward analysis

The analysis of negative ion densities from the H$_\alpha$/H$_\beta$ line ratio is based on the calculations from the CR model and expressed in subsection 2.3 in graphical correlations with electron temperature and electron density as parameter. The parameters are either known by other diagnostic techniques or obtained by OES. The latter requires a simultaneous recording of H$_\beta$/H$_\gamma$ line ratios and H$_\gamma$ line emission, which in turn requires an absolute calibrated spectroscopic system. For the analysis of a sequence of discharges and for plasma monitoring the point-by-point analysis is very time consuming and a formula for a straightforward analysis becomes highly desirable. The self-consistent analysis of spectroscopic signals is the basis for an united formula which allows also for direct monitoring of negative ion densities by the H$_\alpha$/H$_\beta$ line ratio.

Since effective excitation from atomic hydrogen and negative ions are the dominant population processes for quantum numbers $p = 3$ and $p = 4$, only term one and term five of equation (6) have to be taken into account. As discussed before, the H$^+$ density of term five can be replaced by the electron density. Thus, the H$_\alpha$/H$_\beta$ line ratio is given by:

$$\frac{H_\alpha}{H_\beta} = \frac{n_HX_{H_\alpha}^{\text{eff},H} + n_{H^-}X_{H_\alpha}^{\text{eff},H^-}}{n_HX_{H_\beta}^{\text{eff},H} + n_{H^-}X_{H_\beta}^{\text{eff},H^-}}. \tag{8}$$

The direct dependence of the line ratio on electron density has vanished; however, the effective rate coefficients depend on electron density and electron temperature. Using equation (7) for the atomic hydrogen density, a formula for the negative ion density can be obtained:

$$n_{H^-} = \frac{H_\gamma}{n_e} C_1 \left( \frac{H_\alpha}{H_\beta C_2} - 1 \right) \left( 1 - \frac{H_\alpha}{H_\beta C_3} \right)^{-1}. \tag{9}$$
Figure 8. Dependence of the factors $C_1$, $C_2$ and $C_3$ (as defined in equation (9)) on electron density for three electron temperatures.

with

$$C_1 = \frac{X_{\text{eff},H}}{X_{\text{eff},H}^-} \frac{1}{X_{\text{eff},H}^-}; \quad C_2 = \frac{X_{\text{eff},H}}{X_{\text{eff},H}^-}; \quad C_3 = \frac{X_{\text{eff},H}^-}{X_{\text{eff},H}^-}.$$

The factors $C_1$, $C_2$ and $C_3$ represent ratios of effective emission rate coefficients with the consequence that the strong dependence of rate coefficients on electron temperature vanishes in these factors. The factor $C_2$ can be interpreted as the line ratio $H_{\alpha}/H_{\beta}$ which is expected when excitation takes place from atomic hydrogen only (curve labelled with H in figure 4 and parallel lines at low $H^-/H$ density ratios in figure 6). The factor $C_1$ reflects the line ratio $H_{\alpha}/H_{\gamma}$ which is obtained by effective excitation of atomic hydrogen divided by the effective mutual neutralization rate coefficient for $H_{\alpha}$. The latter, i.e. $X_{\text{eff},H}^-$, is almost independent of electron density and electron temperature. The factor $C_3$ represents the line ratio $H_{\alpha}/H_{\beta}$ expected when excitation takes place from negative hydrogen ions only. The three factors are calculated with the CR model and shown in figure 8 as a function of the electron density for three electron temperatures. As expected, these factors show a weak dependence on electron temperature; however, a dependence on electron density still exists.

The second term in parenthesis of equation (9) considers the contribution of negative ions to $H_{\beta}$ radiation and can be interpreted as a correction factor for the negative ion density. For electron densities lower than $5 \times 10^{18}$ m$^{-3}$ and electron temperatures higher than 1 eV, which is the typical parameter range of negative ion sources, this correction is in the range of 15% or less. However, a check for the individual case is necessary. Under these conditions, equation (9) can
be further simplified to:

\[ n_{H^-} = \frac{H_{\gamma}}{n_e} C_1 \left( \frac{H_\alpha}{H_\beta} \frac{1}{C_2} - 1 \right). \]  

(10)

The linear correlation between the line ratio and the negative ion density is illustrated in figure 9 for fixed electron temperature and \( H_\gamma \) line emission using the corresponding factors \( C_1 \) and \( C_2 \) (figure 8). Both formulae (9) and (10), allow for a direct monitoring of negative ion densities by the \( H_\alpha/H_\beta \) line ratio for constant plasma parameters.

For plasma parameters different from these high power, low pressure sources, excitation channels other than direct excitation from H and mutual neutralization of \( H^+ \) with \( H^- \) may contribute to the \( H_\alpha \) radiation. Introducing these channels results in a more complex analysis method than the one presented here. However, the \( H_\alpha/H_\beta \) line ratio may be a useful monitor for changes of the \( H^- \) density even for plasma parameters which does not exactly match the range stated at the beginning of this section.

3. Experiments

The \( H_\alpha/H_\beta \) line ratio method for negative ion densities is applied to and checked in great detail at the RF ion sources at IPP. As mentioned in section 1 these sources are currently under development for neutral beam injection system for the next generation of fusion experiments (ITER).

3.1. The IPP RF ion source for negative hydrogen ions

The plasma is generated in a so-called driver region (cylindrical geometry 24 cm in diameter, 14 cm in length) by inductive coupling (external 6 turn coil) at \( f \approx 1 \text{ MHz} \) with an available
generator power of \( P = 180 \text{ kW} \) at maximum. The plasma expands in a rectangular body (32 \( \times \) 59 \( \times \) 19 cm\(^3\), width \( \times \) length \( \times \) depth), the so-called expansion region. Details of the design of the RF source are given in [43, 44], whereas [12, 13] give an overview of the three test facilities at IPP. Due to the ITER requirements, the discharge operates typically in the pressure region \( p = 0.2–0.6 \text{ Pa} \) in hydrogen and deuterium. In order to increase the formation mechanism of negative ions by the surface effect, caesium is evaporated using a caesium oven mounted at the back plate of the rectangular body. The evaporation rate is adjusted through the oven temperature, being typically around 10 mg h\(^{-1}\). Close to the extraction system a magnetic filter field (\( B \approx 6 \text{ mT} \)) is applied to reduce the temperature of the electrons and, thus, to reduce the destruction rate for \( \text{H}^- \) by electron stripping. The extraction system consists of three grids: the plasma grid, the extraction grid and the grounded grid. The heated plasma grid (up to 250°C) and the water-cooled source body are at high voltage (\(-20 \text{ kV}\)). The extraction voltage (typically 5–11 kV) is applied between the plasma grid and the extraction grid. The extraction grid is equipped with permanent magnets system in order to bend the co-extracted electrons out of the beam and prevent them to be fully accelerated. The size of the extraction area is 74 cm\(^2\), but can be extended up to 390 cm\(^2\).

The RF sources have achieved and even exceeded the ITER requirements regarding extracted current densities of negative ions (\( j_{\text{H}} = 28 \text{ mA cm}^{-2} \), \( j_{\text{D}} = 20 \text{ mA cm}^{-2} \) at the calorimeter) with an electron to ion ratio \( \leq 1 \) at the ITER relevant pressure of 0.3 Pa. At present, discharges are limited to a pulse duration of 6 s, long pulse operation (up to 3600 s) as well as extensions in the source size (half size ITER source without extraction) are under construction [12, 13].

OES is meanwhile a standard diagnostic tool for the various plasma parameters \( (T_e, n_e, T_{\text{gas}}, n_H \text{ and caesium densities}) \) at the IPP test facilities. Details of results are given in [21]–[23]. Typical values for electron density and electron temperature at 100 kW input power and 0.6 Pa filling pressure are: \( T_e = 8 \text{ eV} \), \( n_e = 5 \times 10^{18} \text{ m}^{-3} \) and \( T_e = 3 \text{ eV} \), \( n_e = 5 \times 10^{17} \text{ m}^{-3} \) in the centre of the driver and close to the extraction system (3 cm distance to the grid), respectively. Close to the grid the neutral density of caesium (typically around some \( 10^{14} \text{ m}^{-3} \)) is rather low, about five orders of magnitude lower than the neutral atomic hydrogen density (\( \approx 10^{19} \text{ m}^{-3} \)). Singly ionized caesium is roughly 30 times higher than the neutrals, resulting in a typical density ratio of \( \text{Cs}^+/n_e = 10^{-2} \). The gas temperature is 1200 K and a density ratio of \( \text{H}/\text{H}_2 = 0.2 \) is measured.

3.2. Experimental set-up

The Balmer line emission (\( \text{H}_\alpha, \text{H}_\beta, \text{H}_\gamma \)) is measured with a low resolution survey spectrometer (\textit{PLASUS EmiCon} system) covering a wavelength range of \( \lambda = 200–870 \text{ nm} \) with a spectral resolution \( \Delta \lambda_{\text{FWHM}} = 1–1.8 \text{ nm} \). This system is optimized for plasma monitoring, which means time traces of several line integrals and their ratios, which can be individually defined, are recorded in real-time. The temporal resolution is limited by the exposure time (minimum: 3 ms). The optical fibre is equipped with a collimator lens imaging an plasma volume with 1 cm in diameter. The spectroscopic system is absolutely calibrated by means of an Ulbricht sphere and a deuterium lamp.

The measurements are mainly carried out at one of the three test facilities: Multi Ampere Negative Ion Test Unit (MANITU) [12, 13] with an extraction area of 300 cm\(^2\). A LOS is chosen which is parallel to and close to the extraction system. The optical axis has a distance of 3 cm to the grid, the length of the LOS, i.e. the plasma is 25 cm, covering the whole grid in this
3.3. Analysis of time traces

Figure 10 shows time traces of the line ratios $H_\alpha/H_\beta$ and $H_\beta/H_\gamma$ together with the absolute line intensity of $H_\gamma$ for a hydrogen discharge in MANITU at 0.65 Pa filling pressure and 80 kW input power. As discussed above, the $H_\beta/H_\gamma$ line ratio reflects electron density and electron temperature, whereas $H_\gamma$ emission reflects electron density, electron temperature and atomic hydrogen density (equation (7)). After a start-up phase, both signals are constant during the discharge representing stable plasma parameters.

The line ratio $H_\alpha/H_\beta$, which reflects the $H^-$ density, shows clearly a dip during extraction. This dip reflects a depletion of the $H^-$ particles. Although the extraction mechanism of negative ions is not yet fully understood, the formation of this dip might be interpreted in a qualitative way: initially, ions produced on the grid surface have the direction to flow into the plasma volume (due to the starting condition and the influence of the plasma sheath). They can be turned by magnetic fields and collisions with background particles towards the grid system. With extraction the extraction surface has a drastically changed shape and penetrates into the plasma volume. Ions which hit the extraction surface are immediately removed from the plasma. Thus, the ion density in the observed plasma volume is decreased by the extraction. Since the distance of the LOS to the extraction system is 3 cm, these measurements demonstrate the penetration depth of the extraction on the plasma and confirm the calculated survival length of a few cm for $H^-$ particles [13, 23].
Two additional effects are clearly visible: the increase in the first second, which represents the start-up phase, i.e. until equilibrium conditions for plasma and surfaces are reached, and the spike at 3.4 s which correlates with an electrical breakdown. They demonstrate the fast response of the H_α/H_β on changes in H^- densities.

For a quantification of the H^- density from the H_α/H_β line ratio (subsection 2.3) the plasma parameters have to be determined first. The electron temperature was determined to be 3 eV using another spectroscopic technique: analysis of absolute line emission of the diagnostic gases helium or argon. Since their absolute particle density is known from the partial pressure and since the addition of amounts on the 10% range does not affect the plasma parameters, this analysis allows the determination of T_e provided that the electron density is known [18, 20, 22]. According to figure 4 the measured line ratio of H_β/H_γ = 4 corresponds to an electron density of n_e = 5 × 10^{17} m^{-3}. Knowledge of electron temperature and electron density enables the determination of the atomic hydrogen density from the H_γ line emission. A density of n_H = 1.2 × 10^{19} m^{-3} is obtained.

When considering only effective atomic hydrogen excitation the corresponding H_α/H_β line ratio should be 4.6 at the plasma parameters determined above. However, the measured values are remarkably enhanced: H_α/H_β = 14.1 in the RF only phase (4–4.5 s) and H_α/H_β = 12.8 with extraction. This enhancement is due to the strong influence of H^- on the line ratio. Taking into account in the analysis the contribution of H^- on the H_α/H_β line ratio the measured line ratios yield density ratios of H^-/H = 1 × 10^{-2} and H^-/H = 8.6 × 10^{-3} in the RF only phase and with extraction, respectively. These values represent LOS averaged H^- densities of H^- = 1.2 × 10^{17} m^{-3} (RF only) and H^- = 1 × 10^{17} m^{-3} (during extraction). Thus, the negative ion density is roughly 20% of the electron density at a distance of 3 cm to the grid.

The electrically measured extracted current density of negative ions was j_H^- = 21.9 mA cm^{-2} at an electron to ion ratio of j_e/j_H^- = 0.7, reflecting a well caesium-conditioned source. Estimations show, that extraction of negative ion current densities of the order of 20 mA cm^{-2} require negative ion densities in front of the grid of about 10^{17} m^{-3} [45]. Thus measured negative ion densities are in accordance with measured extracted current densities.

Uncertainties in the measured absolute line intensities, line ratios, electron density, electron temperature and atomic hydrogen density have a direct effect on the precision of the negative ion density. Additional uncertainties arise from using a CR model. The accuracy of a CR model strongly depends on the quality of the input data and, since hundreds of cross section data are used, a determination of the overall accuracy is almost impossible. In total, the error of the absolute negative ion density is estimated to be lower than 40%. Comparison with other methods, such as cavity ring down spectroscopy and laser detachment is highly desirable. This work is in progress. First results of cavity ring down measurements are in agreement within the estimated error bar [46]. However, since some of the uncertainties are systematic ones, relative changes can be obtained within an error bar of 20%.

3.4. Analysis of a sequence of discharges

In order to follow the variation of Balmer line emission for a sequence of discharges with parameter changes (power, pressure, extraction voltage, caesium conditioning of the source, etc) time traces of spectroscopic signals are averaged over the second half of the beam-on-time, representing the phase with extraction (‘beam’), and the beam-off-time, representing the plasma phase without extraction (‘RF only’).
Figure 11. Sequence of hydrogen discharges in the RF source (MANITU) at constant power and pressure (70 kW, 0.44 Pa) with variations in the extraction voltage and during caesium evaporation.

Figure 11 shows measured signals of a sequence of hydrogen discharges with caesium evaporation. An evaporation rate of approximately 10 mg h\(^{-1}\) is chosen to keep the caesium conditioning of the source constant during the scan in extraction voltage, however, due to a complex caesium balance, the caesium conditioning might change either to an improved or degraded caesium effect and thus enhanced or reduced negative ion formation. The time between the pulses is three minutes, the total time for the sequence of discharges plotted in figure 11 is approximately 1 h. The source parameters, such as input power and filling pressure, are kept constant at \(P = 70\) kW and \(p = 0.44\) Pa respectively. The extraction voltage \(U_{ex}\) has been varied between 5 and 7.5 kV as plotted in the upper trace of figure 11. The extracted negative ion current density follows closely the variation in extraction voltage. Measured \(H_\alpha/H_\beta\) line ratios (‘RF only’ and ‘beam’) increase also with increasing shot number, i.e. time, however the flat top phase is reached at a later time. This can be interpreted as an ongoing enhancement of negative ions by the caesium effect and is not inconsistent with the behaviour of the extracted negative ion current density since the beam perveance was not matched in this sequence. The line ratio \(H_\beta/H_\gamma\) and the line emission were constant during one discharge, i.e. a difference between the two time intervals ‘beam’ and ‘RF only’ was not observed (only one symbol appears in figure 11). Both spectroscopic signals, \(H_\beta/H_\gamma\) and \(H_\gamma\)
show a weak variation with increasing discharge number, which results in an almost constant electron density, electron temperature and atomic hydrogen density. Since power and pressure are kept constant this is exactly what one would expect. Measurements show that the small amount of caesium in the discharge does not affect the plasma parameters. This is due to the fact that the density of neutral caesium and caesium ions are very low in comparison to the densities of the other neutral particles (\( \text{Cs}/\text{H} < 10^{-5} \)) and ions (\( \text{Cs}^+ / n_e < 10^{-2} \)), respectively [22].

The quantitative analysis of such a sequence of discharges is best done with the formula for the straightforward analysis (subsection 2.4). From the measured \( \text{H}_\beta/\text{H}_\gamma \) line ratio an electron density of \( n_e = 5 \times 10^{17} \text{ m}^{-3} \) is obtained at the known electron temperature of \( T_e = 3 \text{ eV} \). At these parameters the factors defined in equation (9) and plotted in figure 8 are: \( C_1 = 9.8 \times 10^{14} \text{ s m}^{-3} \), \( C_2 = 4.6 \) and \( C_3 = 133 \). Measured \( \text{H}_\alpha/\text{H}_\beta \) line ratios range from about 8 to 15. This means the second term in parentheses in equation (9) represent a correction of 10% and can be neglected. Using formula (10), the \( \text{H}_\alpha/\text{H}_\beta \) line ratios measured in the two time intervals of a discharge, ‘RF only’ and ‘beam’, results in the negative ion densities shown in figure 12 as a function of the extracted current density. The difference in negative ion density between the ‘RF only’ phase and the ‘beam’ phase is the ‘extracted’ negative ion density which represents the depletion of the ion density caused by the extraction. All three negative ion densities increase with the extracted current density due to continuous caesium evaporation (‘RF only’) and the increase in extraction voltage (‘beam’, ‘extracted’). The factor for the enhancement with respect to the discharge with

Figure 12. Correlation of spectroscopically measured negative ion densities with the extracted current density of \( \text{H}^- \) in the sequence of discharges shown in figure 11. Top: ‘RF only’ phase, middle: ‘beam’ on phase, bottom: difference between ‘RF only’ and ‘beam’.
lowest current density (first discharge of this sequence) is highest for the ‘extracted’ negative ion density, i.e. the difference of the negative ion density measured in the ‘RF only’ phase and the ‘beam’ phase. The ‘extracted’ negative ion density represents the depletion of negative ions in the plasma volume close to the extraction system during extraction. Therefore, a direct proportionality with current density is expected and can been seen in figure 12 (bottom): the current density and the ‘extracted’ negative ion density increase by the same factor, namely 1.41 and 1.46 respectively, which is within the error bar of the fit.

Due to the fact that the negative ions are created at the plasma grid via the surface process and destroyed in the plasma volume by electron stripping and mutual neutralization, the negative ion density should increase towards the extraction system. Hence, the extracted current density represents an average negative ion density with respect to the distance to the grid. This aspect has to be taken into account for a conversion of the measured negative ion densities (plasma cylinder with 1 cm diameter at 3 cm distance to the grid) to current densities. Negative ion densities in the ‘RF only’ interval are close to $10^{17}$ m$^{-3}$. Measurements using two LOS with 3 and 2 cm distance to the grid system, respectively, have been carried out at the second test facility with similar parameters [23]. As expected the negative ion density increases at small distances, a factor of 1.5 was obtained.

The negative ion densities achieved in these ion sources depend strongly on the caesium conditioning. Thus, monitoring the $H_\alpha/H_\beta$ line ratio during caesium evaporation at identical source parameters is the first choice for monitoring the improvement of the caesium conditioning of the source. Measurements carried out in a caesium free source, for example after opening and cleaning the source, result in negative ion densities of $1 \times 10^{16}$ m$^{-3}$, at 80 kW input power and a pressure of 0.6 Pa. In this case the negative ions are produced via the volume process, i.e. the associative detachment, being in agreement with the typical extracted negative ion current densities of 2–4 mA cm$^{-2}$ at this pressure. Thus, the enhancement by the surface effect, already mentioned in the introduction, is an order of magnitude in negative ion densities as well as in extracted current densities.

The line ratio method has been applied to deuterium discharges. The comparison with hydrogen discharges at similar source parameters such as pressure, power, caesium conditioning of the source, results in roughly a factor of 1.5 higher negative ion densities for deuterium [23]. It is explained by the slightly lower electron temperature in deuterium plasmas reducing the destruction of negative ions via the electron stripping. This is confirmed by measurements of the source efficiency, i.e. extracted current density divided by the input power, which is higher in deuterium than in hydrogen [13].

4. Conclusions

A novel diagnostic technique for negative hydrogen ion densities in high power, low pressure ion sources is introduced. The method is based on the contribution of the mutual neutralization process to Balmer line emission, in particular the selective effect on $H_\alpha$ line emission. This selective effect is illustrated by calculations of the individual contributions of five excitation channels to the Balmer line emission. Using the fact that, in the parameter range of interest, effective excitation from atomic hydrogen and mutual neutralization are the dominant population mechanism, a simple method for determining the density ratio $H^-/H$ from the line ratio $H_\alpha/H_\beta$ is obtained. Thus, line-averaged negative ion densities, and in particular their variation can be
measured in situ by OES. The method itself has the advantage of being non-invasive with a simple experimental set-up, providing temporal and (limited) spatial resolution. In combination with measurements of the line ratio $H_\beta/H_\gamma$ and the $H_\gamma$ line emission plasma parameters can be obtained simultaneously. For a straightforward analysis of negative ion densities from the $H_\alpha/H_\beta$ line ratio a formula is presented which gives a linear correlation of the $H_\alpha/H_\beta$ line ratio with the negative ion density and which is the first choice for monitoring negative ion densities.

The method was successfully applied to the RF excited negative ion sources of IPP. A manifold of valuable results are obtained from in situ monitoring of emission lines and line ratios. Negative ion densities of $10^{16}$–$10^{17}$ m$^{-3}$ are obtained, depending on the caesium conditioning of the source, the input power, and the pressure. This corresponds to a negative ion to electron density ratio of 0.02–0.2 at a distance of 3 cm to the extraction system. It was shown, that negative ion densities in the plasma volume correlate well with extracted ion current densities. Since the analysis method depends on the solely usage of atomic data, the method is directly applicable to deuterium.

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