Spectroscopic Study of Neutral Species in a Planar-Coil Inductive Discharge in Hydrogen


This work has been carried out within the framework of the EUROfusion Consortium and has received funding from the European research and training programme 2014–2018 under grant agreement No 633053. The views and opinions expressed herein do not necessarily reflect those of the European Commission.
Spectroscopic study of neutral species in a planar-coil inductive discharge in hydrogen

S. Iordanova, Ts. Paunska and A. Pashov
Faculty of Physics, Sofia University, 5, J. Bourchier, BG-1164 Sofia, Bulgaria
E-mail: snejana@phys.uni-sofia.bg

May 2015

Abstract. This contribution reports on experimental study on characteristics of the neutrals in a single element of a matrix source of negative hydrogen ions. The investigations are carried out in hydrogen discharges maintained at a frequency of 27 MHz, applied power varied in the limits $P = (50 – 150)$ W and pressure in the range $p = (90 – 160)$ mTorr. An optical emission spectroscopy diagnostics, based on analysis of the H$\alpha$ spectral line profile and the intensity distribution of the Fulcher-$\alpha$ molecular band, is used. The profile of the atomic line reveals the existence of thermal as well as of non-thermal (fast) hydrogen atoms in the discharge. For processing the experimental data a line shape model, accounting for details of the plasma kinetics and the fine structure of the line, is used. The temperature of the atoms in the excited $n = 3$ state, the mean energy of the fast atoms, the ratio between the densities of these two groups of atoms and the relative population of the fine structure components of the $n = 3$ hydrogen state are determined. The atomic temperature is estimated to be equal to the temperature of the excited ($n = 3$) atoms. The molecular temperature is determined from the analysis of the intensity distribution of the molecular Fulcher-$\alpha$ band and is found to be about two times lower than the atomic temperature.

1. Introduction

Optical emission spectroscopy is employed as a powerful tool, providing information for various plasma characteristics like type of species, particle densities, temperatures, etc. Investigation of some of them is convenient to be carried out through analysis of the spectral line profiles, registered at high resolution. Plasma parameters obtained from spectroscopic studies are necessary in the optimization of plasma sources and as input data in numerical simulations of gas discharges. The current study is a part of activities on diagnostics of a single element of a matrix source of negative hydrogen ions. The single element is a small radius discharge inductively driven by a planar coil and equipped with a device for detection of the ions. The development of a matrix of such sources [1, 2] is motivated by their potential application in the neutral beam injection systems, used for the additional plasma heating in fusion reactors [3].

The study is mainly dedicated to spectroscopic determination of the temperature of the ground state atoms and molecules at various experimental conditions. Very often due to the lack of information in the simulations, the temperature of the ground state atoms is taken as equal to the molecular temperature, which is not always correct.
The excessive broadening of hydrogen Balmer lines [4, 5] is subject of a series of spectroscopic studies of hydrogen discharges. In general, three components of the line profile may be clearly distinguished [6, 7] and different excitation process could be attributed to each of them. The narrowest central component of the profile results from electron excitation of thermalized atomic hydrogen and its width is related to the velocity distribution of these atoms. A border component, forming a pedestal of the line profile, is related to the excited hydrogen atoms, generated by dissociative excitation or dissociative ionization of H₂ by electron collisions. The products of these reactions are hydrogen atoms in the ground and excited states with a mean kinetic energy of up to 8 eV [5]. The broadest component represents excited hydrogen atoms created in the charge exchange reactions. The positive hydrogen ions (H⁺, H₂⁺, and H₃⁺) are accelerated by the discharge electric field and they can produce neutral atoms, with significant kinetic energies (from tens to hundreds eV [8, 9]) in charge transfer or dissociation collisions with H₂. Then the ground state atoms can be excited and the following emission forms a very broad feature in the base of the spectral line.

Spectroscopic investigations of rotation-vibrational emission bands in diatomic molecules is a well-established technique for determination of rotational temperature [10]. The latter can be related to the temperature of the molecules in their ground state after obtaining the correct relationship between the populations in the excited and ground state. For most low-pressure plasmas the model can be relatively simple [11].

A method, previously developed in [12] and based on interferometric analysis of the Hα spectral line profile, is used for estimation of the temperature of ground state atoms. It includes an experimental part for registration of the spectral line profile with high resolution and a theoretical part including a reliable model which accounts for the various broadening mechanisms. The theoretical model assumes a uniform spatial distribution of the atomic temperature. It takes into account the fine structure of the Balmer Hα line and also the presence of thermal as well as of non-thermal (fast) hydrogen atoms in the n = 3 state. As a result the Hα line profile is described with a non-Gaussian function. Its parameters give information about the temperature of the thermal atoms and the mean energy of the fast atoms in the excited n = 3 state, about the ratio between the thermal and the fast atom densities as well as about the relative population of the fine structure components of the investigated hydrogen state. The temperatures of the hydrogen atoms in n = 3 and ground states are estimated to be equal. In order to obtain the molecular temperature, analysis of the intensity distribution of the rotational spectral lines of the Fulcher-α band is carried out. The results reveal that the molecular temperature is about two times lower than the atomic temperature.

2. Experimental arrangements

A detailed description of the experimental setup – a planar-coil inductive discharge – is given in [2]. As it is described there, the source is supplied with a Faraday cup (FC, the device for detecting the negative ions) positioned at the end of the first chamber (Fig. 1). The latter is a quartz tube with a radius of 2.25 cm and a length of 15 cm. The gas flow rate is controlled by an MKS Mass-Flo controller. It is varied between 7.5 and 22.5 sccm which results in gas pressures in the first chamber between 90 and 160 mTorr. In the volume behind the FC the gas pressure is about 6 mTorr. The discharge is driven by a 3.5-turn planar coil mounted on a quartz window which is at
the front wall of the first chamber. The discharge is maintained in a capacitive mode at 27 MHz by rf power varied in the range from 50 to 150 W.

Plasma characteristics of the neutral hydrogen particles are assessed by applying two setups for optical emission spectroscopy. The first one, based on a Fabri-Perot interferometer [12], is used for registration of the atomic hydrogen H$_\alpha$ line profile, whereas the second experimental setup, including a monochromator, registers the molecular hydrogen Fulcher-\(\alpha\) band.

The light from the discharge is collected by a liquid light guide (LG, 6 mm inner diameter, placed at a distance of about 5 mm from the quartz tube) in radial direction through a movable window in the electromagnetic shield at an axial position from the coil $z = 2 \div 10$ cm. A typical spectrum is shown in Fig. 2. The data are normalized to the H$_\alpha$ line intensity.

2.1. Interferometric setup

The interferometric setup, used for registration of the Balmer H$_\alpha$ spectral line profile, is described in details in [12], so only a brief overview is given here. It consists of a commercial IT51-30 Fabri-Perot interferometer (FPI) with a distance between the mirrors (reflectivity higher than 90\%) $d = 0.6$ mm. The latter ensures a free spectral range (FSR) of about 3.5 Å.

The LG delivers the light collected from the discharge to the optical setup (see Fig. 3). After collimation with the lens L, the light illuminates the FPI through a
band pass interference filter (IF, with central wavelength of 655 nm and 9.5 nm full width at half maximum (FWHM)) for separation of the H\textsubscript{\alpha} spectral line.

![Figure 3. Experimental arrangement for the interferometric diagnostics.](image1)

The light intensity distribution coming out of the FPI is detected by a digital camera Canon EOS 600D (with 14-bit Analog-to-Digital converter) supplied with a Canon lens EF 50 mm f/1.8 Mk II. All the components of the optical setup are arranged and fixed on an optical breadboard with vibration isolation.

For adjustment of the FPI as well as for determination of the apparatus function of the setup a single mode diode laser (Fig. 4), is used as a monochromatic source. In this case the light coming out of the optical fiber is quite anisotropic. A uniform illumination of the FPI is realized by a ground glass diffuser positioned right after the output of the LG (Fig. 3). The laser beam with a wavelength around 650 nm is guided through a beam splitter (BS) to a high finesse confocal cavity (FSR of 750 MHz). The latter is used to control the mode structure of the laser. Images of the interference maxima of the H\textsubscript{\alpha} (Fig. 5) at various experimental conditions are recorded at ISO numbers from 400 to 1600, exposure times from 30 to 240 s and lens apertures f/8 or f/9. The images are collected in a black and white mode of the camera operation. They are saved in a raw image format (i.e. linear gray scale and without modifications by the graphical processor of the camera) and later they are converted into ASCII format.

Due to the circular symmetry of the interference fringes, cylindrical coordinates \( (r, \theta) \) for pixel designation proved to be more appropriate than the Cartesian \( (x, y) \). \( r \) is the distance between the pixel and the center of the rings. A specialized code finds the center of the interferogram and averages the intensity over a preselected range of angles \( \theta \). This leads to significant increase of the signal-to-noise ratio (the ratio between the highest peak and the standard deviation of the noise) from 30-40 to generally above 200. Finally, the output of the interferometric setup is the dependence of the averaged intensity of the interference fringes on \( r \): \( I_{\text{ring}}(r) \) (Fig. 6).

2.2. Spectrometric system including monochromator

The second experimental arrangement, used for registration of the hydrogen Fulcher-\( \alpha \) molecular band \( (d^3\Pi_u \rightarrow a^3\Sigma^+_g) \) (Fig. 7), is described in details in [13]. It consists of a monochromator MDR-23 (Lomo-Russia) with a focal length of 0.6 m, equipped with a Hamamatsu photomultiplier tube PMT R928 and a grating with 1200 grooves/mm. The wavelength tuning and spectra calibration are computerized. The entrance and the exit slits of the monochromator are fixed at 40 \( \mu \)m. The signal from the PMT is detected through a Stanford Research System Lock-in amplifier SR830 in order to reduce its thermal noise.
Figure 5. A negative image of interferogram.

Figure 6. The dependence of the intensity on \( r \) averaged over a preselected range of \( \theta \).

Figure 7. Hydrogen Fulcher-\( \alpha \) rovibrational band at \( p = 130 \) mTorr, \( P = 150 \) W and \( z = 2 \) cm. The Q-branch lines are labeled.

3. Analysis of the experimental observations

3.1. Atomic spectra

Generally, the intensity of the interference fringes \( I_{\text{ring}}(r, a_{\text{line}}) \) is a convolution of the spectral line profile \( I_{\text{line}}(\lambda, a_{\text{line}}) \) and the apparatus function of the setup \( I_{\text{app}}(\lambda_0, r) \). Here with \( a_{\text{line}} \) the set of parameters of the spectral line (central wavelength (\( \lambda_0 \)), amplitude (\( A_{\text{amp}} \)), line width (\( \Delta \lambda \)) etc.) is denoted.

In order to extract information about the line profile from \( I_{\text{ring}}(r, a_{\text{line}}) \) it is necessary to find the apparatus function \( I_{\text{app}}(\lambda_0, r) \). The latter corresponds to the light intensity distribution on the camera sensor in case of a monochromatic illumination (with wavelength \( \lambda_0 \)) of the setup.

3.1.1. Determination of the apparatus function

The apparatus function is determined by the diode laser, tuned at the wavelength of \( \text{H}_\alpha \). A careful examination of the typical interferogram (Fig. 6) shows that \( I_{\text{app}}(\lambda_0, r) \) differs from the idealized FPI transmission function (for details see [12]). In fact, the real transmission function should take into account the possible variations of the reflectivity of the dielectric mirrors \( R \) with the angle of incidence \( \varphi \). The connection between \( \varphi \) and \( r \) is expressed
as:

\[ r = f \tan(\varphi), \]

where \( f \) is the focal length of the camera lens. The dependence of the mirror reflectivity on \( \varphi \) (respectively \( r \)) leads to an \( r \) dependent coefficient of finesse \( F \):

\[ F(r) = \frac{4R(r)}{(1 - R(r))^2}. \]

A linear dependence of the mirror reflectivity on the angle of incidence is assumed:

\[ R(r) = R_0 + R_s \cdot r, \]

where \( R_0 \) and \( R_s \) are the reflectivity of the mirrors at \( r = 0 \) (\( \varphi = 0 \)) and the slope, respectively. In the transmission function it is also necessary to account for the nonuniform illumination of the FPI:

\[ I_{\text{FPI}}(\lambda_0, r) = \frac{I_0(r)}{1 + F(r) \sin^2 \frac{\delta}{2}}. \]

Here \( I_0(r) \) is the integral intensity of the light which propagates at angle \( \varphi \) with respect to the optical axis of the interferometer. Again, a linear dependence (see Fig. 6) on \( r \) is assumed: \( I_0(r) = A_0 (1 + Sr) \). \( A_0 \) denotes the intensity at \( \varphi = 0 \) (\( r = 0 \)) and \( S \) is a parameter accounting for the variation of the intensity with \( r \). In eq. (3) \( \delta \) is the phase shift, expressed as \( \delta = 4\pi d \cos \varphi \lambda_0^{-1} \), where \( \lambda_0 \) is the central wavelength in air.

Regardless of these modifications, the transmission function of the FPI, eq.(3), still can not reproduce the observed intensity of the interference fringes \( I_{\text{ring}}(r, \text{line}) \).

It is important also to consider the nonzero background \( I_{\text{backgr}}(r) \) of the experimental interferograms (Fig. 6) and the possible broadening of the fringes due to residual angle between the mirrors and imperfectness of their surfaces. So finally the intensity of the fringes after illumination with monochromatic light is modeled as [12]:

\[ I_{\text{ring}}(r, \text{line}) = I_{\text{app}}(\lambda_0, r) + I_{\text{backgr}}(r) = \]

\[ = \int_{\lambda} I_G(\lambda, \lambda_0, \Delta \lambda_{\text{Gaf}}) I_{\text{FPI}}(\lambda, r) d\lambda \]

\[ + I_{\text{backgr}}(r), \]

where \( I_{\text{FPI}}(\lambda, r) \) is the transmission function of the FPI from eq.(3). The additional broadening of the fringes is accounted by a convolution of \( I_{\text{FPI}}(\lambda, r) \) with a Gaussian function \( I_G(\lambda, \lambda_0, \Delta \lambda_{\text{Gaf}}) \) with FWHM \( \Delta \lambda_{\text{Gaf}} \). A linear dependence \( I_{\text{backgr}}(r) = a + br \) for the interferogram background is assumed.

By fitting the laser interferogram with the apparatus function described with eq. (4), the standard deviation of the residuals (less than \( 6 \times 10^{-3} \), see Fig. 8) turns out comparable with the estimated noise level. Such a good correlation between experiment and theory is obtained when the first two fringes are fitted. Fixed parameters in the model of the apparatus function are \( \lambda_0 \) and \( d \), whereas \( R_0, R_s, S, A_{\text{amp}}, \Delta \lambda_{\text{Gaf}} \) and the parameters \( a \) and \( b \) are fitted. The parameter values of the apparatus function used further in the study are summarized in Table 1.
3.1.2. Model for the line profile  The H\textsubscript{\alpha} line consists of seven components due to the fine structure of the hydrogen levels with \( n = 3 \) and \( n = 2 \). The exact wavelengths of these components \( \lambda_{0,i} \) are well known \[14\]. When thermal equilibrium is assumed their relative intensities \( I_{\text{rel},i} \) can be calculated from the Einstein coefficients for spontaneous emission and the statistical weights of the corresponding levels \[14\]. The relative population of the \( 3^2S \), \( 3^2P \) and \( 3^2D \) states however may be different, because the rates for their population need not to be equal. This is accounted in the model by introducing correction coefficients \( k_{P/D} \) and \( k_{S/D} \) for adjusting the intensities of the lines emitted from the \( 3^2P \) and \( 3^2S \) states with respect to those emitted from the \( 3^2D \) states.

As it is shown in \[12\], besides the thermal hydrogen atoms, there are fast quasi monoenergetic atoms formed under the investigated gas discharge conditions. They are products of reactions leading to excited hydrogen molecules or molecular ions, followed by decay to repulsive molecular states and, finally, dissociation to a pair of H(1s) (or H\textsuperscript{+}) and H\textsuperscript{*}(\( n = 3 \)) atoms with significant excess energy (up to 8 eV \[5, 15\]). The profile of the H\textsubscript{\alpha} spectral line associated with these atoms due to the Doppler effect is nearly rectangular and appears as a broad pedestal in the base of the Gaussian-like peak emitted by the thermal atoms. Finally, the model of the H\textsubscript{\alpha} which takes into account all these effects can be expressed as \[12\]:


\[ I_{\text{line}}(\lambda, a_{\text{line}}) = A_{\text{amp}} \sum_{i=1}^{7} I_{\text{line},i}(\lambda, \lambda_0, i, I_{\text{rel}}, k_i, \Delta \lambda) + I_{\text{ped}}(\lambda, \Delta \lambda_{\text{ped}}, \delta \lambda), \]

where

\[ I_{\text{ped}}(\lambda, \Delta \lambda_{\text{ped}}, \delta \lambda) = A_{\text{ped}} \begin{cases} 1 & \text{for } |\lambda_0 - \lambda| \leq \frac{\Delta \lambda_{\text{ped}}}{2} \\ \exp \left(-\frac{(\lambda_0 - \lambda)^2}{0.36 \Delta \lambda_{\text{Gaf}}^2}\right) & \text{for } |\lambda_0 - \lambda| > \frac{\Delta \lambda_{\text{ped}}}{2} \end{cases}. \]

Here \( A_{\text{amp}} \) is the overall line amplitude, \( k_i \) is intensity correction coefficient: \( k_i = 1 \) for lines emitted from \( 3^2D \) states whereas \( k_i = k_{P/D} \) and \( k_i = k_{S/D} \) for lines emitted from \( 3^2P \) and \( 3^2S \) states, respectively. \( I_{\text{ped}} \) is the pedestal function depending on the amplitude \( A_{\text{ped}} \), FWHM \( \Delta \lambda_{\text{ped}} \) as well as on \( \delta \lambda \), reflecting the possible energy distribution of fast atoms around a nonzero mean value.

The intensity distribution of the interferogram may be described as a convolution of the line model eq. (5) and the apparatus function eq. (4) as follows:

\[ I_{\text{ring}}(r, a_{\text{line}}) = \int_{\lambda} \frac{I_{\text{line}}(\lambda, a_{\text{line}})I_{\text{app}}(\lambda, r) \, d\lambda}{\lambda} + I_{\text{backgr}}(r), \]

where \( I_{\text{backgr}}(r) \) takes into account possible variation of the image background.

With the determined apparatus function \( I_{\text{app}}(\lambda_0, r) \), the line parameters \( a_{\text{line}} \) are derived by fitting the experimentally observed interferogram \( I_{\text{ring}}^{\text{exp}}(r) \) with the \( I_{\text{ring}}(r, a_{\text{line}}) \) function (eq. (7)). During the fit, fixed parameters are \( \lambda_0 \) and \( d \) as well as the parameters describing the apparatus function (\( R_0 \), \( R_a \), \( A_{\text{amp}} \), \( \Delta \lambda_{\text{Gaf}} \) (see Table 1)). Free parameters are \( \Delta \lambda \), \( S \), \( A_{\text{amp}} \), \( A_{\text{ped}} \), \( \Delta \lambda_{\text{ped}} \), \( k_{P/D} \), \( k_{S/D} \) as well as the parameters \( a \) and \( b \). It is found out that reasonably good fit is obtained by fixing \( \delta \lambda \) to the Doppler width \( \Delta \lambda \) of the H\( \alpha \) components. No difference in the fit quality is observed independently whether the profile of the fine structure components is approximated with a Gaussian or with a Voigt function. Therefore hereinafter in this study Gaussian functions are used for \( I_{\text{line,ii}}(\lambda, \lambda_{0,ii}, I_{\text{rel,ii}}, k_i, \Delta \lambda) \) in eq. (5).

An example demonstrating the application of the fitting procedure is shown in Fig. 9 and the obtained fitted parameters are summarized in Table 2. A comparison between the apparatus function (blue line) and the spectral line profile is presented as a zoomed image in the top part of that figure. The residuals \( I_{\text{ring}}^{\text{exp}}(r) - I_{\text{ring}}(r, a_{\text{line}}) \) presented in the bottom part of Fig. 9, show satisfactory distribution, except around the fringe maxima where similar systematic deviations could be seen at different experimental conditions (Fig. 10). Currently it is difficult to figure out whether these variations are due to the incomplete model or they come from the experimental setup (ghosts or presence of weak spectral lines close to H\( \alpha \)).

Due to the complexity of the experimental setup it is not straightforward to assess the experimental errors of \( I_{\text{ring}}^{\text{exp}}(r) \). Therefore, the estimation of the fit quality is based on the distribution of the residuals. The standard deviation of the latter is taken as experimental error and the estimated uncertainties of the fitted parameters through
Figure 9. Results of the fit of the interference fringes ($z = 2$ cm, $p = 130$ mTorr, $P = 150$ W).

Table 2. Obtained values of the free parameters in the $H\alpha$ simulations at $p = 130$ mTorr, $P = 150$ W and $z = 2$ cm.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>Uncertainty</th>
</tr>
</thead>
<tbody>
<tr>
<td>$A_{amp}$</td>
<td>[arb.u.]</td>
<td>0.0770 $\pm$ 2 $\times$ 10$^{-3}$</td>
</tr>
<tr>
<td>$\Delta \lambda$</td>
<td>[nm]</td>
<td>1.328 $\times$ 10$^{-2}$ $\pm$ 0.5 $\times$ 10$^{-3}$</td>
</tr>
<tr>
<td>$k_{P/D}$</td>
<td></td>
<td>2.300 $\pm$ 0.08</td>
</tr>
<tr>
<td>$k_{S/D}$</td>
<td></td>
<td>1.0 $\times$ 10$^{-5}$ $\pm$ 1.3</td>
</tr>
<tr>
<td>$A_{ped}$</td>
<td>[arb.u.]</td>
<td>6.83 $\times$ 10$^{-2}$ $\pm$ 3 $\times$ 10$^{-3}$</td>
</tr>
<tr>
<td>$\Delta \lambda_{ped}$</td>
<td>[nm]</td>
<td>1.205 $\times$ 10$^{-1}$ $\pm$ 0.03 $\times$ 10$^{-1}$</td>
</tr>
<tr>
<td>$S$</td>
<td>[pix$^{-1}$]</td>
<td>-0.2510 $\pm$ 8 $\times$ 10$^{-4}$</td>
</tr>
<tr>
<td>$a$</td>
<td>[arb.u.]</td>
<td>6.915 $\times$ 10$^{-2}$ $\pm$ 9 $\times$ 10$^{-4}$</td>
</tr>
<tr>
<td>$b$</td>
<td>[pix$^{-1}$arb.u.]</td>
<td>3.59 $\times$ 10$^{-5}$ $\pm$ 2 $\times$ 10$^{-6}$</td>
</tr>
</tbody>
</table>

Figure 10. Residuals from the fit of interference fringes at different experimental conditions.

the matrix of variances and covariances (confirmed also by Monte Carlo simulations) are based on this assumption.
Spectroscopic study of neutral hydrogen species in inductive discharge

3.2. Molecular spectra

One of the molecular bands widely used for diagnostics of hydrogen discharges is the Fulcher-α (d^3Π_u → a^3Σ^+_g). The main reasons are that it is intense and can be clearly identified over the visible wavelength range from 600 to 625 nm (Fig. 2). The rotational components belonging to the Q-branches of the first three vibrational transitions (0-0), (1-1) and (2-2) have well known wavelengths and could be relatively easily resolved. In addition, all necessary molecular constant characterizing the ground and the excited d^3Π_u and a^3Σ^+_g states are available [16]. The spin-orbit and the spin-rotation interactions in the d^3Π_u and the a^3Σ^+_g states are weak, so the splitting of the rotational levels of these states is negligible within the resolution of this study. Due to the Λ-type doubling the energy levels of the d^3Π_u state are twofold degenerated, so two levels of different parities correspond to every combination of rotational and vibrational quantum numbers – the so called e and f levels. In some sources the manifold of the e levels is said to belong to the d^3Π_u state, and the f levels belong to the d^3Π_u state. According to the selection rules for electronic transitions, d^3Π_u → a^3Σ^+_g band system can only have Q-branches, whereas the d^3Π_u → a^3Σ^+_g system possesses only P and R branches. The e levels of the d^3Π_u state are strongly perturbed due to interactions with the neighboring e^3Σ^+_u state and, therefore, the P and R branches of the Fulcher-α band are not suitable for diagnostics.

The analysis of the Q bands, done in the study, relies on the following assumptions: (i) The population of the rotational levels in the ground state follows the Boltzmann distribution; (ii) The d^3Π_u state is excited from the ground molecular state by direct electron impact and decays spontaneously; (iii) The excitation channel does not change the rotational quantum number (ΔJ = 0).

The main mechanisms for establishing a Boltzmann distribution in the population of the rotational levels in the ground state are the inelastic collisions between molecules as well as the collisions with the walls. At temperatures lower than 1500 K (expected in such type of hydrogen discharges [13, 17]) almost all molecules are in the lowest vibrational ground state (v'' = 0) [18, 19] and the population of the rotational states follows the Boltzmann distribution for the given molecular temperature. The electron impact excitation does not change the rotational quantum number J and hydrogen molecules are excited only from one vibrational state (v'' = 0), therefore the population of the rotational states within each v' of the excited state follows the same distribution as for the ground X^1Σ^+_g state. Here it is taken into account that the characteristic time between heavy particle collisions (being of the order of hundreds ms) is much longer than the radiative lifetime of the d^3Π_u state (τ_rad = 31 ns [20]), so the redistribution of the population of the excited state is negligible. As a result, the following expression for the population of the rotational levels n_{v',J'} in the excited d^3Π_u state is obtained:

\[ n_{v',J'} = n_{v''} g_{J'} \exp \left( -\frac{B''_d J'(J' + 1)}{kT_m} \right) ; \]

where \( n_{v''} \) is a factor depending only on the vibrational quantum number of the d^3Π_u state, \( B''_d \) is the rotational constant of the ground state, \( T_m \) is the molecular temperature and \( g_{J'} = (2J' + 1)/(2t + 1) \) is the statistical weight of the level, as \( t = 0 \) for the even \( J' \) and \( t = 1 \) for the odd \( J' \) [10].

Taking into account the population of the excited d^3Π_u state (eq. (8)), the intensity \( I_{v'' \rightarrow v'} \) of the spectral lines of the \( ^3Π_u \rightarrow a^3Σ^+_g \) band can be expressed as:

\[ I_{v'' \rightarrow v'} \propto n_{v'',J''} A_{v'' \rightarrow v'} h \nu_{v'' \rightarrow v'} \propto n_{v',J'} \frac{q_{v''v'} C_{J',J''}}{\lambda^3(2J' + 1)} . \]
Here $A_{\nu'\nu}$ is the Einstein coefficient for spontaneous emission, $q_{\nu'\nu}$ is the Franck-Condon factor, and $C_{J',\mu'} = (2J' + 1)/2$ is the rotational line strength for the Q-lines.

Given the above mentioned assumptions, the relative intensities within the Fulcher-α band can be used for determination of the temperature $T_m$ of the ground state molecules. Detailed description of this procedure is given in [13, 17]. From eq. 9 one can plot the dependence of the quantity $\ln \left( \frac{\lambda^4 I_{\nu'\nu}}{q_{\nu'\nu} (2J' + 1)(2t + 1)} \right)$ as a function of $B_0'' J'(J' + 1)$:

$$
\ln \left( \frac{\lambda^4 I_{\nu'\nu}}{q_{\nu'\nu} (2J' + 1)(2t + 1)} \right) = -\frac{B_0'' J'(J' + 1)}{kT_m} + \ln(n_{\nu'}\text{const}).
$$

(10)

The temperature $T_m$ is determined from the fitted slope of the dependence whereas the free coefficients $\ln(n_{\nu'}\text{const})$ refer to the relative population of the $d^3\Pi_u$ vibrational states. The standard deviation of the linear fit is used for estimation of the experimental error of $T_m$.

4. Results

The results given below are obtained at a distance of 2, 6 and 10 cm away from the coil (Fig. 1). Hydrogen discharges are studied at gas pressures $p = 90, 130$ and 160 mTorr and applied powers $P = 50, 100$ and 150 W. Example of the fitted line profiles, obtained from analysis of the registered interferograms, are presented in Fig. 11. As it could be seen the variation of the experimental conditions leads to changes in the ratio of the areas under the curves describing the central peak and the pedestal. This ratio directly reflects the ratio between the densities $n_{th}$ and $n_f$ of the thermalized and the fast atoms ($n = 3$), respectively (Fig. 12). At the investigated gas pressures and applied powers $n_f$ increases towards the end of the discharge tube, reaching one-third of $n_{th}$. The changes in the ratio $n_{th}/n_f$ with the axial position are more substantial at higher pressures. These results reveal that along the discharge length different mechanisms may be responsible for the formation of thermal and fast atoms in $n = 3$.
state as well as that at lower pressures the existence of fast atom can not be neglected in the plasma kinetic modelling.

The values of the $k_{P/D}$ coefficients (Fig. 13), significantly higher than 1 for all experimental conditions, indicate higher rates for population of the $3^2P$ atomic state than of the $3^2D$ state. The values of the $k_{S/D}$ are statistically equal to zero. This can be explained by the low relative intensities of the $3^2S$ components which play almost no role in the formation of the $H_\alpha$ profile. The obtained $k_{P/D}$ values indicate that the rates for population of $3^2P$ and $3^2D$ states are quite different close to the coil in contrast to the remote axial positions.

The analysis of the Doppler width $\Delta \lambda_D$ of the central peak components of $H_\alpha$ gives information about the temperature of the thermal atoms in $n = 3$ (Fig. 14). The width of the pedestal, which corresponds to the energy of the fast atoms, at all experimental conditions remains nearly the same – approximately 0.13 nm (about 8 eV). The temperature of the thermal atoms rises with the axial position and this dependence is more pronounced at higher pressures. It is worth mentioning that at
z = 2 cm the temperature \( T_a \) decreases with the applied power.

The results for the molecular temperature (Fig. 15), obtained from Fulcher-\( \alpha \) band investigation reveal that the increase of the applied power leads to higher \( T_m \) along the discharge length. At the same time, the dependence of the molecular temperature on the axial position changes with the gas pressure. At \( p = 90 \) mTorr, it seems that \( T_m \) increases with \( z \). By increasing the gas pressure to 130 mTorr, \( T_m \) remains almost constant independently of the axial position. At \( p = 160 \) mTorr, the dependence of the \( T_m \) on \( z \) is not well pronounced, but it is likely to decrease towards the end of the discharge tube.

5. Discussion

The temperature of the thermal atoms \( (n = 3) \) is equal to the temperature of the hydrogen atoms in their ground state only in the case of direct electron-impact excitation of the H(1s) state: \( \text{H}(1s) + e \rightarrow \text{H}^*(n = 3) + e \), followed immediately by spontaneous decay (the so called corona model) [21]. Then the spectral line profile
can be approximated with Gaussian function corresponding to the temperature of the ground state atoms. The existence of pedestal, however, shows that under the investigated experimental conditions the direct electron-impact excitation is not the only process determining the population of \( n = 3 \) excited state. At the same time the processes, apart from direct electron excitation, resulting in production of hydrogen atoms in \( n = 3 \), are characterized with excess energies much higher than the measured thermal energies (in order of 0.1 eV, see Fig. 14). They produce much broader emission lines which can be easily distinguished from the emission of the thermal atoms. In addition, the rate of collisions between all type of particles are orders of magnitude lower than the decay rate of the \( n = 3 \) state, so it is very unlikely that the energy distribution of the excited thermal atoms is influenced by collisions. The thermalization of the hydrogen atoms takes place while they are in their ground state. Therefore it is reasonable to assume that under the investigated experimental conditions the corona model is still valid for the group of thermal atoms and the temperature of the \( n = 3 \) atoms is equal to the temperature of the atoms in the ground state.

As it could be seen from Figs. 14 and 15 the temperature of the hydrogen atoms is about two times higher than that of \( \text{H}_2 \). So it seems that the plasma contains two distinct neutral populations: molecules and atoms at significantly different temperatures. This experimental result is in a qualitative agreement with the theoretical data from a two-dimensional fluid-plasma-model [22], developed for description of the same inductively-driven discharge. However, at the current stage a detailed comparison between theory and experiment can not be done. The simulation describes the inductive mode of the discharge operation, considered as appropriate for efficient negative hydrogen ions production, whereas the current experiments, for technical reasons, have been carried out in the capacitive mode. Since the discharge behaviour significantly changes with the operation mode, different values and dependencies of plasma parameters could be expected. A modification of the experimental set up and investigation of the inductive mode of the discharge operation are planned.

Figure 16 presents results for the correlation between the atomic temperature \( T_a \) and the \( n_{th}/n_f \) ratio at axial position \( z = 2 \) cm. The obtained temperature growth can be connected with the increase of the fast atoms fraction. By increasing the pressure and the applied power, the fast atoms fraction decreases and the dependence of \( T_a \) on \( n_{th}/n_f \) generally becomes smoother. Taking away from the coil the fast atoms fraction become higher which results in higher temperatures of the thermalized atoms. At the same time, the variations of the pressure and the applied power lead to smaller changes in the ratio between the thermalized and the fast atoms densities.

6. Conclusions

An interferometric method, previously developed in [12], is applied for diagnostics of neutrals in a planar coil inductive discharge in hydrogen. The method is based on analysis of the \( \text{H}_\alpha \) spectral line profile, recorded under various experimental conditions. The emission is found to come from two distinct populations of the excited \( n = 3 \) atomic state. The first one is characterized with a temperature which is assumed to be equal to the temperature of thermalized atomic hydrogen in its ground state, whereas the second one is a group of nearly monoenergetic fast atoms with a mean energy of about 8 eV. The established correlation between the atomic temperature and
the ratio between the thermalized and the fast atoms densities in \( n = 3 \) state suggests that the reactions responsible for production of fast hydrogen atoms may play an important role in theoretical models and simulations. The molecular temperature is obtained from analysis of the intensity distribution over the Fulcher-\( \alpha \) rovibrational bands. The experimental results show that the temperature of the hydrogen atoms is about two times higher than that of hydrogen molecules. The values of the atomic and the molecular temperatures are not always correlated and their dependence on the gas pressure, applied power and axial position are different. Currently a collisional-radiative model, describing the same inductive discharge and aiming at determination, among others, of the electron temperature and the electron density, is under development and the temperatures obtained in this study will be used as input parameters. Further experimental work is also needed in order to verify the equivalence of the temperatures of hydrogen atoms in the ground and the excited states. This is assumed in the current study but may be questionable, especially at different experimental conditions. The verification can be done by simultaneous study of several Balmer lines, e.g. \( \text{H}_\alpha \), \( \text{H}_\beta \) and \( \text{H}_\gamma \). The extension of the possibilities of the developed interferometric setup will make it even more attractive for diagnostics of different hydrogen discharges.

7. Acknowledgments

This work has been carried out within the framework of the EUROfusion Consortium and has received funding from the European Union’s Horizon 2020 research and innovation programme under grant agreement number 633053. The views and opinions expressed herein do not necessary reflect those of the European Commission. The authors acknowledge partial support from the Sofia University Grant No 58/2015.

References


