Spectroscopic temperature measurements in hydrogen inductively-driven plasmas at low pressures

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Abstract. Optical emission spectroscopy is applied to obtain the rotational and gas temperatures of an inductively-driven tandem plasma source of negative hydrogen ions. Discharges are maintained in hydrogen at fixed frequency $f = 27$ MHz, applied power $P = 500$ W, and low gas pressure in the range $p = (26 – 60)$ mTorr. The rotational and gas temperatures are determined by analyzing the intensity distribution of the H$_2$ Fulcher-$\alpha$ band ($d^3\Pi_u \rightarrow a^3\Sigma_g^+$) line emission.

1. Introduction

Radio frequency (rf) inductively-driven discharges are involved in plasma processing technologies [1] and in negative hydrogen ion beam production for fusion experiments [2]. In the latter case, the construction of the discharge chamber, known as a tandem plasma source, leads to separation of the discharge vessel into two regions: a driver, where the discharge is produced, and a process chamber, directly connected to the driver, where the plasma exists due to its expansion. This separation provides hydrogen molecules in highly vibrationally excited states in the first chamber that can greatly enhance the production of negative ions, mainly through dissociative attachment of slow plasma electrons in the second chamber. The kinetics of hydrogen vibrationally excited molecules is of great interest in understanding the processes of H$^-$ formation in a volume [3].

This paper deals with the use of optical emission spectroscopy (OES) for gaining information about species and plasma parameters in an inductively-driven tandem plasma source in H$_2$ at low pressure. In order to obtain the gas temperature, the rotational temperature of the excited state H$_2(d^3\Pi_u)$ is determined by analyzing the intensity distribution of the spectral lines of the Fulcher-$\alpha$ system of H$_2$. Usually the rotational temperature is assumed to coincide with the translational gas temperature [4]. However, since in low pressure plasmas the radiative lifetimes of the molecules in the electronically excited states are shorter than the characteristic time of rotational relaxation [5,6], the relation between rotational and gas temperature requires special attention.

2. Experimental set-up

The plasma is generated by an rf inductively-driven discharge. Figure 1 presents the scheme of the experimental set-up which includes: a vacuum vessel, a high-frequency generator and a matching device. The vacuum vessel has two parts: gas discharge dielectric chamber where the plasma is produced (driver region), and a metal chamber providing space for the plasma expansion (expansion region). A nine-turn copper coil is positioned tightly over a quartz cylindrical tube with internal and
external diameters 4.5 cm and 4.9 cm, respectively, and length of 30 cm. The effect of the electrostatic field is minimized by a Faraday shield. The plasma produced in the gas discharge tube expands and fills the metal cylindrical stainless steel chamber with internal and external diameters 22 cm and 23 cm, respectively, and length of 47 cm. The discharges are maintained at fixed high-frequency $f = 27$ MHz and applied power $P = 500$ W in H$_2$ at gas pressures $p = 26, 40$ and 60 mTorr.

![Figure 1. Scheme of the experimental set-up.](image1)

![Figure 2. Arrangements for the OES diagnostics.](image2)

The hydrogen emission spectra from the driver and the plasma expansion region are detected using a portable Ocean Optics HR4000 spectrometer with 25 μm entrance slit. This spectrometer system, shown in figure 2(a), provides fast detection in wide wavelength range $\lambda = (200 – 1100)$ nm. The integration time of the CCD line is $t_{int} = 3.8$ ms and $t_{int} = 250$ ms, for the driver and the plasma expansion region, respectively.

Registration with higher resolution of the H$_2$ Fulcher-α molecular band is achieved with the second spectrometer system shown in figure 2(b), by using an 0.6 m focal length Fasti MDR-2 (Lomo-Russia) monochromator, equipped with a PMT-79 photomultiplier and a grating with 1200 grooves/mm for detection in $\lambda = (400 – 900)$ nm. The slits (entrance and exit) of the monochromator are fixed at 40 μm assuring wavelength resolution of about 0.1 nm. A PC provides the wavelength tuning and calibration, spectra observation in real time and data acquisition. Lock-in amplifier detection is used for measuring the light signal. Spectral responses of the optical and detecting system are tracked by using a tungsten-ribbon lamp.

3. Results and discussion

3.1. Spectra observations

Hydrogen emission spectra taken from the driver near the end of the coil and at a distance of 12 cm from the beginning of the plasma expansion regions are recorded over $\lambda = (200 – 1100)$ nm, at $p = 26, 40$ and 60 mTorr and rf applied power $P = 500$ W. Besides intensive H$_2$ molecular bands, the atomic H$_a$, H$_b$ and H$_c$ spectral lines are also observed in the spectra (figure 3). In the driver region the ratio of the hydrogen H$_a$ line intensity compared to molecular lines of H$_2$ Fulcher-α band is several tens, whereas in the plasma expansion region this ratio decreases to several times. The ratio between the atomic line intensities in the two chambers is of the same order. These observations indicate that the dissociation degree changes from the driver to plasma expansion region. The radiation of molecular bands provides information for the excitation processes as well as for the vibrational and rotational populations of the excited states. As it is known, the Fulcher-α radiative transition ($d^3\Pi_u \rightarrow a^5\Sigma^+_g$) is suitable for diagnostics of H$_2$ [7]. This band is intense and can be identified clearly over $\lambda = (600 – 625)$ nm. The rotational lines observed (figure 4) belonging to the $Q$-branch of the diagonal vibrational states of (0-0), (1-1) and (2-2) transitions in the Fulcher-α spectrum are well separated.
Figure 3. Emission spectra of hydrogen inductive discharge, from driver (in (a)) and plasma expansion (in (b)) regions, at $P = 500$ W and $p = 40$ mTorr.

Figure 4. Observed $\text{H}_2$ Fulcher-α rovibrational band, from driver region, at $P = 500$ W and $p = 60$ mTorr. $Q$-branch lines are labelled.

3.2. Determination of rotational and gas temperatures

Analysis of the relative intensity distribution of the rotational spectral lines of electronically-excited molecules is widely used as a method for determination of rotational excitation temperature $T_{rot}^\ast$ assuming Boltzmann law excitation distribution. The experimentally measured line intensities $I_{\nu \nu' \nu''}$ between the rotational levels of the excited electronic-vibrational (vibronic) states are directly linked to $T_{rot}^\ast$ through the relation: 

$$\ln(\lambda^4 \cdot I_{\nu \nu' \nu''} / S_{\nu \nu' \nu''}) = -(B_{\nu}J'(J'+1)hc)/(kT_{rot}^\ast) + \text{const},$$

where $S_{\nu \nu' \nu''}$ is the
line strength, $\lambda$ and $B_\nu$ are the wavelength and the rotational constant of the excited state, $J'$ is the rotational quantum number, $h$ and $k$ are the Planck and Boltzmann constants and $c$ is the light velocity. The line strength is calculated according to Hönl-London formula: $S_{J,J'} = 0.5(2J+1)(2J'+1)$ with $t = 0$ for even $J'$-values and $t = 1$ for odd $J'$-values. The plot of $\ln(\lambda^4 \cdot I_{J,J'/J,J''} / S_{J,J'})$ versus $B_\nu J'(J'+1)$ gives the so-called Boltzmann plot. The rotational temperature $T_{rot}^0$ can be obtained from the slope of this plot. If the characteristic time of molecular rotational relaxation $\tau_{rel}$ is much shorter than the radiative lifetime $\tau_{rad}$ and that of the kinetic processes of redistribution of rotational excitations, the rotational temperature is equal to the translational gas temperature. However, in low pressure plasmas $\tau_{rad} \ll \tau_{rel}$ is possible in the case of direct electron excitation of ground molecular state H$_2$($\chi^3\Sigma_g^+$) to rotational levels of excited vibronic states followed by spontaneous decay to a lower excited vibronic state. The calculations indicate that at gas pressures $p = (10 - 75)$ mTorr and gas temperatures $(300 - 900)$ K, the characteristic time between heavy particle collisions being of the order of $10^{-7}$ s (the total cross section $\sigma = 2.3 \times 10^{-18}$ m$^2$ for the collisions H$_2$ - H$_2$(d$^3\Pi_u$) is taken from [8]) is much longer than the radiation lifetime $\tau_{rad} = 31$ ns [8] of hydrogen molecules in the H$_2$(d$^3\Pi_u$) state. In that case a Boltzmann rotational distribution in the ground electronic state of the molecules images a Boltzmann rotational distribution in the excited electronic states [5,6] and the relation between rotational temperature of the ground $T_{rot}^0$ and excited $T_{rot}^*$ state is given by: $T_{rot}^0 / T_{rot}^* = B^0 / B^*$, where $B^0$ and $B^*$ are the rotational constant of the ground and excited state, respectively. The gas temperature $T_g = T_{rot}^0$ and since $B^0 = 60.809$ cm$^{-1}$ [6] and $B^* = 30.364$ cm$^{-1}$ [6], $T_g = 2T_{rot}^*$.

To determine the rotational and gas temperatures in an inductively-driven discharge in hydrogen in the driver and the plasma expansion region, the line intensities of $Q_1$ to $Q_4$, belonging to the $Q$-branch of the first three diagonal vibrational state of (0-0), (1-1) and (2-2) transitions in the Fulcher-\(\alpha\) spectrum are measured. The Boltzmann plots for these series presented in figure 5 in the driver and the plasma expansion region are for gas discharge conditions $p = 60$ mTorr, $f = 27$ MHz and $P = 500$ W. The plots give linear fits indicating that the rotational level populations of H$_2$(d$^3\Pi_u$) state follow a Boltzmann distribution. The rotational temperatures obtained from the different diagonal vibrational state transitions coincide quite well. The relative error in $T_{rot}^*$ determination is smaller than 20%. The gas temperature dependence on the gas pressure obtained in the driver and the plasma expansion region is presented in figure 6. The experimental data for the gas temperature will be used in further kinetic modelling of H$_2$ plasmas.

![Figure 5. Boltzmann plot of the first lines of $Q$-branch in the (0-0)(■), (1-1)(●) and (2-2)(▲) rotational bands of the Fulcher-\(\alpha\) system of H$_2$, driver region (full lines), expansion region (dash lines), $P = 500$ W and $p = 60$ mTorr.](image-url)
4. Conclusions
An optical emission spectroscopy technique for determination of rotational and gas temperatures is applied to inductively-driven hydrogen discharges. The technique is based on the analysis of relative intensity distribution of the rotational spectral lines of the Fulcher-α system of H₂.

Acknowledgements
The author wishes to thank Prof. Dr. I. Koleva and Prof. Dr. A. Shivarova for the useful discussions. This work is within the programme of the Bulgarian Association EURATOM/INRNE (task P2), DFG-project 436 BUL 113/144/0-1 and project D01-413 supported by the National Science Fund (Bulgaria).

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