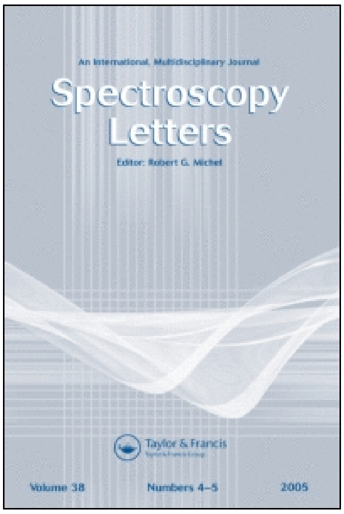

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Hydrogen Degree of Dissociation in a Low Pressure Tandem Plasma Source

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ABSTRACT Hydrogen dissociation degree in an inductively-driven tandem plasma source, operating at low pressure, is measured by applying optical actinometry method with an argon gas as actinometer. The gas temperature, a parameter in the investigations, is obtained from the rotational temperature, analyzing the intensity distribution of the Fulcher-α band. Several actinometric pairs are used for examining the dissociation degree and its pressure dependence. The influence—on the results—of the difference in the excitation cross sections for argon, commonly accepted, is analyzed. Two suitable actinometric pairs are proposed, one of which can also be applied for fast monitoring of the dissociation degree.

KEYWORDS actinometry, degree of dissociation, emission spectroscopy, gas temperature, inductively-driven discharge, tandem plasma source

INTRODUCTION

Radio frequency (rf) inductively driven discharges in molecular gases at low pressure are widely used in the field of plasma technology,[1] for a variety of plasma processes. They have been also deeply involved recently into the problem of the neutral beam injection for plasma heating in fusion reactors.[2] The development of inductively driven tandem plasma source for negative hydrogen ions (H^-) was stimulated since the volume production[3] of H^- generation has been discovered. This source consists of two regions (chambers) with high- and low-energetic electrons, separated by a magnetic filter. In the first region high vibrationally excited hydrogen molecules are created, whereas dissociative attachment of slow plasma electrons to the excited molecules is ensured in the second region. Optimization of the discharge conditions in the first chamber of tandem plasma sources for H^- production in the second chamber is important task. In this aspect the hydrogen atom density in the ground states and respectively dissociation degree of molecules are crucial.

Among the variety of diagnostic methods, the spectroscopy of emission induced by plasma is widespread method,[4,5] due to its no interference in the plasma behavior, easy implementation and ensuring enough accuracy. In spite of all advantages of optical emission spectroscopy methods, a common disadvantage is that they can be used to measure particle concentration directly only in the excited states. With optical actinometry method is

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possible to determine, the particle concentration in their ground states (molecules, atoms and radicals). The actinometry is based on seeding the plasma with a small, known amount of noble gas (actinometer) and monitoring the noble gas emissions concurrently with those of the reactive particles. This method, firstly proposed by Coburn and Chen[6] is continuously developed,[7–10] critically reviewed[11,12] and extended lately[13,14] for obtaining electron temperature (T_e) and electron energy distribution function (EEDF).

The study is focused on optical emission spectroscopy diagnostics of a low pressure inductively driven tandem plasma source. The goal of investigation is determination of hydrogen atom ground state density and degree of dissociation, under various gas discharge conditions, by using actinometry method. With respect to optimization of the negative hydrogen ion production, improvement of the methods for tracking the neutral component densities in the plasma is of current interest[15,16] in the literature for these type of sources.

EXPERIMENTS

Experimental Setup

The plasma studied is of a rf inductively-driven discharge in tandem type plasma source. The experimental set-up (Fig. 1) is as described by Kiss'ovski et al.[17,18] The vacuum vessel consists of two parts: a dielectric chamber where the plasma is produced (a driver region) and a metal chamber providing space for plasma expansion (expansion region). A nine turn copper coil is positioned tightly over the quartz cylindrical tube, which is with internal and external diameters 4.5 cm and 4.9 cm, respectively, and length of 25 cm. A metal cylinder around the driver ensures its electromagnetic shielding. The plasma produced in the gas discharge tube expands and fills the second chamber of the source: a metal stainless steel cylinder with internal diameter 22 cm and length of 47 cm. In the experiment, the discharge is maintained at a frequency of $f = 27$ MHz and power of $P = 700$ W in a H_2:Ar gas mixture at pressures $p = 0.8$, 1.1, 1.3, 3.3, and 5.3 Pa. The magnetic filter is inserted in the expanding plasma chamber (Fig. 1) and the value of field is 100 G. The probe measurements included in the study are performed by a Smart-Probe™ system (Scientific systems, Dublin, Ireland) with a probe movable in the axial direction.

The emission spectra from the driver region are detected by using Ocean Optics HR4000 spectrometer with a 25 μm entrance slit. The spectrometer system shown in Fig. 2(a) provides real time CCD detection in a wavelength range λ of 200 to 1100 nm. Registration of H_2 Fulcher-α molecular band as well as of the atomic hydrogen and argon lines with higher resolution are achieved with the second spectrometer system shown in Fig. 2(b), by using a 0.6 m focal length Fasti MDR-2-23 (Lomo-Russia) monochromator, equipped with a PMT-79 and a grating with 1200 grooves/mm for detection in the region $\lambda = (400–900)$ nm. The entrance and exit slits of the monochromator are fixed at 50 μm assuring wavelength resolution of about 0.1 nm. A PC provides the wavelength tuning and calibration, spectra observation and data acquisitions. Lock-in amplifier

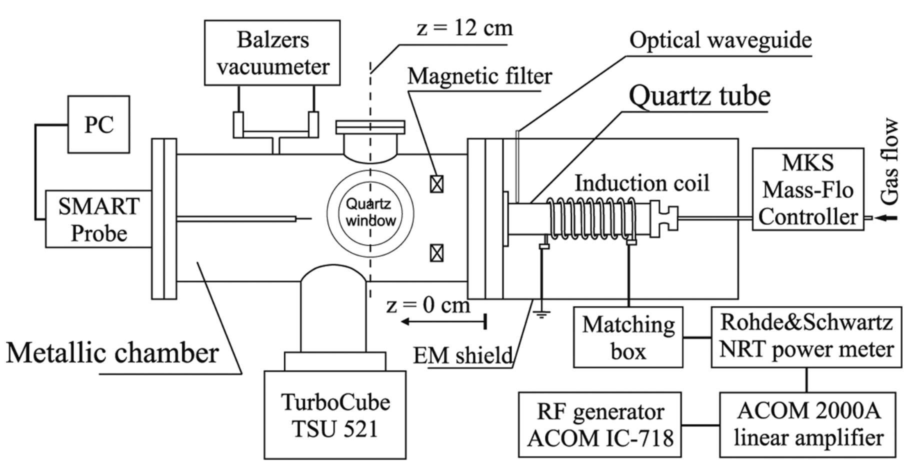
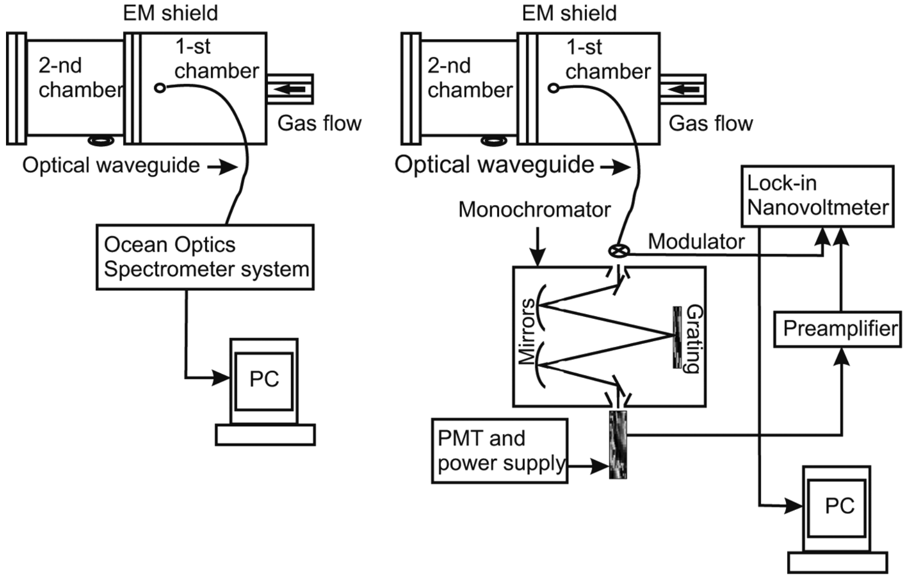


FIGURE 1 Scheme of the experimental set-up.



(a) (b)

FIGURE 2 **Arrangements for the optical emission spectroscopy diagnostics.**

detection is used for measuring the light signal. The spectral detection efficiency is calibrated with a standard tungsten-ribbon lamp. The ambient gas is hydrogen with a high purity (99.99%). Argon gas is added as an actinometer. In all the experiments the maintained flow rates of hydrogen and argon 58.58 sccm and 1.41 sccm, respectively, ensure 2.4% Ar gas admixture.

Analysis of Optical Emission Spectra

Emission spectra of H_2 and H_2:Ar gas mixture taken from the driver region near the end of the inductive coil, are recorded over $\lambda = (350–850)$ nm. The spectra analyses show that the ratios of the atomic line intensities as well as the ratios of atomic to molecular line intensities are of the same order, in both cases (H_2 discharge and H_2:Ar discharge). This implies that the value of selected argon gas admixture does not influence the conditions of hydrogen discharge maintenance. In H_2:Ar discharges (Fig. 3), besides intensive H_2 bands, the atomic H_α, H_β, H_γ and the argon lines with $\lambda = 750.4$ nm and $\lambda = 811.5$ nm are clearly observed. The Balmer line intensities are linearly sensitive to the changes of the discharge-operation conditions, whereas the argon line intensities remain almost the same.

The molecular spectra provide information for the excitation processes as well as for the vibrational and rotational populations of the excited states. As it is known, Fulcher-α radiative transition ($d^3\Pi_u \rightarrow a^3\Sigma_g^+$)

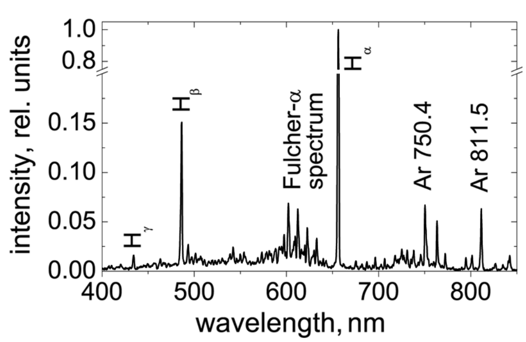


FIGURE 3 **Emission spectra of H_2:Ar inductive discharge at $p = 1.1$ Pa.**

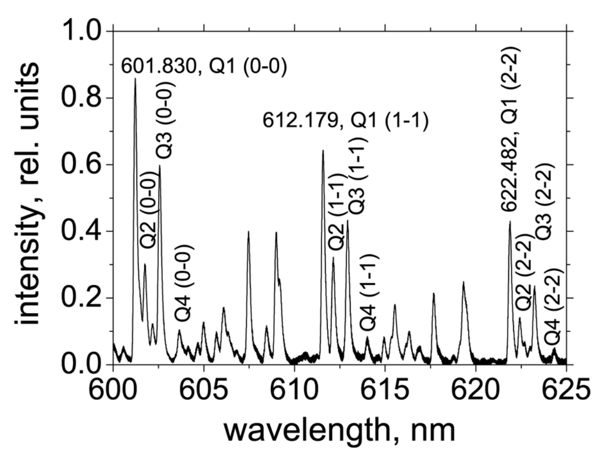


FIGURE 4 **Hydrogen Fulcher-α rovibrational band at $p = 5.3$ Pa. The Q-branch lines are labeled.**

is suitable for diagnostics of H_2 discharges, since this band is intense and can be identified clearly over $\lambda = (600–625)$ nm. The observed rotational lines (Fig. 4) belonging to the Q-branch of the diagonal states of the (0-0), (1-1), and (2-2) transitions in the Fulcher-α spectrum are well separated.

IMPLEMENTED METHODS

Optical Actinometry Method

The actinometry is a straightforward technique for determining concentration of neutral species in the plasma, but certain conditions must be satisfied in order to obtain reliable results. If the emitting states of the actinometer and of the examined gas are excited by direct electron impact from the ground state and deactivates spontaneously, and if they have similar excitation potentials, then the same group of electrons will take part in populating these states. Consequently, the rate constants of excitation of the emitting states should depend in the same manner on the plasma parameters. As a result, the density of the hydrogen atoms in the ground state N_H is determined from the corresponding spectral line intensity ratio I_H/I_{Ar} of excited hydrogen (j) and argon (i) states, and the density of argon atoms N_{Ar}, which is known:

$$N_H = K\frac{I_H}{I_{Ar}}N_{Ar}, \quad (1)$$

where $K = (\sigma \cdot A \cdot \tau \cdot \nu \cdot S)_{Ari}/(\sigma \cdot A \cdot \tau \cdot \nu \cdot S)_{Hj}$ and presented as $K = (\sigma^{\max} \cdot C \cdot A \cdot \tau \cdot \nu \cdot S)_{Ari}/(\sigma^{\max} \cdot C \cdot A \cdot \tau \cdot \nu \cdot S)_{Hj}$ is a constant that depends only on the properties of the specific atoms, A are radiative transition probabilities of the respective spectral lines with frequencies ν, generated by transition from the upper excited states with radiative lifetimes τ, σ are the electron impact excitation cross sections with maximum values $\sigma_{Ari}^{\max}$ and $\sigma_{Hj}^{\max}$, S contains spectral and geometrical factors of the detection system, C is related to the behavior of the cross sections, near to the threshold region. In this way the method of actinometry is very attractive for fast monitoring of N_H and consequently dissociation degree.

In all cases, when the contributions of the dissociative channel, quenching by heavy particle collisions as well as other processes, taking place in the population kinetics of the excited states, the ratio N_H/N_{Ar} depends nonlinearly on I_H/I_{Ar}.

To obtain hydrogen atom density in the first chamber of tandem plasma source by the method of actinometry, the contribution of the molecules (dissociation excitation and deexcitation by heavy particle collisions) in the excited state kinetic balance is included. That requires EEDF and T_e to be known.

The kinetic processes, considered in the study, are given in Table 1. In this case the stationary rate balance equations for the excited hydrogen and argon states, N_{Hj} and N_{Ari} are expressed as:

$$\frac{dN_{Hj}}{dt} = n_e N_H \alpha_H^{dir} + n_e N_{H_2} \alpha_H^{diss} - N_{Hj}\tau_{Hj}^{-1} - N_{Hj} N_{H_2} q_{H_2}^{H} = 0, \quad (2)$$

$$\frac{dN_{Ari}}{dt} = n_e N_{Ar} \alpha_{Ar}^{dir} - N_{Ari}\tau_{Ari}^{-1} - N_{Ari} N_{H_2} q_{H_2}^{Ar} = 0, \quad (3)$$

where α^{dir} and α^{diss} are the rate coefficients for direct and dissociative excitation; $q_{H_2}^{H}$, $q_{H_2}^{Ar}$ are the quenching coefficients for the excited hydrogen and argon levels, by H_2 molecules and N_{H_2} is the density of hydrogen molecules. From the expressions (2) and (3), the density of hydrogen atoms is deduced as:

$$N_H = N_{Ar}\frac{I_H S(\lambda_{Ar})\alpha_{Ar}^{dir} A_{Ar}\lambda_H \tau_{Ari}}{I_{Ar} S(\lambda_H)\alpha_H^{dir} A_H \lambda_{Ar}\tau_{Hi}} \times \left(1 + \frac{N_{H_2}\alpha_H^{diss}}{N_H \alpha_H^{dir}}\right)^{-1} \times \frac{\tau_{Hj}^{-1} + N_{H_2} q_{H_2}^{H}}{\tau_{Ari}^{-1} + N_{H_2} q_{H_2}^{Ar}}. \quad (4)$$

This form of the equation has clear physical meaning. The first term is similar to expression (1) as the

TABLE 1 Kinetic Processes, Cross Sections σ, Rate Coefficients α, Einstein Coefficients A, Radiative Lifetimes τ, and Corresponding References

No	Kinetic processes	Reference
Direct excitation:		
1	$H + e \rightarrow H^*(n = 3, 4, 5) + e$	σ[19,20]
2	$Ar + e \rightarrow Ar^*(4p) + e$	σ[21,22,23]
Radiative decay:		
3	$H^*(n = 3, 4, 5) \rightarrow H^*(n = 2) + h\nu$	A, τ[24]
4	$Ar^*(4p) \rightarrow Ar^*(4s) + h\nu$	A, τ[25]
Dissociative electron excitation of H_2:		
5	$H_2 + e \rightarrow H^*(n = 3, 4, 5) + H(n = 1) + e$	σ[19,26]
Quenching of excited atoms:		
6	$H^*(n = 3, 4, 5) + H_2 \rightarrow H + H_2 + \Delta E$	σ[27]
7	$Ar^*(4p) + H_2 \rightarrow Ar + H_2 + \Delta E$	σ[27]

cross sections are replaced by the rate coefficients. The second term accounts for the relative contribution of dissociative excitation with respect to those of direct excitation. The third term reveals to the process of quenching of excited hydrogen or argon atoms by H_2 molecules. The references used for cross-sections, radiative transition probabilities, lifetimes of excited states as well as for quenching coefficients data are also given in Table 1. The quenching coefficient of $2p_9$ (Paschen notation) argon excited level (spectral line $\lambda = 811.5\,\text{nm}$) by H_2 is unknown and thus is not taken into account.

Then dissociation degree D of the hydrogen molecules in the plasma is given by the relationship:

$$D = \frac{N_H}{N_H + N_{H_2}}. \tag{5}$$

Determination of the Gas and Electron Temperatures

In order to obtain the gas temperature (T_g), the rotational temperature of the excited state $H_2(d^3\Pi_u)$ is determined by analyzing the intensity distribution of the spectral lines belonging to the hydrogen Fulcher-α band. Usually the rotational temperature is assumed to coincide with the translational gas temperature.[28] When the radiative lifetimes of some excited electronic-vibrational (vibronic) states is shorter than the characteristic time of the rotational relaxation, especially in low pressure plasmas, then the relation between rotational and gas temperatures requires special attention. The method applied in the study is described in details by Iordanova[29] and involves the assumption that under the given gas-discharge conditions the hydrogen molecular state $H_2(d^3\Pi_u)$ is excited from the ground molecular state $H_2(X^1\Sigma_g^+)$ by electron impact and decays to $H_2(a^3\Sigma_g^+)$ spontaneously. The calculations indicate that the characteristic time between the heavy particle collisions ($H_2(X^1\Sigma_g^+) - H_2(d^3\Pi_u)$), being of the order of $10^{-7}\,\text{s}$ (with a total cross section $\sigma = 2.3 \times 10^{-18}\,\text{m}^2$[28]) is much longer than the radiation lifetime $\tau_{rad} = 31\,\text{ns}$[30] of the hydrogen molecules in the $H_2(d^3\Pi_u)$ state. Then the Boltzmann rotational distribution in the ground electronic state images to a Boltzmann rotational distribution in the excited electronic states[31,32] and the relation between the rotational temperatures of the ground T^0_{rot} and excited T^*_{rot} states is expresses as $T^0_{rot}/T^*_{rot} = B^0/B'$ where $B^0 = 60.809\,\text{cm}^{-1}$ and $B' = 30.364\,\text{cm}^{-1}$ are rotational constants of the ground and excited state, respectively. That gives $T_g = T^0_{rot} = 2T^*_{rot}$.

For determining the rotational and gas temperatures of H_2 plasmas in the driver region, the line intensities of Q1 to Q4 belonging to the Q-branch of the first three diagonal vibrational state of the (0−0), (1−1), and (2−2) transitions in the Fulcher-α spectrum (Fig. 4) have been measured. The Boltzmann plots for these series, obtained at $p = 5.3\,\text{Pa}$ and $P = 700\,\text{W}$ are presented in Fig. 5. The rotational temperatures obtained from the different diagonal vibrational state transitions coincide quite well. The relative uncertainties in the determination of T^*_{rot} are smaller than 20%. The obtained dependence of the gas temperature on the gas pressure is presented in Fig. 6.

The data for T_e, carried out from probe diagnostics in the driver region of the plasma source, correspond well to those determined at similar plasma sources.[16,33,34] The probe is positioned on the discharge axis at the axial position of the cross section from which the spectra emission is collected. The electron temperature is determined from the transition region of the probe characteristics as it is described in.[35,36] The obtained pressure dependence of the electron temperature is given in Fig. 6. The probe measurements indicate also that in the studied gas pressure range, the EEDF could be approximated with a Maxwellian distribution.

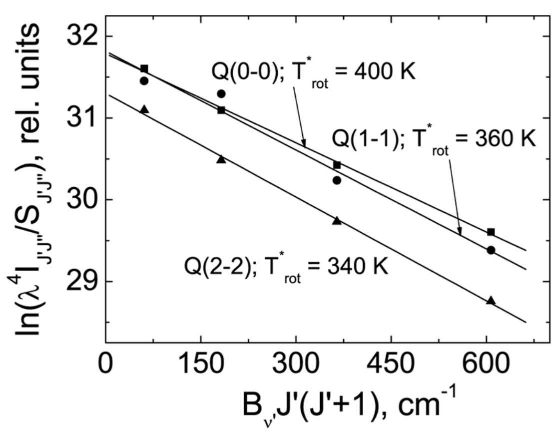


FIGURE 5 Boltzmann plot of the first lines of the Q-branch in the (0–0)(■), (1–1)(●), and (2–2)(▲) rotational bands of the Fulcher-α system of H_2. Experimental conditions: driver region, $p = 5.3\,\text{Pa}$.

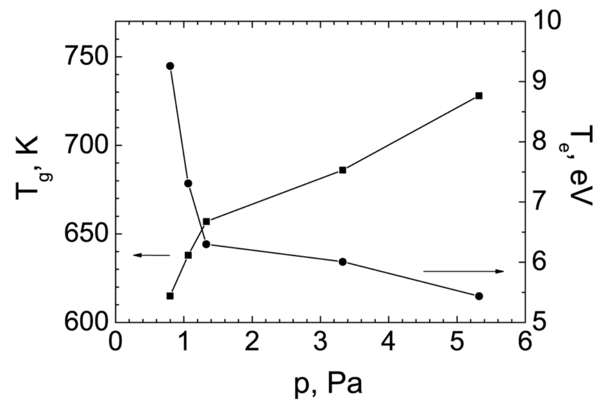


FIGURE 6 Temperature dependences on the gas pressure.

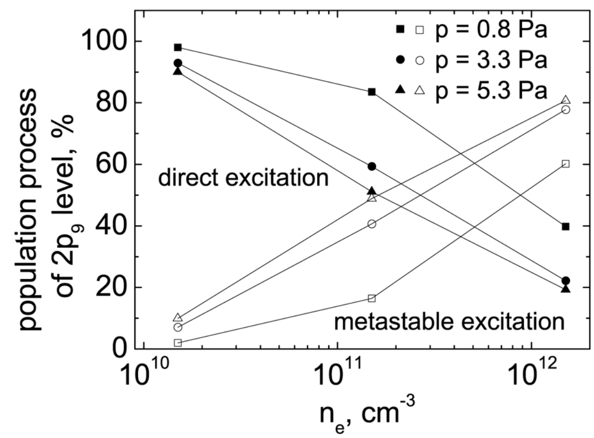


FIGURE 7 Dependence of direct (full symbol) and step (open symbol) excitations of $2p_9$ level on electron density in the pressure range $p = (0.8 - 5.3)$ Pa.

RESULTS FOR HYDROGEN ATOM DENSITY AND DEGREE OF DISSOCIATION

The results for the density of ground states hydrogen atoms N_H as well as for the degree of dissociation D are obtained as a solution of Eq. (4) employing four actinometric pairs (H_α, Ar_{750}), (H_α, Ar_{811}), (H_β, Ar_{750}), (H_γ, Ar_{750}), commonly used in the literature (Table 2). Emission cross sections of direct and dissociative excitation of the H_α and H_β lines by electron impact, with account for the fine structure of hydrogen atom levels,[19] and for direct[20] and dissociative[26] excitation of the H_γ line are used. The excitation cross sections of the argon lines are those given in the literature.[21–23] According to our previous investigations[37,38] the argon excited $2p_1$ and $2p_9$ (Pashen notation) levels (Table 2) are mainly populated by direct and metastable electron excitation, respectively. The contribution of population processes of $2p_9$ level is estimated and presented in Fig. 7 in the typical for inductively driven hydrogen discharges electron density range ($n_e = (10^{10} - 10^{12})\,cm^{-3}$). Since, according to probe measurements the electron density in the driver region of plasma source is of the order of $10^{10}\,cm^{-3}$, the contribution of metastable pooling of $2p_9$ level even at the highest pressure is less then 10% thus it is neglected in the modelling.

The contribution of the processes listed in Table 1 is examined in the calculations of N_H and D by using the excitation cross sections for the argon lines.[21] Fig. 8 shows the density of the hydrogen atoms in the ground state (dash curves), calculated taking into account the processes ((1) to (5)) in Table 1. At the same figure, with full curves, are presented the results obtained by accounting only the direct excitation and radiative decay (Table 1, processes (1) to (4)) of the hydrogen and argon atoms. In accordance with Eq. (4) the dissociative excitation leads to a reduction of the ground state hydrogen atom density.

TABLE 2 Spectral Data Relevant to the Investigated Emission Lines

Emission line	H_α	H_β	H_γ	Ar_{750}	Ar_{811}
Upper state (u)	$n=3$	$n=4$	$n=5$	$3p^54p2p_1$	$3p^54p2p_9$
Lower state (l)	$n'=2$	$n'=2$	$n'=2$	$3p^54s1s_2$	$3p^54s1s_5$
E_u (eV)	12.08	12.75	13.05	13.48	13.08
E_l (eV)	10.20	10.20	10.20	11.82	11.55
Wavelength (nm)	656.3	486.1	434.0	750.4	811.5

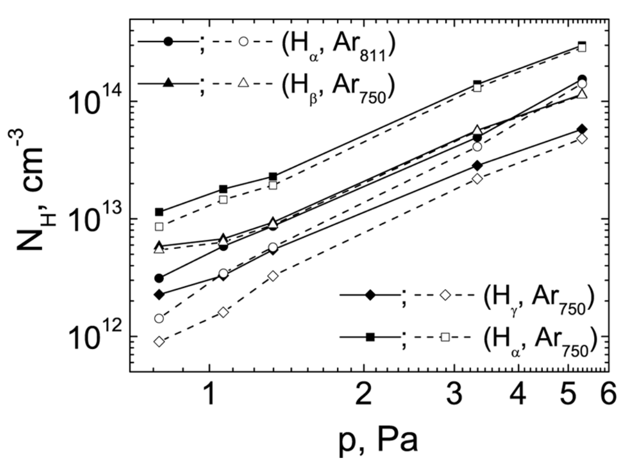


FIGURE 8 Dependence of density of hydrogen atoms in ground state on pressure.

The effect is pronounced at pressure $p < 1.3$ Pa especially in the results obtained from for the pairs (H_α, Ar_{811}) and (H_γ, Ar_{750}), whereas at $p > 3$ Pa, for the whole actinometric pairs, the contribution of dissociative excitation is negligible with respect to those of direct excitation. The obtained pressure dependence of the influence of the dissociative excitation is also according to equation (4, second term) since, (as it is shown in Fig. 9), the impact frequency ratios of dissociative to direct excitations depends strongly on the pressure and have higher values at low gas pressure. The results of N_H, obtained through the line intensity ratios $I(H_\alpha)/I(Ar_{811})$ and $I(H_\beta)/I(Ar_{750})$ coincide quite well, whereas those from $I(H_\alpha)/I(Ar_{750})$ and $I(H_\gamma)/I(Ar_{750})$ give, respectively, at about two times higher/lower values.

Dependence of the dissociation degree on the pressure, presented in Fig. 10, is calculated with the complete set of processes (1) to (7) in Table 1 (open symbols) and without (full symbols) accounting for the quenching of the excited hydrogen and argon atoms by H_2 (processes (1)-(5) in Table 1). The results reveal that the contribution of the quenching process is negligible in the pressure range studied. The degree of dissociation depends linearly upon the pressure, and it changes from 3% (at $p = 0.8$ Pa) to 20% (at $p = 5.3$ Pa), as it follows from the results obtained through the actinometic pairs (H_α, Ar_{811}), (H_β, Ar_{750}).

The next step in the study is to obtain an actinometric pair suitable (not depending on the EEDF and T_e) for fast monitoring. The results for degree of dissociation D^* (obtained according to expression (1)) and those D (obtained according to expression

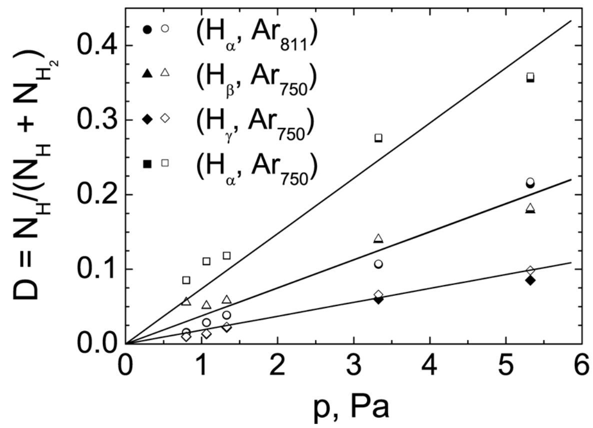


FIGURE 10 **Dependence of dissociation degree on pressure.**

(4)) are compared through their ratio (Fig. 11). The variation of the D^*/D value at lower pressure is due to the influence of the dissociative excitation on the excited hydrogen atom population, taken into account in expression (4). The results reveal that the ratio D^*/D depends strongly of the actinometric pair choice and the requirements for using the actinometry method in its simplified form are fulfilled—in the pressure range studied—for the spectral pair (H_β, Ar_{750}).

For checking the influence over the results for N_H, D and D^*/D cross section, caused from the discrepancy of the Ar cross section data, calculations are also conducted. The results, obtained with the line intensity ratios $I(H_\alpha)/I(Ar_{811})$ and $I(H_\beta)/I(Ar_{750})$, are in agreement with those, presented in figure 10, but with values of D higher up to 1.3 times ($D \approx 4\%$ at $p = 0.8$ Pa, $D \approx 24\%$ at $p = 5.3$ Pa). As should be expected the choice of data for the cross sections is

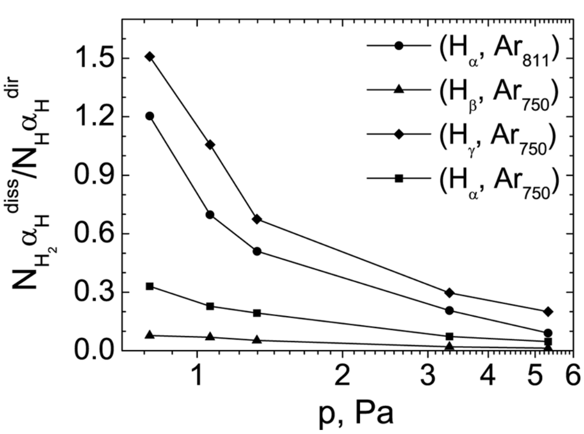


FIGURE 9 **Dependence on pressure of the impact frequency ratios of dissociative to direct excitations.**

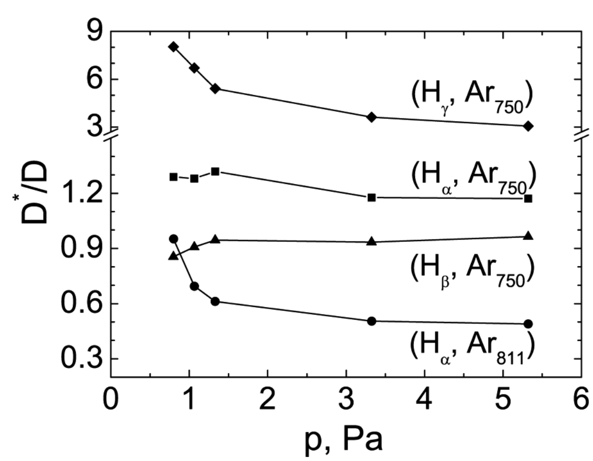


FIGURE 11 **Dependence on pressure of ratios D^*/D.**

crucial for finding a suitable pair for fast actinometric monitoring. With the use of cross sections for argon lines[22,23] such pairs could not be extracted.

CONCLUSIONS

The degree of dissociation in the driver region of tandem plasma source is measured by applying the method of optical actinometry. The gas temperature, an essential parameter in the investigations, is obtained from the rotational temperature, reflecting the rotational level distribution of the electronically excited state $H_2(d^3\Pi_u)$. The gas temperature in the driver region is twice higher than the rotational temperature. The actinometry method is applied with 2.4% argon gas admixture, which does not disturb the excitation kinetics of hydrogen plasma. Four actinometric pairs (H_α, Ar_{811}), (H_β, Ar_{750}), (H_γ, Ar_{750}) and (H_α, Ar_{750}) are examined. The results obtained by including extended kinetics in the balance equations of the excited states imply that the contribution of the dissociative excitation of atomic hydrogen levels is pronounced and in the same order for actinometric pairs (H_α, Ar_{811}) and (H_γ, Ar_{750}), at lower pressure. The contribution of the excited atoms impact by H_2 is negligible with respect to the radiative decay, in the whole pressure range of investigation. The results for dissociation degree obtained through actinometric ratio of spectral line intensities $I(H_\alpha)/I(Ar_{811})$ and $I(H_\beta)/I(Ar_{750})$ coincide quite well. They determine a linear increase of degree of dissociation from 3% to 20% in the pressure range $p = (0.8 - 5.3)$ Pa. The influence of argon excitation cross sections on the obtained results is small, but for practical demands is essential in finding suitable pair for fast monitoring of the hydrogen dissociation degree.

In conclusion two actinometric pairs (H_α, Ar_{811}), (H_β, Ar_{750}) are proposed to examine, hydrogen dissociation degree of this type of discharge. The second is suitable also for fast monitoring of dissociation degree, using the cross sections from Lavrov and Pipa[19] and from Hayashi,[21] knowing only the gas temperature and accounting for spectral sensitivity of the detecting system.

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REFERENCES

1. Lieberman, M. A.; Lichtenberg, A. J. *Principles of Plasma Discharges and Materials Processing*; 2nd ed.; Wiley-Interscience: New Jersey, 2005.
2. Brown, I. G. *The Physics and Technology of Ion Sources*; Wiley-VCH: Weinheim, 2004.
3. Bacal, M.; Hatayama, A.; Peters, J. Volume production negative hydrogen ion sources. *IEEE Trans. Plasma Sci.* **2005**, *33*, 1845–1871.
4. Fantz, U. Emission spectroscopy of molecular low pressure plasmas. *Contrib. Plasma Phys.* **2004**, *44*, 508–515.
5. Hippler, R.; Pfau, S.; Schmidt, M.; Schoenbach, K. H. *Low Temperature Plasma Physics: Fundamental Aspects and Applications*; Wiley-VCH: Berlin, 2001.
6. Coburn, J. W.; Chen, M. Optical emission spectroscopy of reactive plasmas: A method for correlating emission intensities to reactive particle density. *J. Appl. Phys.* **1980**, *51*, 3134–3136.
7. St-Onge, L.; Moisan, M. Hydrogen atom yield in RF and microwave hydrogen discharges. *Plasma Chem. Plasma Process.* **1994**, *14*, 87–116.
8. Rousseau, A.; Granier, A.; Gousset, G.; Leprince, P. Microwave discharge in H_2: Influence of H-atom density on the power balance. *J. Phys. D: Appl. Phys.* **1994**, *27*, 1412–1422.
9. Otorbaev, D. K. ''Simple'' diagnostics for characterization of low-pressure chemically active plasmas. *Pure Appl. Chem.* **2002**, *74*, 453–457.
10. Abdel-Rahman, M.; Schulz-von der Gathen, V.; Gans, T.; Niemi, K.; Döbele, H. F. Determination of the degree of dissociation in an inductively coupled hydrogen plasma using optical emission spectroscopy and laser diagnostics. *Plasma Sources Sci. Technol.* **2006**, *15*, 620–626.
11. Schulz-von der Gathen, V.; Döbele, H. F. Critical comparison of emission spectroscopic determination of dissociation in hydrogen RF discharges. *Plasma Chem. Plasma Process.* **1996**, *16*, 461–486.
12. Gicquel, A.; Chenevier, M.; Hassouni, Kh.; Tserepi, A.; Dubus, M. Validation of actinometry for estimating relative hydrogen atom densities and electron energy evolution in plasma assisted diamond deposition reactors. *J. Appl. Phys.* **1998**, *83*, 7504–7521.
13. Malyshev, M. V.; Donnelly, V. M. Trace rare gases optical emission spectroscopy: Nonintrusive method for measuring electron temperatures in low-pressure, low-temperautre plasmas. *Phys. Rev. E* **1999**, *60*, 6016–6029.
14. Donnelly, V. M. Plasma electron temperatures and electron energy distributions measured by trace rare gases optical emission spectroscopy. *J. Phys. D: Appl. Phys.* **2004**, *37*, R217–R236.
15. Fantz, U.; Wünderlich, D. A novel diagnostic technique for $H^-(D^-)$ densities in negative hydrogen ion sources. *New J. Phys.* **2006**, *8*, 301 (23 pp).
16. Fantz, U.; Falter, H.; Franzen, P.; Wünderlich, D.; Berger, M.; Lorenz, A.; Kraus, W.; McNeely, P.; Riedl, R.; Speth, E. Spectroscopy-a powerful diagnostic tool in source development. *Nucl. Fusion* **2006**, *46*, S297–S306.

17. Kiss'ovski, Z.; Kolev, St.; Shivarova, A.; Tsankov, T. Expanding plasma region of an inductively driven hydrogen discharge. *IEEE Trans. Plasma Sci.* **2007**, *35*, 1149–1155.
18. Kiss'ovski, Z.; Kolev, St.; Müller, S.; Paunska, T.; Shivarova, A.; Tsankov, Ts. Expanding hydrogen plasmas: photodetachment technique diagnostics. *Plasma Phys. Control. Fusion* **2009**, *51*, 015007 (12 pp).
19. Lavrov, B. P.; Pipa, A. V. Account of the fine structure of hydrogen atom levels in the effective emission cross sections of Balmer lines excited by electron Impact in gases and plasma. *Opt. Spectrosc.* **2002**, *92*, 709–719.
20. Walker, Jr. J. D.; John, R. M. St. Design of a high density atomic hydrogen source and determination of Balmer cross sections. *J. Chem. Phys.* **1974**, *61*, 2394–2407.
21. Hayashi, M. *Bibliography of Electron and Photon Cross Sections with Atoms and Molecules – Argon*; NIFS-DATA-72, TOKI, Japan, 2003.
22. Chilton, J. E.; Boffard, J. B.; Schappe, R. S.; Lin, C. C. Measurement of electron-impact excitation into the $3p^54p$ levels of argon using Fourier-transform spectroscopy. *Phys. Rev. A* **1998**, *57*, 267–277.
23. Bartschat, K.; Zeman, V. Electron-impact excitation from the ($3p^54s$) metastable states of argon. *Phys. Rev. A* **1999**, *59*, R2552–R2554.
24. http://nist.gov/
25. Wiese, W. L.; Brault, J. W.; Danzmann, K.; Helbig, V.; Kock, M. Unified set of atomic transition probabilities for neutral argon. *Phys. Rev. A* **1989**, *39*, 2461–2471.
26. Möhlmann, G. R.; De Heer, F. J.; Los, J. Emission cross sections of Balmer – α,β,γ radiation for electrons (0–2000 eV) on H_2 and D_2. *Chem. Phys.* **1977**, *25*, 103–116.
27. Wouters, M. J.; Khachan, J.; Falconer, I. S.; James, B. W. Quenching of excited Ar I and H by H_2 in a gas discharge. *J. Phys. B: At. Mol. Opt. Phys.* **1999**, *32*, 2869–2880.
28. Biberman, L. M.; Vorobev, V. S.; Yakubov, I. T. *Kinetics of Nonequilibrium Low-Temperature Plasma*; Nauka: Moscow, 1982.
29. Iordanova, S. Spectroscopic temperature measurements in hydrogen inductively-driven plasmas at low pressures. *J. Phys.: Conf. Series* **2008**, *113*, 012005 (5 pp).
30. Marechal, M. A.; Jost, R.; Lombardi, M. Lifetimes, g factors, and collision cross sections of hydrogen molecules in the (1s3p) $^3\Pi_u$. *Level. Phys. Rev. A* **1972**, *5*, 732–740.
31. Lavrov, B. P. Determination of the gas temperature of a low-pressure plasma from the intensities of the H_2 and D_2 molecular bands. Relationship between the intensity distribution in a band and the gas temperature. *Opt. Spectrosc.* **1980**, *48*, 375–380.
32. Qing, Z.; Otorbaev, D. K.; Brussaard, G. J. H.; van de Sanden, M. C. M.; Schram, D. C. Diagnostics of the magnetized low-pressure hydrogen plasma jet: Molecular regime. *J. Appl. Phys.* **1996**, *80*, 1312–1324.
33. McNeely, P.; Dudin, S. V.; Christ-Koch, S.; Fantz, U. NNBI Team A Langmuir probe system for high power RF-driven negative ion sources on high potential. *Plasma Sources Sci. Technol.* **2009**, *18*, 014011 (17 pp).
34. Volynets, V. N.; Park, W.; Tolmachev, Yu. N.; Pashkovsky, V. G. Spatial variation of plasma parameters and ion acceleration in an inductive plasma system. *J. Appl. Phys.* **2006**, *99*, 043302 (5 pp).
35. Lishev, St.; Shivarova, A.; Tsankov, T. Experiments on the detection of negative hydrogen Ions in a small-size tandem plasma source. *AIP Conf. Proc.* **2009**, *1097*, 127–136.
36. Djermanov, I.; Kolev, St.; Lishev, St.; Shivarova, A.; Tsankov, Ts. Plasma behaviour affected by a magnetic filter. *J. Phys.: Conf. Series* **2007**, *63*, 012021 (6 pp).
37. Iordanova, S.; Koleva, I. Optical emission spectroscopy diagnostics of inductively-driven plasmas in argon gas at low pressures. *Spectrochim. Acta Part B* **2007**, *62*, 344–356.
38. Crintea, D. L.; Czarnetzki, U.; Iordanova, S.; Koleva, I.; Luggenhölscher, D. Plasma diagnostics by optical emission spectroscopy in argon and comparison with Thomson scattering. *J. Phys. D: Appl. Phys.* **2009**, *42*, 045208 (11 pp).