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TEMPERATURES OF EXCITED HYDROGEN ATOMS IN THE ABNORMAL GLOW DISCHARGE

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Abstract. The results of the spectroscopic measurements of excited hydrogen atoms temperatures in the cathode fall and beginning of the negative glow region of the Grimm-type abnormal glow discharge in pure hydrogen and argon-hydrogen mixture are reported. The origin of energetic hydrogen atoms, excited in collisions with matrix gas, is explained. Higher temperatures at the beginning of the negative glow region are related to the additional excitation in collisions with electrons.

1. INTRODUCTION

Recent studies of atomic-hydrogen line shapes in the vicinity of the cathode, in various types of glow discharges (Benesch and Li, 1984; Cappelly et al., 1985; Baravian et al., 1987; Li Ayers and Benesch, 1988; Barbeau and Jolly, 1990; Kuraica and Konjević, 1992), has shown Balmer lines shapes with an extraordinary wings development. The extensive far wings indicate the presence of excited hydrogen atoms with very high velocities. As shown by both theory and experiment (Petrović et al., 1992 and references [4,10] therein), those energetic hydrogen neutrals originate from incident H^+, H, H_2^+, H_2 and H_3^+, whose backscattered fragments from the cathode are almost entirely H atoms. For low incident energies, the energy of reflected H atoms increase with incident energy. On their way back through the discharge, they collide mainly with matrix gas and excite (Kuraica and Konjević, 1994).

In this work, temperatures of excited hydrogen atoms are spectroscopically measured in the cathode fall and at the beginning of the negative glow region of the plane cathode and cylindrical hollow anode abnormal glow discharge of the Grimm-type (Grimm, 1968). Experimental setup is fully described elsewhere (Kuraica et al., 1992; Videnović et al., 1995), so only minimum theoretical details will be given here for clearness.

In the cathode fall region, the presence of external electric field predominantly determines the shape of hydrogen Balmer lines. Therefore, the theory of polarization dependent Stark splitting has to be employed (see more details in Videnović et al., this Volume). Placing the polarizer parallel or perpendicular to the discharge axis, we have selected components with $\Delta m = 0$ or π, and $\Delta m = \pm 1$ or σ polarization respectively. All components (10 π and 10 σ for Balmer H_β line, used throughout this work) form the appropriate π or σ overall profile. In the cathode fall region, we assume Doppler broadening only, since plasma broadening in this region is negligible. Therefore, to the each Stark component we have assigned Gauss function, which takes into account Doppler and instrumental broadening. Considering overall profile as the sum of all gaussians, we have fitted it to the experimental recordings, varying electric field intensity E end temperature T of hydrogen atoms. In the negative glow region, the difference between π and σ profiles disappears ($E \approx 0$), and the fitting procedure is reduced to the varying of T only. Here we shall discuss only the best-fit temperature results.

2. RESULTS AND DISCUSSION

The measurements have been performed at pressures of 195, 228 and 250 Pa and discharge currents of 20, 30 and 40 mA in pure hydrogen, and 240, 320 and 425 Pa of argon-hydrogen mixture (97% Ar : 3% H_2), at currents of 20 and 30 mA.

Typical results of Balmer H_β spectra recordings and fitting procedures in the cathode fall and negative glow regions of Grimm discharge in both investigated gases are shown in Fig. 1. Two graphs on the far right-hand side refer to the negative glow region, where no difference between two polarizations occurs. As reported before (Videnović et al., 1995), in pure hydrogen, best fits were achieved starting with assumption that two groups of excited atoms with considerably different velocities exist: so-called "slow" and "fast" neutrals with temperatures about 5 eV and 100 eV respectively. In argon-hydrogen mixture, only one group of energetic neutrals is detected, with temperatures about 40 eV. All the results, obtained in various experimental conditions are given in Table 1.

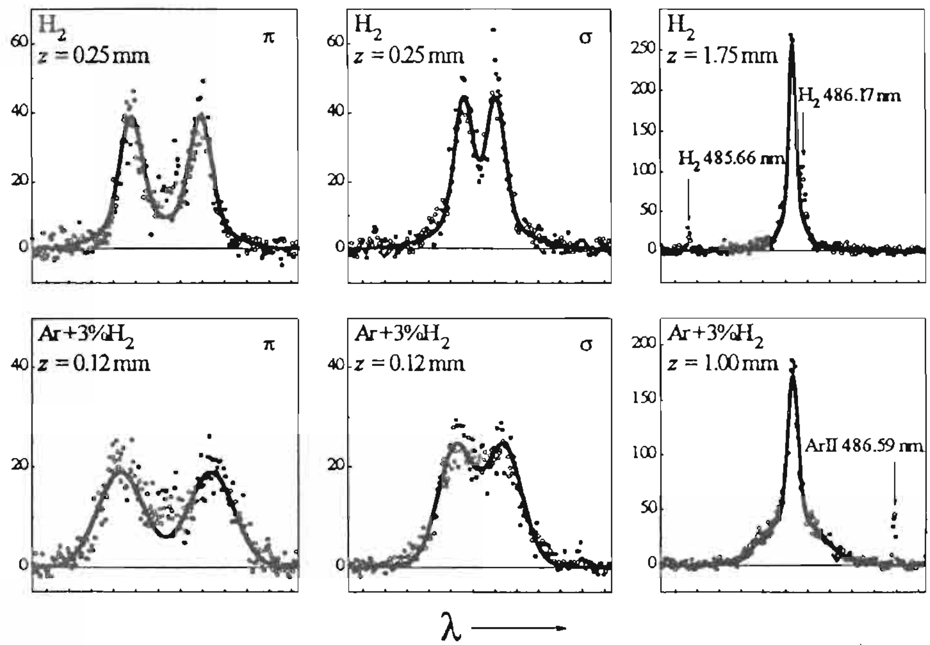

Fig. 1. Typical measured H_β line profiles and their best fits. z is the distance from the cathode. Two graphs on the far right-hand side refer to the negative glow region where no difference between polarized profiles occurs. The wavelength scale is given in 0.1 nm units. Discharge conditions: pure H_2: 228 Pa, 30 mA, 920 V; Ar+3%H_2: 320 Pa, 30 mA, 820V.

Table 1. Best-fit temperatures (in eV) of excited hydrogen atoms in investigated regions, in various experimental conditions.

Working gas		Cathode fall	Negative glow
H_2	"slow"	3.4 - 8.2	5.0 - 10.4
	"fast"	80 - 190	90 - 160
Ar+3%H_2		32 - 43	49 - 59

2.1. PURE HYDROGEN DISCHARGE

In the cathode fall of pure hydrogen discharge, three principal ion species exist: H^+, H_2^+ and H_3^+. On their way to the cathode, they involve following charge exchange reactions:

$$(H^+)_{fast} + (H_2)_{slow} \rightarrow H^*_{fast} + (H_2^+)_{slow} \qquad Q \approx 3.7\cdot 10^{-17}\ \mathrm{cm}^2,$$
$$(H_2^+)_{fast} + (H_2)_{slow} \rightarrow H^*_{fast} + H_{fast} + (H_2^+)_{slow} \qquad Q \approx 8.4\cdot 10^{-16}\ \mathrm{cm}^2,$$
$$(H_3^+)_{fast} + (H_2)_{slow} \rightarrow H^*_{fast} + (H_2)_{fast} + (H_2^+)_{slow} \qquad Q \approx 2.1\cdot 10^{-17}\ \mathrm{cm}^2.$$

where Q are the cross section data at about 100 eV laboratory ion energy (Phelps, 1990). Due to largest charge exchange cross section, H_2^+ has the lowest energy at the cathode surface. Using mass spectrometric results of Dexter et al. (1989) in a hydrogen glow discharge operating at 133 Pa, 6 mA and 500 V, Barbeau and Jolly (1990) calculated the mean energy of ions reaching the cathode surface (in eV):

$$\bar{\varepsilon}_{H^+} = 170, \qquad \bar{\varepsilon}_{H_2^+} = 35, \qquad \bar{\varepsilon}_{H_3^+} = 75.$$

According to previous investigations (Petrović et al., 1992 and refs. [4,10] therein), the ions loose about 1/3 of the incident energy in the collision with the cathode. Therefore, energy of the backscattered hydrogen atoms at the cathode surface are (in eV):

$$\bar{\varepsilon}_{H(H^+)} = 114 \qquad \bar{\varepsilon}_{H(H_2^+)} = 11.7 \qquad \bar{\varepsilon}_{H(H_3^+)} = 16.8$$

Hence, two groups of hydrogen atoms, originated from H^+ and H_3^+ ions, have enough energy to exceed threshold for Balmer lines excitation (for H_β, $\varepsilon_{ext} = 12.76$ eV). The result of this simple calculation suggests the origin of "slow" and "fast" excited hydrogen atoms, see Table 1. "Slow" H atoms are, most likely, created after neutralization and fragmentation of H_3^+ ions. Temperatures of "slow" atoms varies in our case from 3.4-8.2 eV what corresponds to H_3^+ energies in the range 72-94 eV. On the other hand, the origin of "fast" excited H atoms, with temperatures between 80 eV and 190 eV, may be related to energetic H^+ ions only, with incident energies ranging from 140-300 eV. After the reflection from the cathode, both groups of neutrals collide mainly with H_2 and excite, see e.g. Kuraica and Konjević (1994).

2.2. DISCHARGE IN ARGON-HYDROGEN MIXTURE

In argon-hydrogen discharge, the presence of argon contributes to the efficient production of H_3^+ ions through following reactions:

$$Ar^+ + H_2 \rightarrow ArH^+ + H,$$
$$ArH^+ + H_2 \rightarrow H_3^+ + Ar,$$

and

$$Ar^+(3p^5\ {}^2P^0_{1/2}) + H_2 \rightarrow Ar + H_2^+,$$
$$H_2^+ + H_2 \rightarrow H_3^+ + H,$$

see Kuraica and Konjević (1994) and the cross section data for above reactions (Phelps, 1990, 1992). Large concentration of H_3^+ ions increases the intensities of hydrogen line wings, see Fig. 1. Therefore, H_3^+ is now the dominating hydrogen ion and backscattered H atoms from the cathode originate mainly from this ion. The temperatures in range 32-43 eV corresponds to the incident H_3^+ energies of 134-168 eV. In comparison with pure hydrogen, where "slow" H atoms are formed in the same way, here H_3^+ ions gain more energy in the cathode fall region. This could be related to the higher argon transparency for H_3^+ (mass ratio 3:40) in comparison with transparency of hydrogen matrix (3:2).

2.3. NEGATIVE GLOW REGION

In the negative glow region, the central narrow peak is induced by Stark and Doppler (gas temperature $T_g \approx 0.1$ eV) broadening, see Fig. 1. Analyzing lower, broader part of the profiles, in both investigated gases, the temperature increase have been detected, see Table 1. Although unexpected, this effect could be explained by the additional excitation of hydrogen atoms in collisions with electrons, whose concentration is about 10^{14} cm^{-3} in this part of the discharge (Kuraica et al., 1992). Since the electron-atom collisions change internal energy of atoms only, their temperature remains high. Actually, in this region, one has, most probably, the superposition of two profiles: one emitted by H atoms excited in collisions with matrix gas, and another, broader, induced by electron excitation of H atoms. Unfortunately, with present spectral resolution (see Videnović et al., 1995), we could not resolve these profiles.

Another argument in favour of this explanation can be supplied on the basis of expected exponential decrease of energetic H atoms number, due to collisions with matrix gas, along their path from the cathode towards negative glow region. The calculation, using total cross section data (Phelps, 1990) for collisions of neutral hydrogen atoms at 133.4 eV with H_2 (228 Pa, T_g = 1000 K and cathode dark space length L = 0.16 cm - typical value in pure hydrogen) shows that about 18% of reflected "fast" H atoms arrive to the negative glow without any collision. Similar calculation using total cross section data (Phelps, 1992) for collisions of H atoms at 75 eV with argon (320 Pa T_g = 1000 K and L = 0.08 cm - typical for Ar+3%H_2), shows that about 67% of reflected neutrals reach negative glow region without collisions. These percentages agree well with areas of lower, broader parts in comparison to overall profiles, see far right-hand side graphs in Fig. 1, and also explain the more pronounced effect of temperature increase in argon-hydrogen then in pure hydrogen discharge, see Table 1.

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