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EMISSION AND ADSORPTION SPECTROSCOPY MEASUREMENTS OF ATOMIC HYDROGEN LEVEL POPULATIONS IN AN EXPANDING MAGNETIZED PLASMA

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The absolute density of atomic hydrogen excited states in two different regimes of a magnetized expanding plasma is determined using emission and absorption spectroscopy. First evidences have been found for the presence of high densities of the negative ions in the "atomic" (recombining) regime an expanding hydrogen plasma. Several clear observations of the presence of "hot" electrons have been measured for the "molecular" (ionizing) regime of an expanding plasma. For both regimes the absolute density of atomic hydrogen in the ground state and plasma dissociation degree have been determined.

INTRODUCTION

Development and characterization of high intensity sources of reactive short-living radicals are of great interest to surface physics, fusion and low-temperature plasma physics community. As a rule, kinetics of the radicals in the plasma is difficult to simulate since it requires accurate knowledge of all the relevant kinetic processes, including heterogeneous phenomena on the walls [1]. The aim of this paper is to present the results of diagnostics and to describe the radical kinetics in the two different regimes of an expanding magnetized hydrogen plasma.

EXPERIMENTAL

The experiments are carried out in an expanding pure hydrogen plasma produced in a cascaded arc described in detail elsewhere [2,3]. For the present studies a magnetic coil (diameter $\approx$ 0.5 m) is added at the location of the anode of the cascaded arc. This leads to an diverging magnetic field with a maximum induction between 8 and 40 mT on the axis. The motivation to apply a magnetic field is to extend the parameter range to lower pressure and thus to avoid the observed strong recombination in a freely expanding hydrogen plasma [2]. So-called "molecular" regime of an expanding plasma has been realized for the magnetic field induction B_{max} = 8 mT, so-called "atomic" regime for the B_{max} = 40 mT. In comparison with previous experiments on the expanding hydrogen plasma jet, the hydrogen flow was 8 scc/s, and with that the pressure in the cascaded arc was low as well (about 20 mbar in the cathode region). This makes the assumption of a thermal plasma questionable [2]. The pressure in the vessel and the arc current were kept constant at respectively 5 Pa and 50 A (V_{arc} = 130 V).

The measurements of an expanding plasma characteristics were carried out using a movable Langmuir electrostatic probe and spectroscopic system. The probe characteristics were interpreted using classical Langmuir theory, assuming a negligible sheath thickness. The lateral scans of the hydrogen Balmer spectral lines emissivities were performed at axial positions from 18 to 30 cm downstream of the arc nozzle. An Abel inversion was applied to derive local values of the emission coefficients and hydrogen excited state absolute densities. To measure the absolute density of the first excited states of H and H_2 a method of reabsorption with a mirror (which is identical to the method of two identical light sources [4] has been used. A concave mirror of reflectance r was placed behind the plasma beam, and the line intensity was measured with a mirror covered, or not covered [5]. The mirror reflectance coefficient $r = r(\lambda)$ has be measured using a spectral line for which the plasma was optically thin.

RESULTS AND DISCUSSION

"Atomic" regime of an expanding plasma

The prominent characteristic of the visible spectrum of an expanding hydrogen plasma in the "atomic" regime was a strong emission of atomic hydrogen spectral lines. The radiation from the quantum state up to p = 18 is easily detected. In Fig. 1 the absolute population densities on the plasma beam axis at a position x = 18 cm from the nozzle as derived from emission and absorption

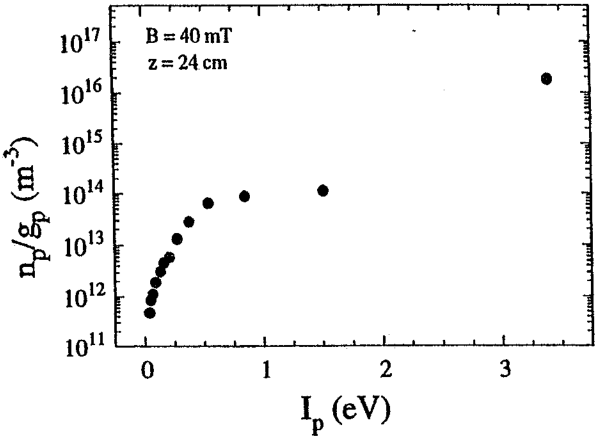

Fig. 1

spectroscopy measurements are shown as a function of the ionization potential I_p of the level p. Note the large population density per statistical weight for the levels $3 < p < 6$. Also an inversion is observed for the level $p = 7$ for large distances from the anode nozzle. In general the inversion is more pronounced downstream in the plasma jet and the maximum of n_p/g_p vs. I_p occurs for the higher quantum numbers.

Atomic hydrogen density in the first excited state H*(p = 2) has been determined by $H_\alpha(\lambda = 656.3$ nm) spectral line absorption. To the same spectral range as H_α belongs the spectral line of argon: $\lambda = 660.5$ nm (radiative transition $Ar(3p^5 4d \rightarrow 3p^5 4p)$). It has been shown, that the plasma was optically transparent for this line, and this line can be used to determine the reflectance coefficient of the mirror. In Fig. 1 the absolute population density of H*(p = 2) per statistical weight is shown as well.

Comparing the measured population densities (Fig. 1) with the densities calculated on basis of the measured n_e and T_e (see later), using a purely atomic collisonal-radiative model, leads to the conclusion that purely atomic recombination processes can not account for the large population densities observed. It is argued that molecular induced recombination reactions in which the negative ion participates should be taken into account [6,7]:

$$H_2^+[H^+] + H^- \longrightarrow H_2[H] + H^*(p). \qquad (1)$$

Note that the reaction of mutual neutralization of H^+ and H^- can only lead to the excitation of the quantum states $H(p \leq 3)$. Hydrogen negative ions H^- and positive molecular ions H_2^+ are either generated by the arc or formed in the reactions with the participation of rovibrationally excited $H_2^{v,J}$ molecules:

$$H_2^{v,J} + e \longrightarrow H^- + H, \qquad (2)$$

$$H_2^{v,J} + H^+ \longrightarrow H_2^+ + H. \qquad (3)$$

Kinetic analysis of the rates of various processes in the expanding plasma show, that in the discussed regime of an expanding magnetized hydrogen plasma only spontaneous radiative processes are controlling the density of H*(n = 2) state; namely, radiative decay from the high-lying atomic states, and partially trapped resonance radiation to the ground state. Therefore the general balance equation for H*(n=2) can be transformed to the following:

$$\sum_{n=3}^{\infty} \Phi_{n\rightarrow 2} = \Phi_{2\rightarrow 1} = n_2 A_{21} \Lambda_{21}, \qquad (4)$$

where $\Phi_{k\rightarrow i}$ are the radiation fluxes between the quantum states k and i, n_2 is the absolute density of H*(p = 2) state, A_{21} is the absolute transition probability of the resonance Lyman-α spectral line, and Λ_{21} is the escape factor for resonance radiation. Another important conclusion can be derived from the kinetic analysis. Namely, in the plasma it should be a strong production source of highly excited hydrogen atoms H*(p $\geq$ 3), which then radiatively decay to H*(p = 2) state. This statement is confirming the conclusion made earlier about the excitation mechanism of the highly excited hydrogen states.

The experimental information on the population density of hydrogen atoms in the excited states $H^*(n \geq 2)$ (cf. Fig. 1), and known radiative transition probabilities [8], allows to determine from (4) the escape factor Λ_{21} for resonance Lyman-α radiation. For the conditions mentioned of an expanding hydrogen plasma at a distance of $x = 24$ cm from the nozzle of the arc the escape factor was equal to $\Lambda \simeq 1.8 \cdot 10^{-3}$. So, in other words the plasma under investigation can be described as an optically thick medium for the resonance Lyman-α radiation. The escape factor is related to the optical depth $\overline{kR}$ of the absorbing medium. The relation between the escape factor and optical depth has been calculated for Voigt emission profiles in [9] for the case of cylindrically symmetric plasmas. Since the radius of the plasma beam is known one can determine an over the line of sight average of the absolute population density of $H(n = 1)$ state. In the "atomic" regime of an expanding hydrogen plasma at the position $x = 24$ cm from the arc nozzle, the averaged atomic hydrogen ground state density will be $n_{H(n=1)} \simeq 1.7 \cdot 10^{20}$ m^{-3}. The corresponding dissociation degree of hydrogen plasma is equal to $\beta \simeq 11$ %. It is essential that an accuracy of the discussed procedure is determined practically only by the experimental accuracy, but not by the uncertainties in the kinetic coefficients, since the radiative transition probabilities, which were used in the kinetic scheme are known with the high accuracy (better than 1 % [8]).

The correctness of the Langmuir probe diagnostic has been checked by comparison of the results with the Thomson scattering measurements, which is a non-intrusive optical diagnostic to determine similtaneously the electron density and temperature [3]. The mean ion energy were approximately 0.3 eV, whereas the ion density were in the range $(1.4 - 2.4) \cdot 10^{17}$ m^{-3}.

"Molecular" regime of an expanding plasma

It has been shown, that electron temperature in the expansion as determined by Langmuir probe diagnostics in the "molecular" regime is rather high, and for all positions in the expansion exceed 10^4 K, i.e. 1 eV. In the emission spectrum in this regime the strong emission of the Fulcher-α band of $H_2(d^3\Pi_u \rightarrow a^3\Sigma_g^+$ transition) (excitation potential is $\Delta E_1 = 13.87$ eV) can be observed. Rotational (excitation) temperature and neutral particle (gas) temperature of an expanding plasma can be derived from the analysis of rotational spectra of Fulcher- α system [10].

Typical results are shown in Fig. 3. The Boltzmann plot of the rotational lines intensities clearly shows that within the experimental error all three curves, corresponding to (0-0), (1-1) and (2-2) bands of Fulcher-α system can be considered as linear with approximately the same slope. The rotational excitation temperature of $H_2(d^3\Pi_u)$ state, derived from slope of the lines (Fig. 3), is in the range of 245 - 280 K, therefore the gas temperature in the plasma should be very low and in the range of 490 - 560 K (see [10]).

It is important to mention the experiments with a local excitation of the Ar and He spectral lines. In this case the transportation of Ar and He to the particular local points in the plasma active zone has been made through the thin cylindrical cyramic tube. In the "atomic" regime of an expanding plasma within a detection limit of the optical system we did not observe any presence of argon and helium spectral lines. However in the "molecular" regime one can see an appearance of the spectral line both of neutral argon Ar I $(7d \rightarrow 4p)\lambda = 4876.26$ A and $\lambda = 4887.95$ A, and of ion argon Ar II $(4p^2D \rightarrow 4s^2D)\lambda = 4879.86$ A. The excitation potential of these lines $\Delta E_2 = 15.45$ eV, and $\Delta E_3 = 19.68$ eV, respectively. An appearance of the spectral lines with even higher excitation potentials has been observed in the case of helium, namely the spectral lines of He I $(3d \rightarrow 2p)\lambda = 5875.70$ A ($\Delta E_4 = 23.07$ eV), and He I $(5d \rightarrow 2p)\lambda = 4026.20$ A ($\Delta E_5 = 24.04$ eV) can be seen in the emission spectra of an expanding plasma.

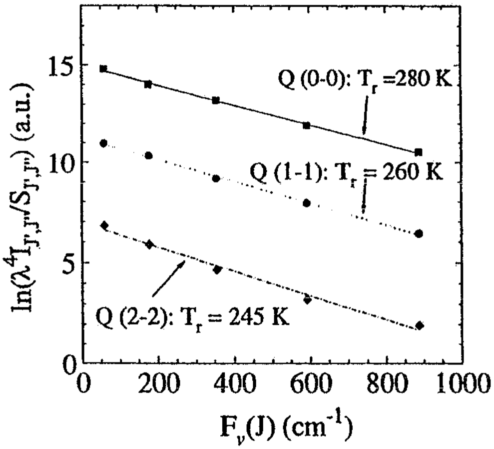

Fig. 2

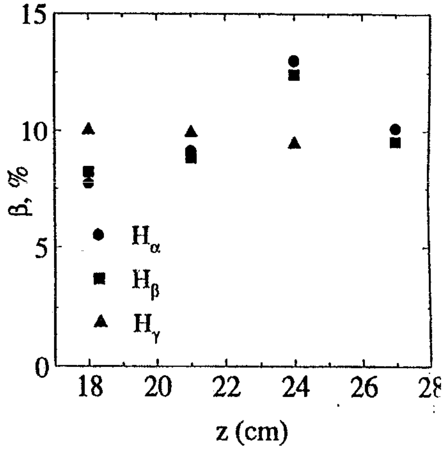

Fig. 3

Optical actinometry has been applied to measure the absolute density of hydrogen atoms and hydrogen dissociation degree in an expanding magnetized plasma. It has been shown that the atomic H*(n = 3, 4, 5), and molecular $H_2(d^3\Pi_u)$ radiative quantum states are excited by a direct electron impact from the ground states H(n = 1) and $H_2(X^1\Sigma_g^+)$, respectively, and depopulated via spontaneous radiative decay (corona balance). Method of reabsorption with a concave mirror has been used in order to determine the population density of the first excited states of H and H_2, and thus to estimate influence of the spepwise electron excitation. The densities were lower than 10^{15} m^{-3}, and it can easily be shown, that in this case the stepwise electron excitation was unefficient.

The results of the determination of the dissociation degree on the axis of an expanding hydrogen plasma are shown in Fig. 3. It is essential to mention that the dissociation degree do not strongly depend the atomic spectral lines selected, i.e. H_α, H_β or H_γ. We believe that the presented results provide a strong support to the corona balance approximation, and to the optical actinometry method as a whole. The measured absolute density of hydrogen atoms are in the $(1 - 1.4) \cdot 10^{20}$ m^{-3} range, and dissociation degree of the hydrogen plasma are in the range of 8 - 13 %. It can be seen that within the experimental accuracy the dissociation degree does not depend on the axial position. That probably indicates that for given experimental conditions the recirculation gas flows in the vessel are ineffective, and both processes of H_2 dissociation, and of H recombination are slow, and do not influence the kinetics of hydrogen atoms in the plasma.

CONCLUSIONS

Two different regimes of an expanding magnetized hydrogen plasma have been investigated. Several clear observations of the presence of "hot" electrons have been measured for the "molecular" (ionizing) regime of an expanding plasma. For both regimes the absolute density of atomic hydrogen in the ground state and plasma dissociation degree have been determined.

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