

EMISSION SPECTROSCOPY OF DIPOLAR PLASMA SOURCE IN LOW PRESSURE HYDROGEN

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Introduction

Vibrational and rotational excited hydrogen molecules in the ground $X^1\Sigma_g^+$ state and electronic states play important role in the in the formation of the negative atomic hydrogen ions, positive ions and hydrogen atoms in gas discharges in hydrogen. Chemical activity of hydrogen plasma increases with increasing concentrations of excited particles. Translational (gas) temperature is an important plasma parameter as rate coefficients of most of plasma processes depends on gas temperature.

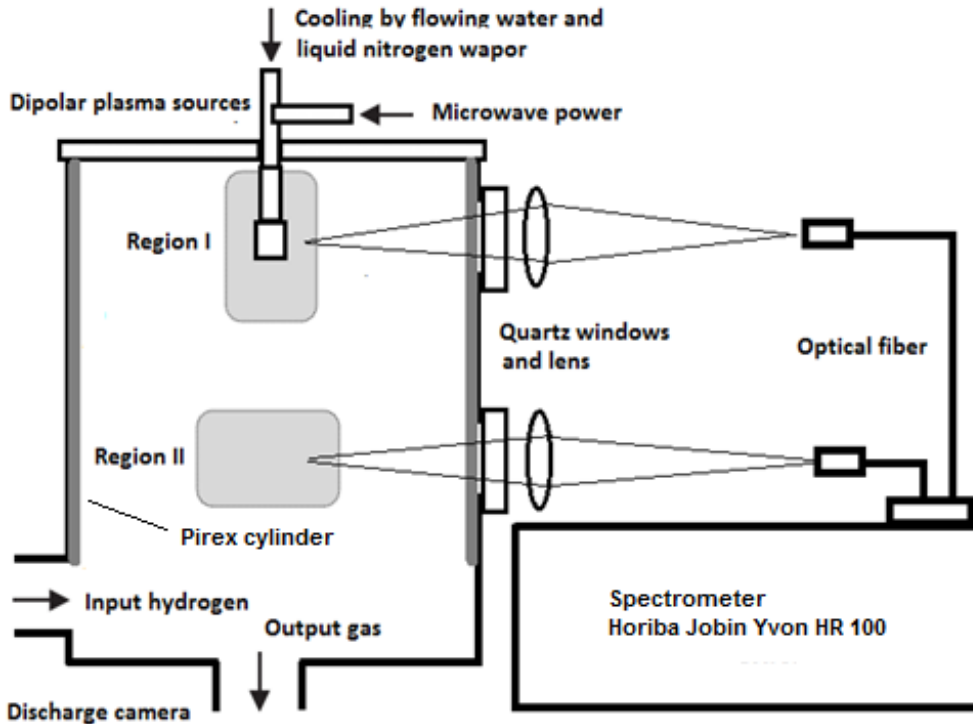
The aim of this paper is study of rotational and vibrational distribution functions and corresponding temperatures in low pressure hydrogen plasma generated by the dipolar plasma source.

OUTLINE

- **Introduction**
- **Experimental setup**
- **Composition of emission spectra of dipolar plasma source.**
- **Rotational distribution of the $d^3\Pi_u$ state and rotational temperature of excited and ground state.**
- **Vibrational distribution of hydrogen molecules in $d^3\Pi_u$ state.**
- **Comments on determination of vibrational temperature of hydrogen in the ground state from experimental data for the $d^3\Pi_u$ state.**
- **Conclusions**

Experimental setup

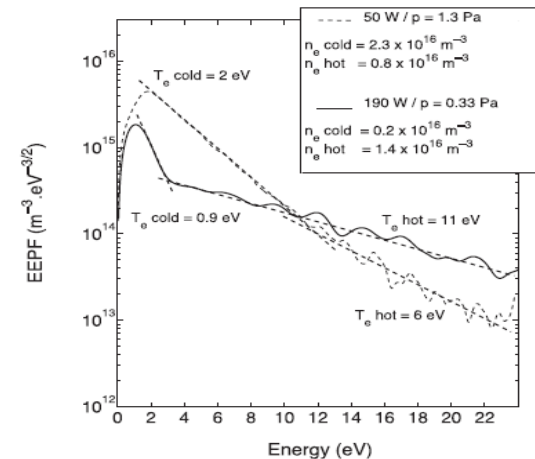
Dipolar plasma source



Experimental setup

Discharge chamber is a stainless steel cylinder with a diameter of 100 mm. Walls are insulated from the plasma by glass cylinder. Method of the relative intensities is used for determination of the rotational (RDF) and vibrational (VDF) distribution functions of the hydrogen molecule in the $d^3\Pi_u$ state.

- Plasma gas H_2
- Residence time of plasma particles $< 10^{-4}$ s
- Frequency 2.45 GHz
- Absorbed power 50 190 W
- Pressure 0.01 2.5 10^{-3} Torr
- Electron energy distribution function is bi-maxwellian
- Region near the dipolar source (region I)
- T_e (cold) = 2.0 0.9 eV
- T_e (hot) = 6.0 11. eV
- N_e (cold) = 2.3 10^{10} 0.2 10^{10} cm^{-3}
- N_e (hot) = 0.8 10^{10} 1.4 10^{10} cm^{-3}
- Diffuse region (region II)
- T_e (cold) = 0.6 0.7 eV
- T_e (hot) = 2.0 2.7 eV
- N_e (cold) = 1.4 10^{10} 1.3 10^9 cm^{-3}
- N_e (hot) = 0.3 10^{10} 4.6 10^9 cm^{-3}



Experimental setup

Dipolar plasma source

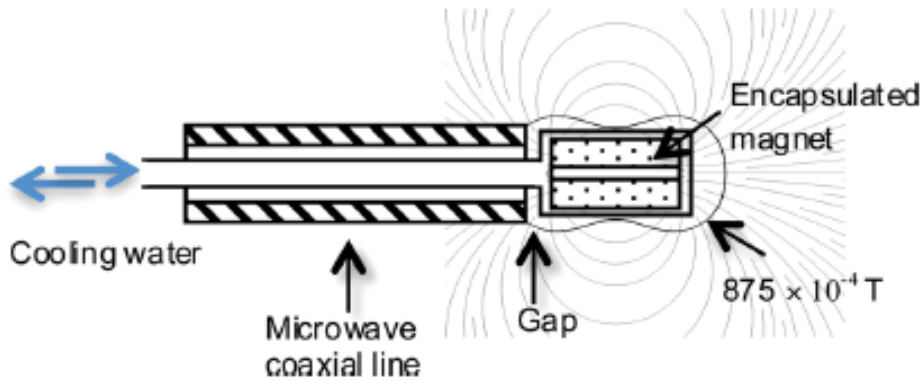
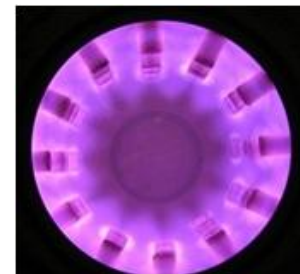
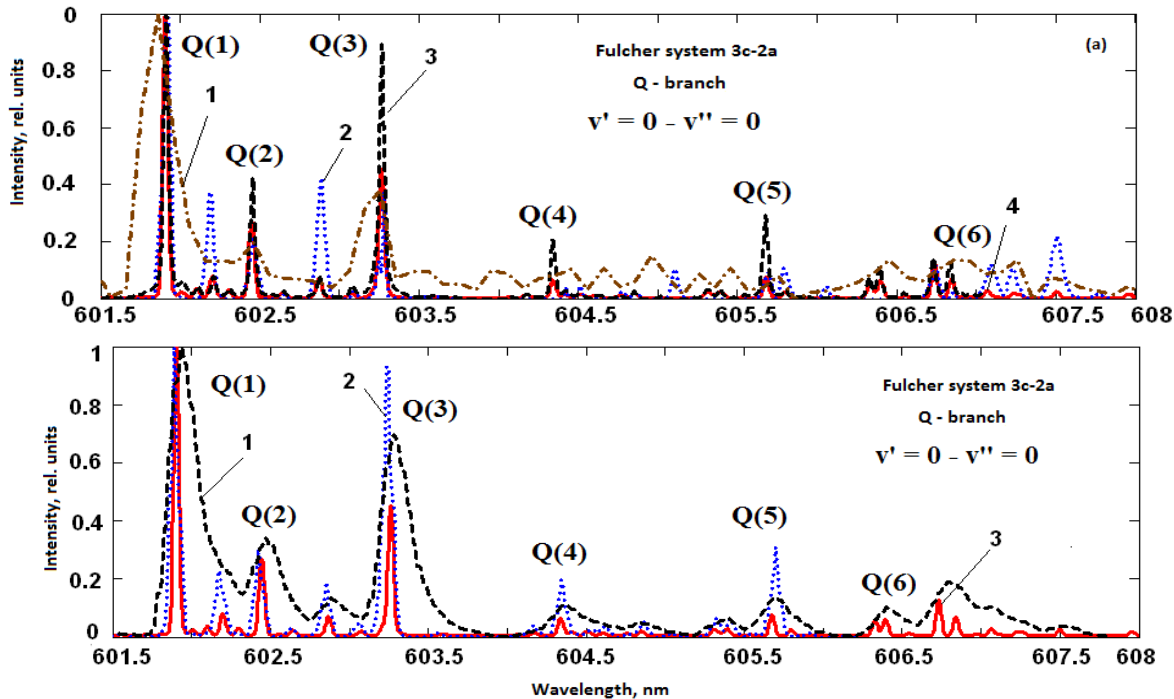


Photo of plasma of dipolar source in hydrogen at pressure of 0.01 Torr and the incident microwave power of 190 W

Conceptual design of an elementary plasma source. The microwave line is plugged on the left side of the source. At the right side, the microwave line ends at the rear side of the encapsulated magnet. The size of the gap between the coaxial line and the magnet is 4.5 mm. Magnetic field lines (thin continuous lines) and line of field equal intensity (875104 T) (peanut shape line) are also represented. [S. Beshu et al, PHYSICS OF PLASMAS 20, 101601 (2013)]



Emission spectra of plasma



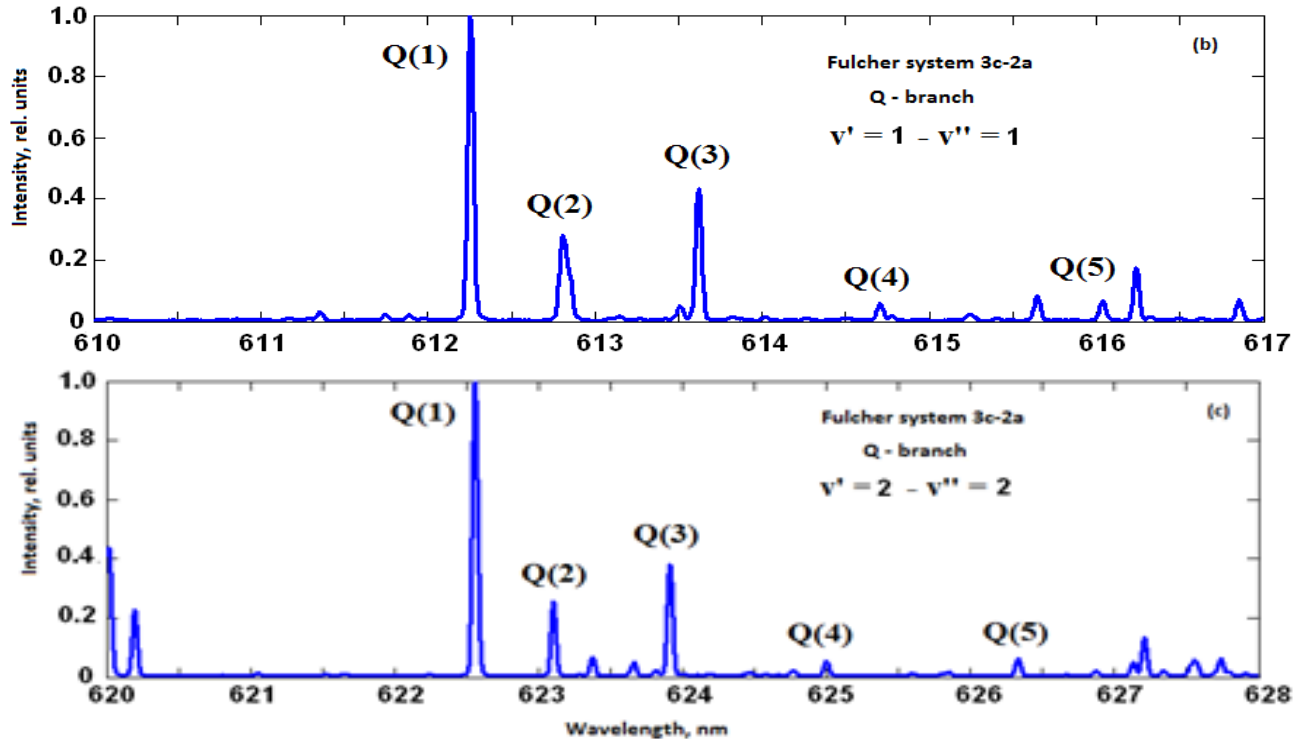
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4. Present study

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3. Present study.

Spectral composition of emission of the ECR discharge coincides with those which are observed in RF discharges [8-11].

The spectral composition of the ECR discharge is more saturated with the lines of the **singlet and triplet transitions** of the hydrogen molecule as compared the emission of the glow DC discharge [1]. This can be due to the difference of the electron concentrations and electron energy distribution functions (EEDFs) in both discharges. The EEDF in the ECR discharge, contrary to that in the positive column of the DC glow discharge [1], is enriched with the "hot" electrons in the energy range of thresholds of excitation of the singlet ($N^1\Lambda_{\sigma, v_{\Lambda}}$, 11.4–14.8 eV) and triplet ($N^3\Lambda_{\sigma, v_{\Lambda}}$, 6–11 eV) states of the hydrogen molecule by direct electron impact.

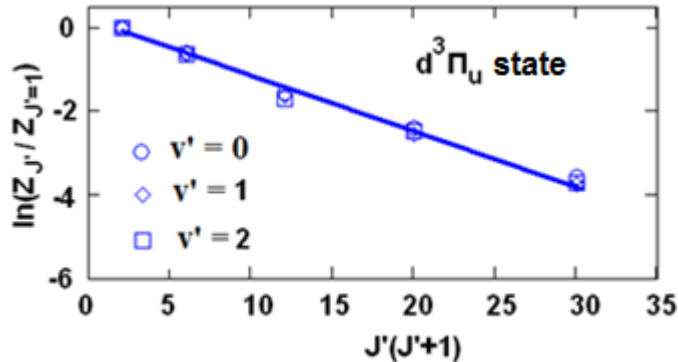
Emission spectra of plasma



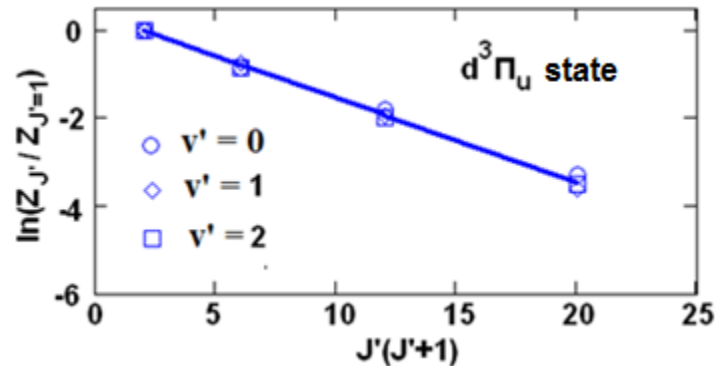
The intensive lines of the Q - branches (Q(1), Q(2), Q(3), Q(4) and Q(5)) of the sequences with $\Delta v=0-2$ of the Fulcher α - system $H_2(d^3\Pi_u, v_d \rightarrow a^3\Sigma_g^+, v_a)$ were selected for the study of the rotational and vibrational distribution functions of hydrogen molecule in the state $d^3\Pi_u$.

Rotational distribution function of the $d^3\Pi_u$ state

ECR region



Diffuse region



- Rotational distribution functions of the $d^3\Pi_u$ state for vibrational levels $V=0-2$ coincide with the Boltzmann distribution, both in ECR and diffusion regions
- Upon cooling of dipolar plasma source by running water the rotational temperatures in the ECR region corresponding the “ortho-“ and “para-“ modifications of hydrogen molecule are identical within error limits for the vibrational levels $v_d=0-2$ and equal to 315 ± 30 K. Upon cooling of plasma source by vapor of liquid nitrogen the temperatures are only slightly differ from those obtained by cooling by running water.
- The similar conclusions are valid for the region II. and values are consistent within the error, but their magnitude is smaller than those obtained in region I and equal to 230 ± 35 K for the same vibrational levels.

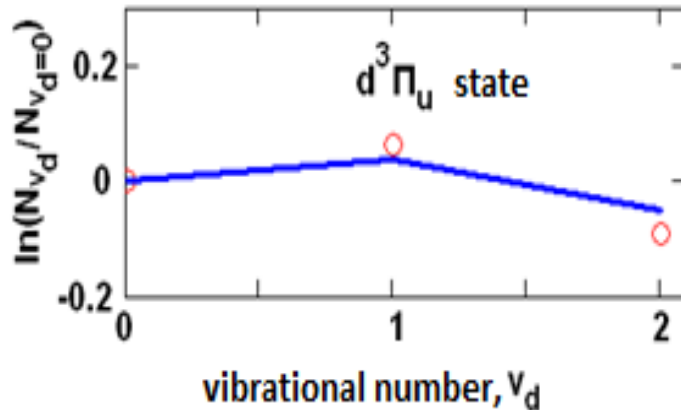
Translational (gas) temperature

Rotational temperature is equal to the gas temperature if:

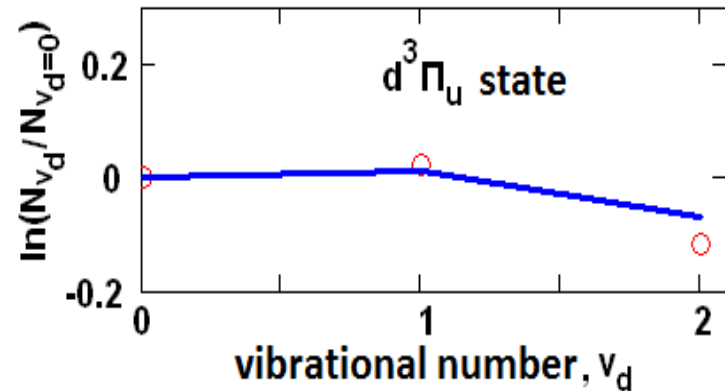
- The $d^3\Pi_u$ state, $v=0-2$, $J=1-5$ of the hydrogen molecule is formed in result of electron impact excitation from the ground state and quenched via radiative decay without reabsorption.
- Rotational distribution of molecules on the rotational levels $J=1-5$ in the $d^3\Pi_u$ state corresponds to the Boltzmann distribution. It is true for the $v=0-2$, $J=1-5$ states of the hydrogen molecule in both regions of the ECR discharge.
- Equilibrium between translational and rotational degrees of freedom exists for the hydrogen molecules in the ground electronic state $X^1\Sigma_g^+$. The RDF in the $X^1\Sigma_g^+$ state of the hydrogen molecule is described by the Boltzmann formula.
- Process of excitation does not distort the RDFs in the state $d^3\Pi_u$. The rotational temperature in the ground state can be calculated from the known relation between the rotational temperatures and rotational constants:
$$\frac{T_{rot}(X^1\Sigma_g^+)}{B_{rot}(X^1\Sigma_g^+)} = \frac{T_{rot}(d^3\Pi_u^-)}{B_{rot}(d^3\Pi_u^-)}$$
- The time of the rotational relaxation $\tau_R = (6-9) \times 10^{-5}$ s of the excited hydrogen molecules must be less than the residence time molecules in the ECR discharge $\tau_D = 10^{-4}$ s.
- Lifetime of the $d^3\Pi_u$ state ($\tau_e = (3-7) \times 10^{-8}$ s) should be noticeably larger than the time of rotational relaxation. This requirement is not satisfied for the pressure of 10^{-3} Torr and translational-rotational relaxation of molecules of hydrogen on hydrogen molecules in the plasma is not completed.
- Nevertheless if one will use the above formula, it give the gas temperature in the ECR region 620-650 K both for cooling with running water and vapor of liquid nitrogen. The gas temperature in the diffusion region is 430 ± 50 K.

Vibrational distribution functions (method of related intensities)

ECR region



Diffusion region



1. VDF in $d^3\Pi_u$ state are non-Boltzmann both in ECR and diffusion region and independent of cooling method. It is overpopulated at $v=1$.
2. Coronal model is valid for vibrational population of $d^3\Pi_u$ state.
3. Defined vibrational temperature of the ground state is 3000 - 3100 (\pm 550).
4. There is a problem in reconstruction of the vibrational temperature of the ground state from the measured distribution in $d^3\Pi_u$ state.

Vibrational temperature of the ground state

Vibrational temperature of the ground state of hydrogen molecules under different conditions

Experimental conditions					Results	Discharge	Ref.
T_{gr} , eV	p , Torr	N_{gr} , cm^{-3}	Method	T_{gr} , K	$T_v(X^1\Sigma_g^+)$, K		
1.3	15	$1.5 \cdot 10^{10}$	Spectr. of Comb. Scatt.	600±50	1800-2300(±50)	DC	[1]
1-2	2 1.5 8.0	10^{10} - 10^{12}	CARS	330±20 540±120 750±50	3400±540 2700±420	RF	[2-3]
0.65	$2 \cdot 10^{-2}$	$1.0 \cdot 10^{11}$	LIF	370±50	1600±100	Hot cathode	[4-6]
1.5	$3 \cdot 10^{-2}$	$5.7 \cdot 10^{11}$	CARS	470±50	1780±100	Hot cathode	[7-9]
1.5	19	$9.3 \cdot 10^{11}$	CARS	2150	2350±400	Microwave	[10]
3.1	0.1	$5.4 \cdot 10^{10}$	Emission spectroscopy	410±50	2400±100	Hollow cathode	[11]
3.1	10^{-3}	10^{10}	Emission spectroscopy	420-650	3100±400	ECR	Present paper

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Conclusions

1. Data obtained show that dipolar plasma source can produce conditions for effective generation of negative atomic hydrogen ions.
2. There is a problem in reconstruction of the vibrational distribution of hydrogen molecules in the ground state from the known distribution in $d^3\Pi_u$ state.
3. Two possible reasons: (1) violations of conditions of applicability of coronal model for $d^3\Pi_u$ state, and (2) uncertainty in the calculated factors of Frank-Condon for radiative transitions from this state. This problem requires detailed study.

**Thank you for your
attention**