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COMPARATIVE SPECTROSCOPIC TEMPERATURE MEASUREMENTS IN HYDROGEN HOLLOW CATHODE GLOW DISCHARGE

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Abstract. We report results of optical emission spectroscopy measurements of rotational T_{rot} and translational temperature T_{tr} of hydrogen molecules. The light source was hollow cathode glow discharge with titanium cathode operated in hydrogen at low pressure. The rotational temperature of excited electronic states of H_2 was determined either from relative line intensities of the R branch of the $GK \rightarrow B$ band or from the Q branch of the Fulcher-α diagonal band. The population of excited energy levels, determined from relative line intensities, was used to derive ro-vibronic temperature of the ground state of hydrogen molecule.

1. INTRODUCTION

The hollow cathode glow discharges (HCGD) operated in hydrogen and hydrogen seeded gas mixtires are of interest not only for fundamental research, but also for plasma assisted technologies (Da Silva et al. 2006). For both, fundamental and applied research, the knowledge of discharge parameter like the translational gas temperature T_{tr} of molecules and radicals is of particular importance since it determines the rate of chemical reactions. Different laser techniques were developed to measure T_{tr} of such an important molecule: coherent anti-stokes Raman scattering (CARS) see e.g. P´ealat et al. (1985), laser-induced fluorescence (LIF) see e.g. Chu et al. (1991) and optical emission spectroscopic (OES) technique see e.g. Goyette et al. (1996). If OES technique can be applied with standard laboratory equipment in visible region of spectrum it would be of great practical importance for monitoring and control of various hydrogen plasma assisted technologies.

Recently, the rotational, vibrational and translational temperatures of hydrogen molecules in HCGD (Majstorović et al. 2007) were determined using relative line intensities within Fulcher-α band. The rotational temperature of excited electron energy levels is determined from the Boltzmann plot of rotational line intensities, belonging to Fulcher-α diagonal bands ($d^3\Pi_u \rightarrow a^3\Sigma^+_g$) electronic transition; P, Q and R branches ($\nu'=\nu''=0$). The ro-vibronic temperature of hydrogen molecule ground state was evaluated from results of rotational line intensities, see below.

The aim of this paper is to compare the results of spectroscopic measurements of rotational and translational temperature in hydrogen HCGD from $GK^1\Sigma^+_g$, $\nu'\rightarrow B^1\Sigma^+_u, \nu''$ and Fulcher-α band.

2. EXPERIMENTAL

In this experiment, hollow titanium cathode with two symmetrically positioned kovar anodes operated in hydrogen is used as discharge source. The HC tube was 100 mm long with 6 mm internal diameter and 1 mm wall thickness. The discharge source is described in (Šišović et al. 2005, Majstorović et al. 2007). Here, only few important details related to the optical setup for spectra recordings will be described. The light from the discharge was focused with an achromat lens (focal length 75.8 mm) onto the entrance slit of 2 m focal length Ebert type spectrometer with 651 g/mm refection grating (the reciprocal dispersion of 0.74 nm/mm in first diffraction order). All spectral measurements were performed with an instrumental profile very close to Gaussian form with measured full half-width of 0.018 nm. Signals from CCD detector (29.1mm length, 3648 channels) are A/D converted, collected and processed by PC. During the discharge operation, HC was air cooled by a fan. The outer wall HC temperature (about 320 K) was controlled by a K-type thermocouple.

3. RESULTS AND DISCUSSION

Within the framework of model discussed in (Astashkevich et al. 1996) the logarithm of scaled rovibrational population density should be linear function of the rotational energy in hydrogen ground $X\,^1\Sigma^+_g$, $\nu=0$ vibronic state:

$$\ln N^*_{n'\nu'J'} \equiv \ln \frac{N_{n'\nu'J'}}{g_{a.s.}(2J'+1)\tau_{n'\nu'N'}} = -\frac{hcE_{X0J}}{kT_0(n',\nu')} + const. \tag{1}$$

In this case $T_0\,(n',\nu')$ is the rotational temperature of ground vibronic state determined from the rotational population density distribution in an excited (n',ν') vibronic state. The temperature $T_0\,(n',\nu')$ can be considered as a valid estimation of the ground state rovibronic temperatutre i.e. H_2 translational temperature T_{tr}.

In the recent study we used Q-branch ($\nu'=\nu''=0$) of the Fulcher-α diagonal bands as the most reliable for temperature estimation in HCGD (Majstorović et al. 2007). Now, we investigate the possibility of using R or P branches of the $GK^1\Sigma^+_g$, ν', $J'\rightarrow B^1\Sigma^+_u, \nu''$, J'' molecular system for temperature measurement.

Semilogarithmic plots of rotational population density distribution for $G^1\Sigma^+_g$, 0, J' ro-vibronic levels of H_2, evaluated from measured line intensities within R and P branch, have different dependencies on $J(J+1)$. The population distributions of lower levels used for these plots show different slope in comparison with higher levels see Fig. 1. The higher levels are most likely overpopulated in respect to the trend of low energy levels. The non-Boltzmann character of the plots prevented reliable temperature measurements. Therefore, another attempt is made to estimated temperature, but now, from the Boltzman plot of R0-R6 lines.

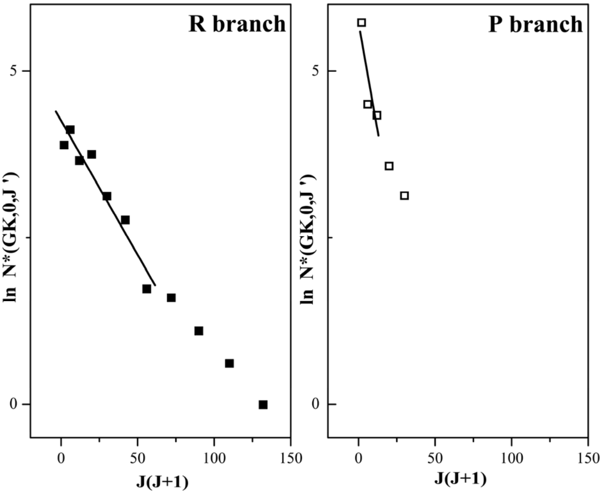

Figure 1: Semilogarithmic plot of rotational population density distribution for $G^1\Sigma^+_g$, 0, J' rovibronic levels of H_2 calculated from measured intensities for spectral lines of: (a) R branch and (b) P branch. Discharge conditions: titanium HC discharge in H_2 at p=3mbar; I=90 mA; U=400V.

The temperature obtained using limited number of spectral lines from R branch of $GK \rightarrow B$ transition agrees well with temperature measured from Q branch of Fulcher-α band (Majstorović et al. 2007). Thus, only values of the rotational temperature derived from the Q branch population of $d^3\Pi^-_u,0 \rightarrow a^3\Sigma^+_g,0$ and from R branch population of $GK^1\Sigma^+_g, 0 \rightarrow B^1\Sigma^+_u ,0$ were used in conjunction with (1) to determine T_0 of the ground vibronic state $X\ ^1\Sigma^+_g$ ($\nu = 0$) . The results in Fig. 2 show both temperatures which agree within estimated experimental uncertainties.

The result of this work at low pressure, based on intensity of limited number of spectral lines, show an agreement between rotational T_{rot}(d,0Q) and T_{rot}(GK,0R) temperatures as well as an agreement between T_0 (d,0) and T_0 (GK,0) of the ground vibronic state $X\ ^1\Sigma^+_g$ ($\nu = 0$) of hydrogen. The rotational temperatures derived from the population of Q branch of Fulcher-α and R branch of $GK \rightarrow B$ band, $\nu'=\nu''=0$ were (480 ± 70)K and (503 ± 70)K, respectively. The molecular hydrogen temperatures $T_0(d^-,0)$ and $T_0(GK,0)$ at the axis of Ti HCGD is (980 ± 100)K and (1050 ± 100)K, respectively. Under present experimental conditions the temperature T_0 for the ground vibronic state $X^1\Sigma_g^+(\nu=0)$ of hydrogen molecule is two times larger than the rotational temperatures of excited states $d^3\Pi_u$ and $GK^1\Sigma_g^+$.

For quick measurement of rotational and ground state T_0 temperature monitoring it is necessary to use first five lines of R-branch of $GK \rightarrow B$, ($\nu' = \nu''$=0) band.

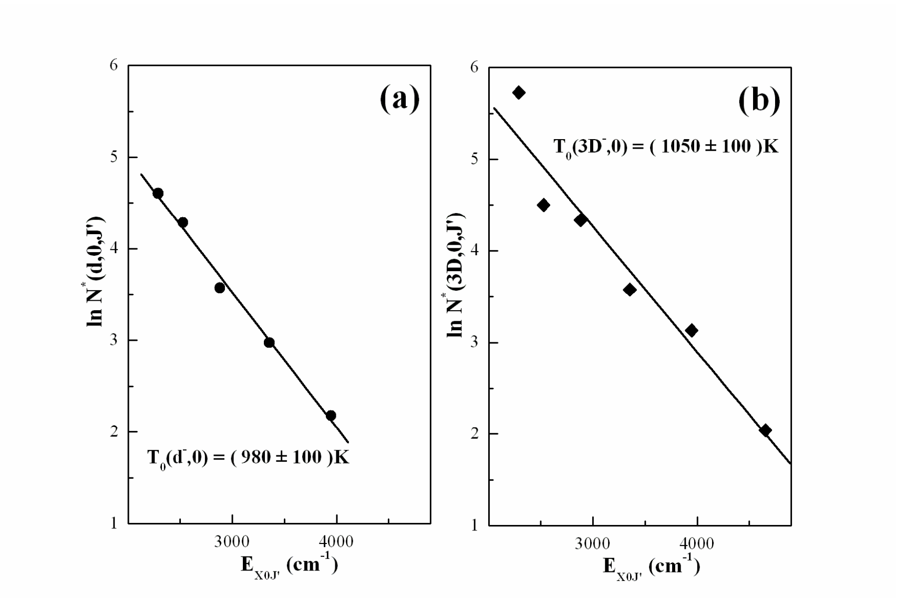

Figure 2: Plots of rotational levels population densities versus rotational energy of the molecular hydrogen ground state ($X\,^1\Sigma^+_g$, 0) of: (a) $d^3\Pi^-_u$,0 and (b) $GK^1\Sigma^+_g$, 0. Discharge conditions: same as in Fig. 1.

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