SPECTROSCOPIC TEMPERATURE MEASUREMENTS IN A H_2 MICROWAVE DISCHARGE

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Gas temperature and rotational temperature T_{rot} in a H_2 discharge are compared. The gas temperature is obtained by Doppler broadening of well defined atomic and molecular line giving atomic T_H and molecular T_{H2} temperature respectively.

Experiment.

Rotational experimental set-up used to study the gas temperature of H_2 discharges is described in [1, 2]. The plasma is created in a quartz tube (inner diameter being equal to 8mm or 16 mm). The microwave power is brought to the plasma at the gap. Photons emitted by the plasma are collected from the plasma via an optical fiber to the entrance slit of a spectrometer. We have used a monochromator JY HR1000 (1000 mm focal length) equipped with a Hamamatsu R928 photomultiplier and a 1200 groves/mm grating. The slits are 10 µm wide for T_{rot} (first order, resolution of 0.019 nm at 500 nm) and 20 µm wide for Doppler broadening temperatures measurements (second order, resolution of 0.013 nm at 500 nm). To improve the precision of Doppler measurements, experiments have been performed using a Fourier Transform Spectrometer (FTS) Bruker IFS 120HR providing a very high resolution.

Temperature measurements.

Rotational temperature measurements have been determined from the Fulcher α rotational spectrum ($d^3\Pi_u$ (v=0) --> $a^3\Sigma_g^+$ (v'=0), Q Branch : 612.0 nm to 616.5 nm).

For $d^3\Pi_u$ state, the spin-orbit interaction is well described by Hund's case b [3]. Therefore, assuming a Boltzmann distribution of the rotational levels, we have :

$$I_{emi} \propto (2K+1)(2\Gamma+1).e^{\frac{-B_v K(K+1)hc}{kTrot}} \qquad (1)$$

$\Gamma = 0$ and 1 for K even and odd respectively ($2\Gamma+1$: is a statistic weight). The rotational temperature T_{rot} is determined by plotting $\ln(I_{emi}/(2K+1).(2\Gamma+1))$ as a function of K(K+1). T_{rot} is inversely proportional to the slope ($B_v.h.c/k.T_{rot}$) of this straight line.

We have estimated the temperature of H_2 molecules (T_{H2}) and H atoms (T_H) using Doppler broadening of molecular and atomic line intensities. Two lines were considered:

- the singulet-singulet transition line $G^1\Sigma_g^+ \rightarrow B^1\Sigma_u^+$ (the J=2 and J=4 lines of the R Branch from 461.5 nm to 464.0 nm for v=v'=0 and $\Delta\Lambda=0$), since there is no fine structure ($\Lambda=0$ and S=0), and the hyperfine structure can be neglected.
- the balmer β line (Hβ) because the wavelength of these lines are close to the $G^1\Sigma_g^+ \rightarrow B^1\Sigma_u^+$ molecular system. For this latter, the fine structure has to be considered [4].

The measured broadening is defined by the FWHM $\Delta\lambda_m$. The apparatus function has a gaussian shape whose FWHM is $\Delta\lambda_{app}$. The FWHM Doppler broadening $\Delta\lambda_D$ is :

$$\Delta\lambda_D^2 = \Delta\lambda_m^2 - \Delta\lambda_{app}^2 \qquad (2)$$

The corresponding temperature (in °K) is [5] : $T = M.\left[\frac{\Delta\lambda_D}{\lambda}.\frac{1}{716.10^{-9}}\right]^2 \qquad (3)$

with the molar mass M (M = 2 g for H_2), and λ is the wavelength of the transition.

Results.

T_H, T_{H2}, and T_{rot}, decrease from the gap to the end of the plasma at 1Torr, 750W in a 8/10 quartz tube [Fig. 1]. This axial evolution of the gas temperature is explained by the fact that the electron density (and therefore the microwave power density) decreases along the plasma column, as shown previously [2].

a) Signification of rotational temperature.

T_{rot} is calculated in considering the rotational constant Bv* of the excited state in equation (1). Experimental results show that T_{rot} is systematically much lower than T_H and T_{H2} [Fig. 1]. T_{rot} can be considered as the gas temperature when the destruction frequency of the $d^3\Pi_u$ state ν^* is lower than the neutral/neutral collision frequency ν_{coll} in such a way that the thermalisation of this upper state is made possible. From [6], $\nu_{coll} / P = 8.10^6\ s^{-1}Torr^{-1}$. The emission probability of $d^3\Pi_u$ is $\nu^* = 4.\ 10^7\ s^{-1}$. Thus, at 1Torr, $\nu_{coll} < \nu^*$ and there is no equilibrium of rotational distribution. If there are not any collision between the gas and excited molecules before the molecular desexcitation (case of low pressures), the rotational distribution of $d^3\Pi_u$ reflects the ground state population, provided the excited state is created by electronic collision without modification of molecular rotational energy, and that there are not any radiative cascades from upper states to populate $d^3\Pi_u$ state. Under these conditions, the rotational constant B_v^g of the ground state has to be considered instead of the rotational constant Bv* of the $d^3\Pi_u$ state ($B_v^g = 2$ Bv*).

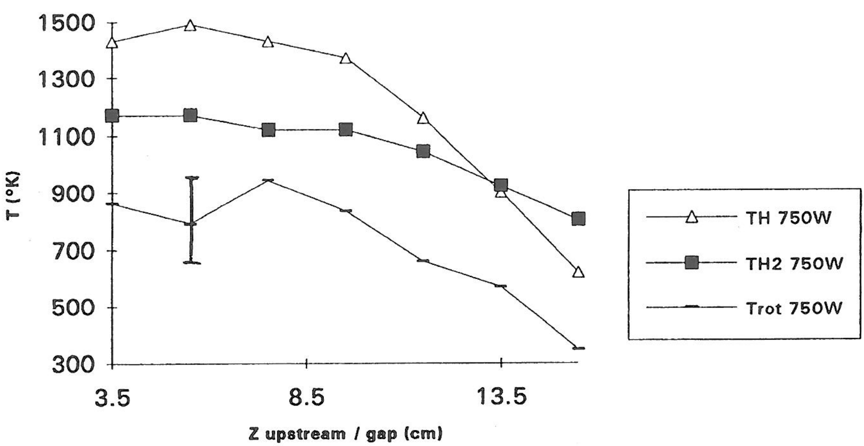

Figure 1 : T_H, T_{H2}, and T_{rot} along the plasma column at 1Torr in a 8/10 quartz tube.

To check this, experiments at lower pressure (0.5 Torr) were performed [Fig.2]. Indeed, T_{H2} is very close to $2xT_{rot}$, the rotational distribution of the excited state is the image of the ground state. However, at 1Torr, $T_{H2}=1.7T_{rot}$ and therefore T_{rot} can not be considered as the gas temperature, because there are a few collisions, but not enough to accomplish a total redistribution of the rotations.

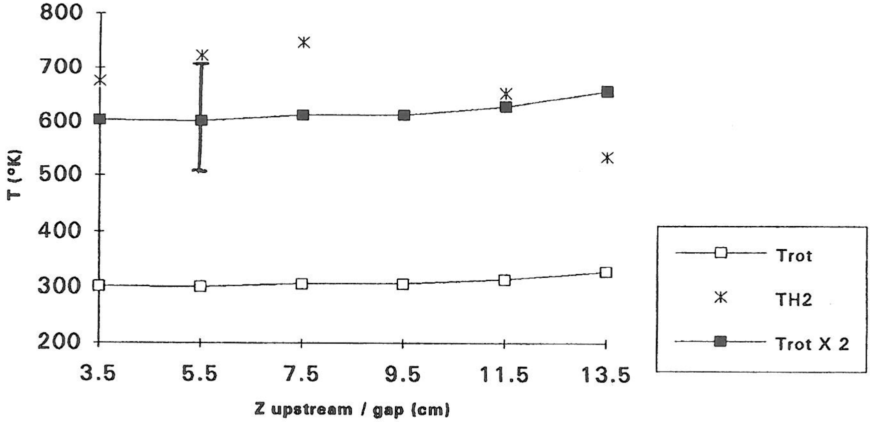

Figure 2 : Comparison at 0.5Torr and 550W between T_{rot} and T_{H2}.

b) Comparison between atomic and molecular kinetical temperature.

At 1 Torr, T_H is greater than T_{H2} **[Fig. 3]**. Moreover, we have shown in [7], that the dissociative excitation does not occur under our experimental conditions, so that the main process to create excited H atoms is the direct excitation from the ground state. It seems therefore that both excited and ground state H atoms have the same kinetical energy, so that the temperature determined from Doppler broadening measurements can be considered as the kinetical temperature of the whole H atom population. To explain why $T_H > T_{H2}$, one as to consider that H atoms are produced by electron impact dissociation of the molecules with an average kinetical energy of about 3.5 eV. Hence, these hot atoms relax part of their energy, by atom-molecular collisions, before they are lost by wall recombination. The molecular-neutral collision frequency for the relaxation has to be compared with the loss frequency of the H atoms in the discharge. Under our low pressure conditions, the collision frequency is to low to achieve a complete equilibrium between T_H and T_{H2}.

Moreover, one can therefore expect that, the higher is the H_2 fraction, the more efficient is the relaxation of hot atoms and the cooler is the gas. In order to check this, atomic temperature was measured both in continuous and pulsed power, for the same instantaneous power (750W).

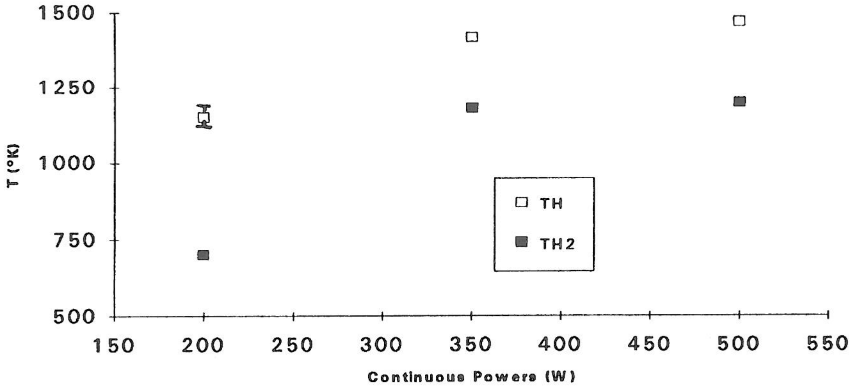

Figure 3 : Comparison for different continuous powers between T_H and T_{H2} at 1Torr in a 16/19 quartz tube with the FTS.

c) Comparison of T_H between continuous and pulsed power.

We have shown recently that the H atom molar fraction is much higher in a pulsed power regime that in a continuous one [7]. T_H is higher in the pulsed discharge (Fig 4).

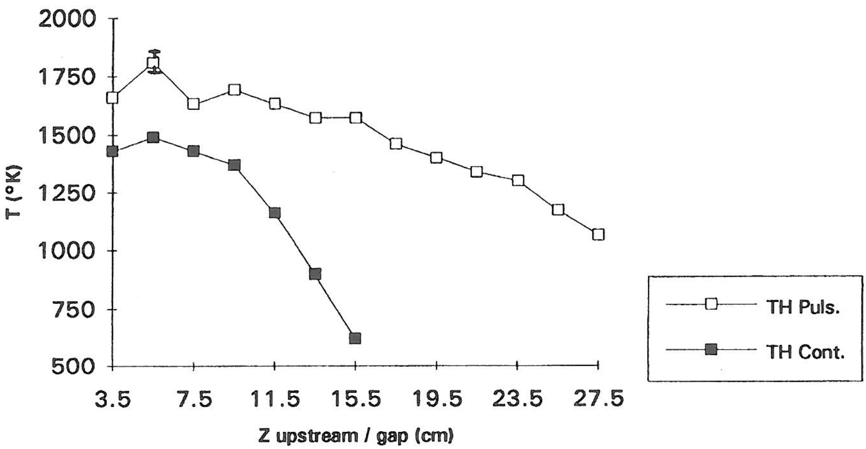

Figure 4 : Comparison at 1Torr between continuous (750W) and pulsed discharge (same high power) for T_H with the FTS in 16/19 quartz tube.

However, the length of the plasma is greater in a pulsed regime, since the instantaneous volumic power density injected in the plasma column is more important for the continuous regime than for the pulsed regime. Then, one would have expected the gas temperature being higher in case of continuous power. To explain this paradox, we have to take into consideration the whole power balance in the plasma slab. In this case, the H atom molar ratio is higher than 80 % [7]. The H atoms are less efficiently cooled by the molecules so that T_H is higher in the pulsed discharge than in the continuous power.

Conclusion.

We have shown that T_{rot} is not equal to the gas temperature under our discharge conditions.

T_H is higher than T_{H2} because this seems to be related to the H atom relaxation process.

The H atom temperature is higher in a pulsed power discharge : this might be related to a most important H atom density in the pulsed regime.

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