MEASUREMENT OF VIBRATIONAL POPULATIONS IN HYDROGEN PLASMA BY COHERENT ANTI-STOKES RAMAN SCATTERING.

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ABSTRACT

Coherent anti-Stokes Raman Scattering (CARS) has been applied to the measurement of vibrational populations in a low-pressure H_2 plasma. For an electron density of 2×10^{11} cm^{-3} and a total pressure of 0.13 mbar, the rotational temperature is found to be 475 K. The population of vibrational states $\nu = 0$, 1 and 2 has a non-Boltzmann distribution.

I - INTRODUCTION

The production of negative deuterium ions is of interest because it is a necessary step towards injection of high energy neutral beams into controlled fusion devices. Evaluation of the capabilities of negative deuterium (and hydrogen) ions sources is therefore essential for optimizing the design of a fusion reactor.

It has been observed recently that low-pressure hydrogen plasmas are characterized by negative ion densities much higher than expected from computations based on conventional dissociative attachment cross-sections (1).

Recent theoretical (2) and experimental (3) work has shown that an important increase of cross-section is obtained when dissociative attachment is produced from hydrogen molecules on vibrationally excited levels. In order to verify if the high negative ion densities observed were due to the relative importance of the population of vibrationaly excited molecules, the measurement of this population had to be performed.

Only optical techniques can perform nonintrusive measurements in these highly tenuous media. Two techniques are contending for this task : fluorescence and coherent anti-Stokes Raman scattering (CARS). However, owing to the high luminosity of the plasma under study, CARS is better adapted because of its excellent background light rejection capability. CARS, which had been proposed some time ago for temperature and concentration measurements in reactive media (4,5) has been used here for measuring precisely the density of vibrationally excited hydrogen molecules in low pressure plasma.

II - EXPERIMENTAL SET-UP

A - The plasma generator

The plasma is produced in a magnetic multipole (5) which is operated under a bell jar (Fig. 1). The thermionic electrons emitted from a thoriated tungsten filament are accelerated to $V_d = 90$ eV and ionize the hydrogen gas, flowing at a pressure which can be varied between 0.1 and 300 mbars. The gas pressure is measured using a Pirani gauge and a Penning gauge. At low pressure the magnetic field, created by permanent magnets near the walls of the device, confines the primary electrons and thus a high gas ionization efficiency is achieved. The multipole walls are made of stainless steel and are cooled by circulating water. Openings in the walls allowed the laser beams to cross the plasma.

The electron density and electron temperature of the plasma in the region traversed by the laser beams have been measured using a 4-mm diam plane probe. For this purpose the multipole structure was transferred to another glass vessel.

The effect of gas pressure P upon the electron density n_e and electron temperature kT_e has been studied in the pressure range between 3.10^{-3} and 3.10^{-1} mbars, while maintaining a constant discharge power (i.e., a constant discharge voltage of 90 V and a constant discharge current of 3 A). The results are shown in Fig. 2. It was found that the electron density attains a maximum of about 2.5×10^{11} cm^{-3} at a pressure of 1.5×10^{-2} mbars.

The variation of the electron temperature with gas pressure is also shown in Fig. 2. The electron temperature decreases from 0.75 to 0.15 eV when the pressure is increased in the studied range.

The cross section σ_{01} for excitation on the state $v = 1$ by electron collisions with molecules in the state $v = 0$, measured by Ehrhardt et al (6), has been used to estimate the reaction rate for electron temperatures below 1 eV (7). The reaction rates corresponding to the measured electron temperatures have been used to calculate the production rate ϕ of the states $v = 1$. Note that this production rate is maximum at a pressure of about 0.1 mbar (Fig. 2).

B - THE CARS SPECTROMETER

The CARS set-up used has been described elsewhere. The characteristics are as follows : the laser beam has a maximum energy of 170 mJ at 532 nm, emitted in 15 ns. The tunable Stokes beam has an energy varying from 0.5 to 1.5 mJ in the spectral domain from 685 to 650 nm, which has been explored in this study. The linewidth is less than 10^{-2} cm^{-1} at 532 nm and less than 0.1 cm^{-1} for the Stokes. The beams are focused at the center of the magnetic multipole. The transverse resolution is of the order of 60 µm and the longitudinal resolution is about 80 mm.

The detection of CARS signal is performed as described in Refs. 8 and 9 but use of optical fibers reduces the e.m interference problems.

C - DATA REDUCTION

The anti-Stokes power P_3 collected in a CARS experiment is approximately given by :

$$P_3 = K \left|\chi^{(3)}\right|^2 P_1^2 P_2 , \quad (1)$$

where K is a proportionality constant, and P_1 and P_2 are the pump powers at the laser, ω_1, and Stokes, ω_2, frequencies, respectively. The CARS nonlinear optical susceptibility $\chi^{(3)}$ associated with an isolated Q branch transition can be given the expansion (9).

$$\chi^{(3)} = K'N\left[\rho(v,J) - \rho(v+1,J)\right](v+1)\frac{d\sigma}{d\Omega}\, h(\omega_1 - \omega_2), \qquad (2)$$

where K' is a proportionality constant
N is the number density
$\rho(v,J)$ is the probability of occupation of the rovibrational state v J (lower state of the Raman transition under probe)
and $\rho(v+1,J)$ is that of the upper state.

The $v+1$ factor reflects the growth of the cross-section as a function of vibrational quantum number.

$\frac{d\rho}{d\Omega}$ is the spontaneous Raman-scattering cross-section for the $v = 0 \rightarrow 1$ transition.

$h(\omega_1 - \omega_2)$ is the lineshape function, cast in a form adequate for Doppler-broadened Raman lines. Actually, the dye laser linewidth is larger and the vibrational lineshape is broadened by an instrumental effect.

The computation taking into account this effect is given in Ref. 9, together with a discussion of the influence of rotational temperature changes, and also with a description of the special procedure used when computing the population from the recording of very faint lines in the presence of stray light which gives a constant background pedestal. Such procedure is made necessary by the non-linear nature of the relationship giving the populations from the values of the measured powers.

III - RESULTS AND DISCUSSIONS

The instrument was first tested and calibrated on the fundamental Q lines of low-pressure hydrogen at room temperature. The conditions for optimal vibrational excitation in the discharge were then established by changing the pressure at constant discharge power ; the value of 0.13 mbar was selected (9).

Figure 3a) gives the profiles of fundamental Q lines without the discharge, together with calculated peak intensities for Boltzmann equilibrium at 290 K shown as horizontal bars. The uncertainty on rotational temperature measurements is ± 5 K by eyeball fitting (i.e., calculated peak line intensities at 285 and 295 K depart significantly from the measured ones). The vertical scale in number densities of rotational state populations is adjusted to match the reading of the pressure gauge assuming a temperature of 290 K.

Figure 3b) gives the experimental profiles of the same lines at the same pressure, but under the optimum discharge conditions. The result of a calculation for rotational Boltzmann equilibrium at 475 K, which gives the best fit, is also shown. The uncertainty on the temperature measurement is here ± 10 K. The vertical calibration is given in number densities.

The Q(1) lines for $v = 1$ and $v = 2$ are plotted in Fig. 4. The $v = 3$ line could not be detected. The vertical scales are given in units of number densities. They were adjusted like those of Fig. 3, with due corrections as discussed in Ref. (9).

If we call $n(v)$ the total number density for vibrational state v and if we assume that a Boltzmann rotational equilibrium is established at 475 K for all vibrational states, we have the following results :

$$\begin{aligned} n(0) - n(1) &= 1.86 \times 10^{15}\ \mathrm{cm}^{-3}, \\ n(1) - n(2) &= 4.04 \times 10^{13}\ \mathrm{cm}^{-3}, \\ n(2) - n(3) &= 3.63 \times 10^{12}\ \mathrm{cm}^{-3}, \\ n(3) - n(4) &\leqslant 1.0 \times 10^{12}\ \mathrm{cm}^{-3}, \end{aligned}$$

which are good to a relative accuracy of ± 10 % approximately. From these results, we deduce $n(0) \simeq 1.9 \times 10^{15}\ \mathrm{cm}^{-3}$. The precise determination of $n(1)$ and $n(2)$ is impossible without the knowledge of $n(3)$ or without an assumption on the type of equilibrium which is established among the vibrational states. On the basis of a theoretical calculation by Garscadden and Bailey (10) which takes anharmonic pumping into account, we assume $n(3) = 0.8 \times 10^{12}\ \mathrm{cm}^{-3}$ from which we derive

$$\begin{aligned} n(2) &\simeq 4.4\ 10^{12}\ \mathrm{cm}^{-3}, \\ n(1) &\simeq 4.5\ 10^{13}\ \mathrm{cm}^{-3}. \end{aligned}$$

Therefore about 2.5 % of the H_2 is vibrationally excited. Furthermore, molecular H_2 including excited states constitutes 88 % ± 7 % of the plasma. We thus deduce that ions, atoms, electronically excited H_2, and other vapors which make up the balance range from 5 to 19 % mole fraction. Our experimental uncertainty on this quantity would be reduced appreciably by taking averages over a larger number of laser shots while recording the spectra.

Several other remarks can be made regarding these results.

(i) - The close agreement between the observed peak line intensities on $v = 0$ in the plasma and the calculated Boltzmann factors for 475 K indicates that our spatial resolution is adequate and that the plasma is uniform over the active length of the laser beams.

(ii) - Our detection sensitivity for $v = 2$ is on the order of 0.5×10^{-7} bars ; appreciable gains would be obtained by reducing the intensity of the parasitic signal generated in the windows and enhancing the transmission of the monochromators. Under these conditions we could monitor the populations of up to $v = 5$ assuming that a Treanor-like equilibrium is established.

IV - CONCLUSION

The vibrational excitation of H_2 in a discharge at low pressure has been measured with a fair degree of accuracy. Some technical improvements should enable us to monitor the populations of states of still higher energy and to confront the results of computational models. A systematic analysis of the influence of pressure and static temperature as well as discharge conditions and wall collisions should also be conducted so that useful conclusions can be drawn on pumping and relaxation rates.

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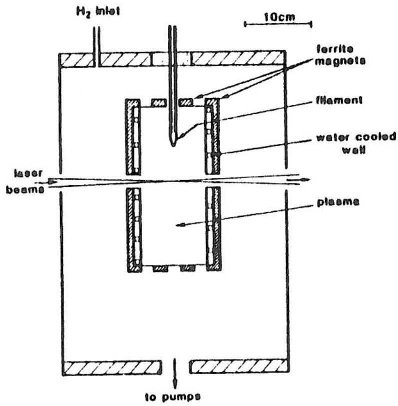

Fig. 1 — Schematic of the plasma generator, drawn to scale.

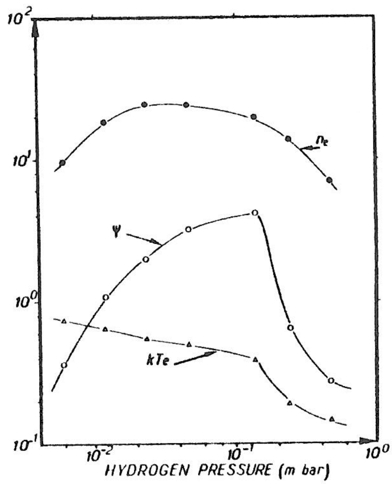

Fig. 2 — Variation with gas pressure of the electron density n_e (10^{10} cm^{-3}), the electron temperature kT_e (eV) and the production rate ψ (mbar s^{-1}).

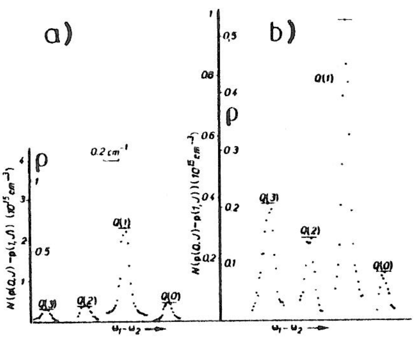

Fig. 3 — Profiles of lines Q(0) to Q (4) for the $\nu = 0 \rightarrow 1$ fundamental transition of neutral H_2 : (a) without the discharge ; (b) with a discharge of 90 V, 3 A. The strongest portions of the lines only are shown, in steps of 0.02 cm^{-1}, and with 10 consecutive measurements averaged at each point. The spectral resolution of the dye laser was 0.07 cm^{-1}.

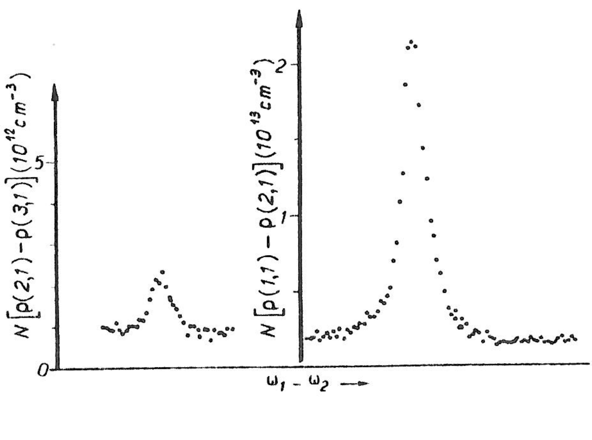

Fig. 4 — Profiles of excited states lines, with 30 measurements averaged at each point.