NITROGEN ACTIVE SPECIES $N, N^*(^2D, ^2P), N_2(A^3\Sigma_u^+)$ IN GLOW DISCHARGE

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ABSTRACT

Concentrations of the atomic nitrogen metastable states $N^*(^2D, ^2P)$, the molecular metastables $N_2(A)$ and the atomic nitrogen ground state $N(^4S)$ have been measured in d.c. nitrogen glow discharges. A kinetic model shows that the atomic metastable states are created by electronic collisions on N_2 and $N(^4S)$ and also, for the $N^*(^2P)$ state, by the $N_2(A)+N(^4S)$ reaction. We deduce the coefficient of creation of the metastable states $N(^2D, ^2P)$ by electronic collisions on N_2.

1. INTRODUCTION

The long lived reactive species of nitrogen play an important role in the chemistry of the upper atmosphere and in laboratory plasmas. The more important are: the atomic nitrogen in ground state $N(^4S)$, in the metastable states $N^*(^2D)$ and $N^*(^2P)$ and the molecular nitrogen in the first metastable state $N_2(A)$. The concentrations of these reactive species are measured in direct current glow discharges in pure nitrogen.

2. EXPERIMENTAL

The measurement is performed in the positive column of a d.c. glow discharge. The internal diameter of the discharge tube is R=1cm, the pressure is in the range from 5×10^{-2} to 2 Torr and the electrical current can reach 120 mA. In this positive column, the electron density is measured by a microwave cavity, the neutral gas temperature is deduced from the rotational spectrum of the second positive band of N_2 and the axial electric field is measured by electrostatic probes.

The metastable concentration is measured by optical absorption and the atomic nitrogen $N(^4S)$ by NO titration.

For the optical absorption two experimental devices are used.

An atomic line optical absorption method gives the $N^*(^2D)$ and $N^*(^2p)$ densities (figure 1). The absorbed lines are in the vacuum ultraviolet: 1492 Å for $N^*(^2D)$ and 1742 Å for $N^*(2P)$. A detailed description of this experimental device can be found in the reference [1].

The molecular metastable $N_2(A)$ is measured by optical absorption of the (1-0) band of the nitrogen first positive system. The optical arrangement (figure 2) [2] is a two light beams. The light emitted by the source is divided in two beams modulated in phase opposition. One of them is weakly absorbed in the positive column and the other one is the reference beam. The two beams are focused on the entrance slit of a monochromator. The output signal of the photomultiplier is detected by a lock-in amplifier. This device can measure absorption down to 0.1%.

The $N(^4S)$ ground state nitrogen is measured by NO titration in a low speed ($v<7ms^{-1}$) afterglow. The upstream discharge have the same characteristics than the previous steady discharges. By varying the gaz velocity, we check that the $N(^4S)$ concentration has reached his steady value.

3. EXPERIMENTAL RESULTS

3.1. - Plasma parameters

The electron density is nearly proportional to the discharge current for a fixed pressure and increases with pressure for a fixed current. At 2 Torr and 50mA, the electronic density n_e is $2x10^9 cm^{-3}$.

The electric field, for a fixed pressure, decreases when discharge current increases, but it is constant at low pressure. In our experimental conditions E varies form 4 to 60 Vcm^{-1}. The neutral gas temperature increases with the current but is nearly constant and equal to 300 K at low pressure.

From this measurements we found that the ratio E/N (electric field over neutral gas density) is constant for a fixed pressure and decreases from $3x10^{-15}$to $6x10^{-16}Vcm^2$ when the pressure varies from $5x10^{-2}$ to 2 Torr.

3.2.- Atomic metastable $N^*(^2D)$ and $N^*(^2P)$

The measurements of $N^*(^2D)$and $N^*(^2P)$ concentrations have been published previously [1]. The densities increase linearly with the current and reach a saturation, of about $6x10^{10}cm^{-3}$ for N^* (^2D) and $10^{11}cm^{-3}$ for $N^*(^2P)$.

3.3. - Molecular metastable $N_2(A^3\Sigma_u^+)$

The optical absorption is proportional to the $N_2(A)$ concentration. The calculation of the proportionality factor is given in reference [2]. The figure 3 shows the $N_2(A)$ concentrations as a function of discharge current and pressure. The density reaches a saturation value of $1.5x10^{12}cm^{-3}$;

3.4. - Ground state atomic nitrogen $N(^4S)$

The $N(^4S)$ concentration is measured for pressure from 0.5 to 2.2 Torr. Even for current up to 120mA, the $N(^4S)$ density does not reach a saturation but increases whith the current; it is equal to $10^{15}cm^{-3}$ at 2.2 Torr and 120mA.

4. BALANCE OF THE ACTIVE SPECIES

The concentrations of the active species, in steady state conditions, are given by the balance equation.

From the experimental results whe found that the main processes are the following :

- $N_2(A)$ is created by electronic collisions on the ground state molecule N_2 and lossed by diffusion to the wall, reaction with $N(^4S)$ and pooling reaction;
- $N(^4S)$ comes from dissociation of N_2 by electronic collision and vibration-vibration transfert [3] and is lossed by the $N_2(A)$ reaction;
- N^* (^2D) and $N^*(^2P)$ are created by electronic collisions on N_2 and $N(^4S)$ and for N^* (^2P), by the $N_2(A)$ + N (^4S) reaction.

From the balance equations we deduce the coefficient of creation of $N^*(^2D,^2P)$ by electronic collisions on N_2. The results are presented on the figure 4 as a function of the experimental E/N ratio.

5. CONCLUSION

Creation an destruction mechanisms for some metastable species and N(^{4}S) were studied in this work and numerical coefficients derived. Future work should also include the detailed analysis of the behaviour of individual vibrational levels.

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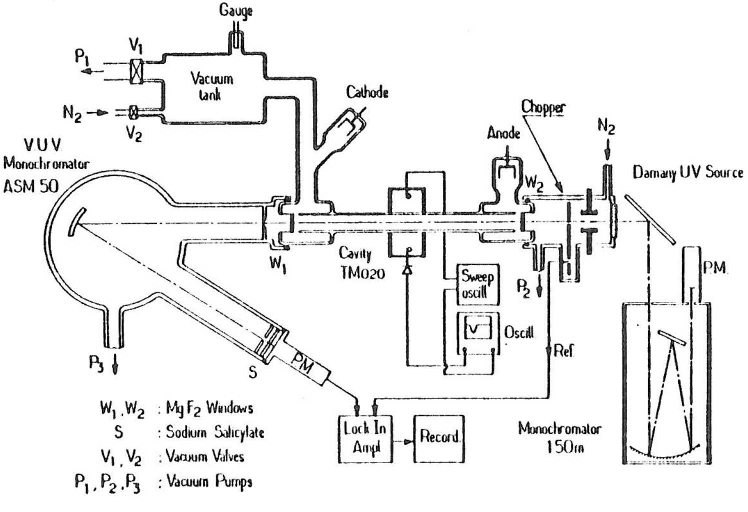

FIGURE 1

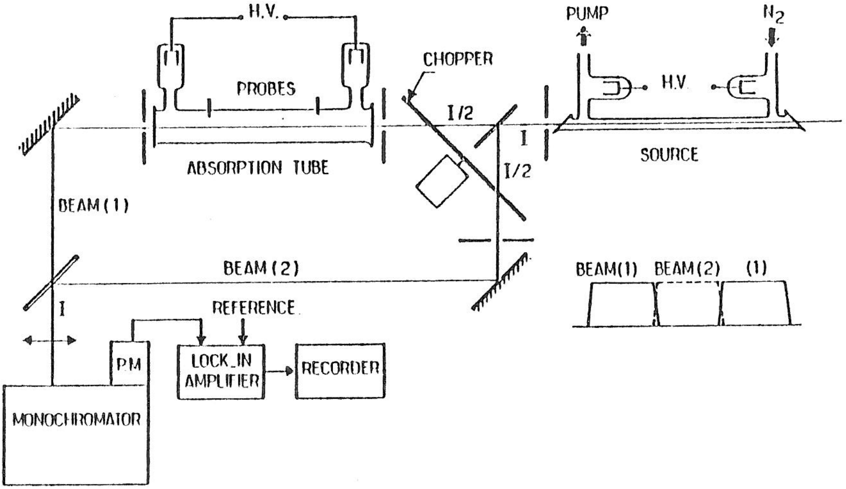

FIGURE 2

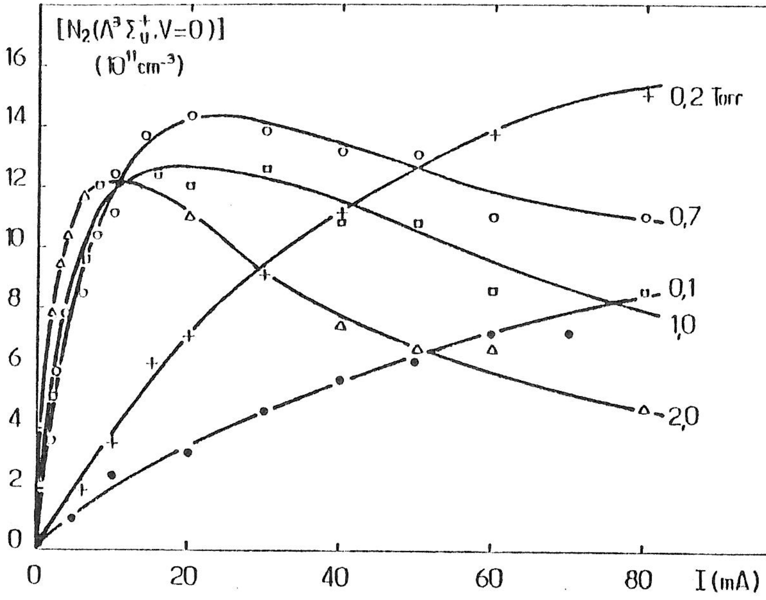

FIGURE 3

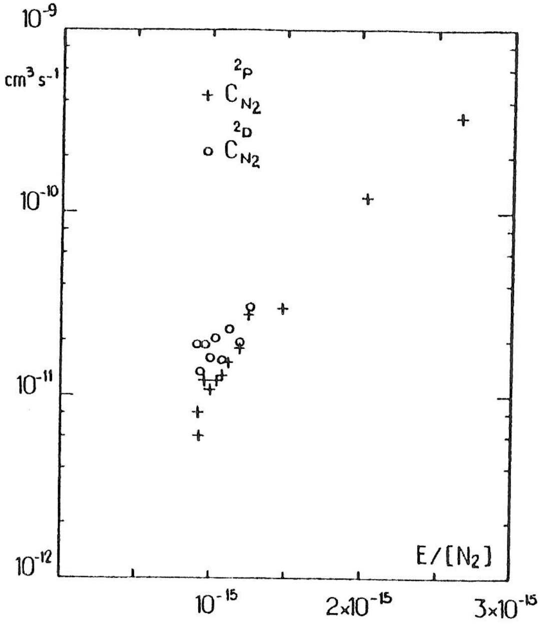

FIGURE 4