Optical emission spectroscopy study of N_2 dc discharges at low pressure.

G. Cicala [1], E. De Tommaso [1], A. C. Rainò[2] and A. Boggia[3]

[1] *IMIP-CNR (Istituto di Metodologie Inorganiche e dei Plasmi, Consiglio Nazionale delle Ricerche), Via Amendola 122/D, 70126 Bari, Italy*

[2] *Dipartimento di Fisica/INFN Sezione di Bari, University of Bari, Via E. Orabona 4, 70126 Bari, Italy.*

N_2 dc discharges at 3 Torr and discharge current of 17.5-100 mA have been studied by OES at four positions: 25, 75, 125 and 175 mm far from the cathode. The distribution of the excited species of N_2 second and first positive systems and N_2^+ first negative system, strongly changes going from the cathodic dark space to the positive column. In the cathodic dark space (25 mm) the emissions of N_2 SPS and FPS systems are weaker than those in positive column (75, 125 and 175 mm), whereas the emissions of N_2^+ FNS prevail at positions very close to the cathode (3 mm) and decrease down to disappear by going away from the cathode. The analysis of emission intensities of SPS sequence Δv =-2 allows to determine the vibrational temperature (T_v), only if the emissions are not too weak. Reliable values of T_v are found to increase from 3250 to 5750 K by increasing the discharge current from 17.5 to 100 mA and to maintain constant in the the positive column.

1. Introduction

The study of low pressure discharges has been and still has many interest in different field such as aviation, astronautics, surface treatment and thin film deposition. So studying the relationship between the plasma properties and discharge parameters is important both in plasma science and plasma techniques. In this study we have used the non-invasive optical emission spectroscopy (OES) for investigating the distribution of the various excited species along the interelectrode gap.

In this contribution we report on the results of optical emission spectroscopy (OES) at four positions: 25, 75, 125 and 175 mm far from the cathode by varying the discharge current from 17.5 to 100 mA . In the same experimental conditions the vibrational temperature has been determined.

2. Experimental section

The schematic diagram of the experimental setup is given in Fig. 1. It comprises a pyrex tube (800 mm long and 43 mm inner diameter) in which a non equilibrium dc discharge has been ignited by a dc supplier (Alintel SHV 1000, 200mA/5000V) between a pair of annular electrodes. The distance between the two electrodes has been 200 mm and the electrode polarity can be reversed.

The emission radiation in the range 200-1000 nm has been collected by an optical fiber and recorded with an AvaSpec 2048 spectrometer (focal length of 75 mm and wavelength resolution of 0.8 nm) equipped with a CCD array. In spite of the great operative range of the spectrometer, the UV emission below 315 nm has been cut off because of the pyrex glass tube.

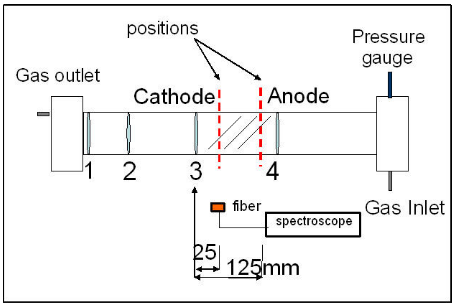


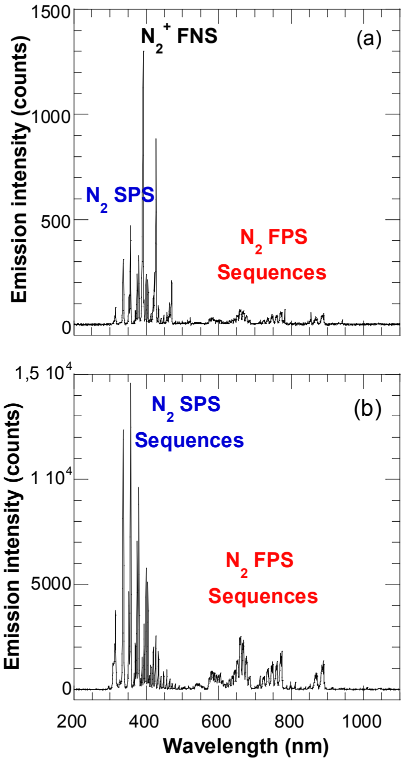
Figure.1 Schematic of experimental apparatus.

The optical fiber can be moved to any position parallel to the axis of discharge tube, from the cathode to about the full length of the interelectrode gap. If the polarity has been *3°Cathode-4°Anode* the positions respect to the cathode are 25 mm and 125 mm, by reversing the electrode polarity, *3°Anode - 4°Cathode,* the positions respect to the cathode became 75 mm and 175 mm. Thus, the investigated four positions have been 25, 75, 125 and 175 mm with respect to the cathode.

The N_2 flow has been fixed at 200sccm by MKS gas flowmeter, the pressure has been maintained constant at 3 Torr and the discharge current has been varied from 17.5 to 100 mA.

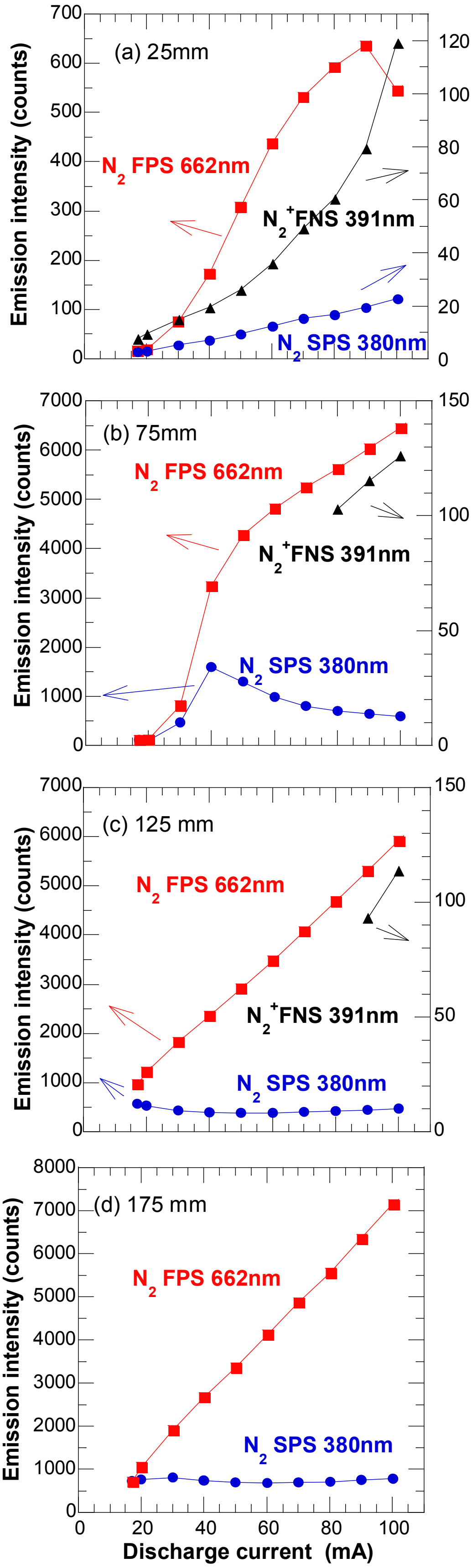
3. Results and discussion

Figures 2a-b show the OES spectra gathered in two distinct regions very close to the cathode and anode of N_2 dc discharge. The spectrum near the cathode (Fig. 2a) is characterized by the presence of emitting vibrational bands of the second positive system (SPS $C^3\Pi_u - B^3\Pi_g$) and the first positive system (FPS $B^3\Pi_g - A^3\Sigma_u^+$) of the N_2 molecule, and the first negative system (FNS $B^2\Sigma_u^+ - X^2\Sigma_g^+$) of N_2^+ molecular ions. From Fig. 2a, it is clear that the emission intensities of N_2 molecules (SPS and FPS) are very low, whereas those of N_2^+ molecular ion (FNS) are dominant. In the anode region (Fig. 2b) the emission intensities of the SPS and FPS are one order of magnitude higher than those in the cathode region and the FNS system becomes undetectable by the spectroscope operating in the same conditions of Fig. 2a.



Figures 2a-b. OES spectra of N_2 dc discharges very close to (a) the cathode, 3mm, and (b) the anode, 1mm, obtained at 3 Torr and 50 mA.

Figures 3a-d show the evolution of emission intensities of SPS and FPS of N_2 and FNS of N_2^+ versus discharge current at four fixed positions along the interelectrode gap: 25, 75, 125, 175 mm from the cathode. In each of these three systems we follow the emission of one wavelength: 380.4, 662.4 and 391.4 nm, respectively. Near the cathode (Fig. 3a) the SPS (380.4 nm) emission is very low and increases with the current, whereas in the positive column (75, 125 and 175 mm) it becomes 20-50 times higher; by keeping almost constant for all the current (see Figs. 3b-d). The excited N_2^+ ions are detectable at all the currrent only near the cathode (Fig. 3a) and absent a long way from cathode (Fig. 3d); in the positive column they appear only at the



Figures 3a-d Emission intensities of SPS (380.4 nm), FPS (662.4 nm) of N_2 molecule, FNS (391.4 nm) of N_2^+ molecular ion vs discharge current at (a) 25 mm, (b) 75 mm, (c) 125 mm and (d) 175 mm far from the cathode.

highest current (Fig. 3b-c). The dominant band along the positive column is the FPS: in all the positions this emission increases with the increasing current and strongly increases, one magnitude order, going from the cathodic dark space (Fig. 3a) to the glow of positive column (Fig. 3b-d).

From the relative values of the band intensities of a given band sequence a measure of the vibrational excitation can be obtained. The relationship between total band intensity and temperature, when a Boltzmann distribution is assumed, is the following [2]:

$$I_{em}^{v'-v''} \propto A_{v'-v''} \cdot \nu^{4}_{v'-v''} \cdot \exp\left[-\frac{G_0(v')\cdot hc}{kT_v}\right] \qquad (1)$$

where $A_{v'v''}$ is the transition probability i.e. Franck-Condon factor [3], $\nu_{v'v''}$ is the transition frequency and $G_0(v')hc$ is the energy of the vibrational level v'.

For example the relative intensities (divided by $A_{v'v''}$ times the fourth power of $\nu_{v'v''}$) of the SPS sequence (Δv=-2) have been plotted against $G_0(v')$ in a semilogaritmic plot and the vibrational temperature can be deduced from the slope of the experimental data. For each plot of the sequence the

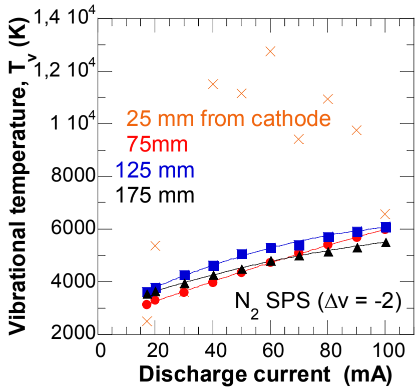


Figure 4. Vibrational temperatures as a function of discharge current at the four axial positions far from the cathode.

value of the correlation coefficient gives us a measure of the validity of our assumption, i.e. the levels are populated according to a Boltzmann distribution.

Figure 4 shows the vibrational temperature against discharge current at the four positions. In the positive column (75, 125 and 175 mm from the cathode) the vibrational temperature regularly increase with increasing current and the values of the temperature are almost the same for the three positions. At 25 mm from the cathode the trend is irregular and this behaviour is due principally to the very weak intensity emissions and a non Boltzmannian distribution.

The distributions of the excited N_2 and N_2^+ species representative of FPS, SPS and FNS systems, strongly change going from the cathodic dark space to the positive column because of the relative different excitation threshold energies 7.4, 11.1 and 18.7 eV and kinetic processes. However, since the physical phenomena occurring near the cathode constitute essential processes to the maintenance of dc discharge current, in the cathode region the presence of ions as detected by OES is justified. In fact, the high potential fall typical of the cathode region is responsible for the ionization processes. By going away from the cathode the extent of potential decreases and the ionization is allowed only at the highest current and not too far from the cathode. This explanation is consistent with the data shown above.

The vibrational temperatures in the positive column (75, 125 and 175 mm) increase with increasing the discharge current and this is principally due to the expected increase in the electron density with the current. These results agree with those reported in ref. [4].

Conclusions

Low pressure dc discharges of N_2 have been investigated by varying the discharge current from 17.5 to 100 mA. The optical emission spectroscopy has been used to detect the excited species at four axial positions far from the cathode. The distributions of excited N_2 molecules and N_2^+ molecular ions change at the various positions accordingly to the potential distribution along the interelectrode gap because of their different excitation threshold energy. The excited N_2^+, with the highest excitation energy (18.7 eV), indeed is observed in the region close to cathode, where the ion density is expected higher than that of the neutral species.

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