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# Atomic radiation from a recombining nitrogen plasma

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**Shock layer radiation from atomic lines can represent the main source of radiation in the afterbody region of a space capsule during Earth entry. Studying the kinetics of excited states of atomic nitrogen is therefore important to correctly predict its corresponding radiation. The CentraleSupélec plasma torch facility provides a stable equilibrium plasma and can be used to study plasma kinetics and radiation in a controlled environment. Previous experiments were performed to supply measurements of excited states of molecular nitrogen in a nonequilibrium recombining flow. We extend these measurements to atomic lines to get measurement of excited states of atomic nitrogen. We performed optical emission spectroscopy from VUV to NIR and obtain measurements of atomic nitrogen states above 10eV. The VUV region allow access to low energy states of nitrogen. A departure from Boltzmann distribution is observed for the high excited states and their densities tend to be equal to the Saha-Boltzmann distribution. This was previously predicted and is experimentally observed in this paper. These results may be used to validate kinetic codes.**

## I. Introduction

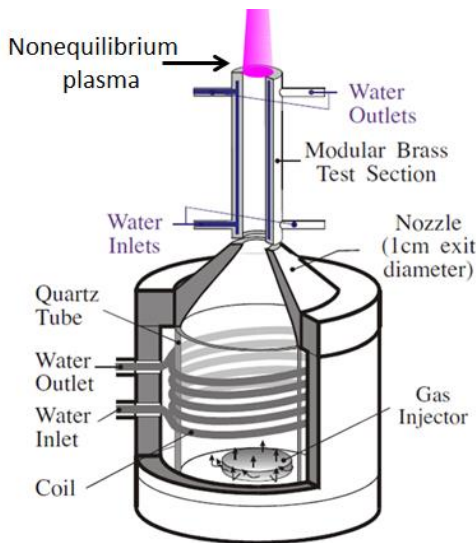
The prediction of the radiation from the shock layer that forms in front of a space capsule during atmospheric entry is required for the design of thermal protection systems [1] [2]. Depending on the entry conditions (atmosphere, velocity...) the radiation may be dominated by molecular bands or atomic lines. For a Mars entry scenario, the radiation from atomic lines is expected to be insignificant (< 3% of the total radiation) [3]. However, for an Earth entry, atomic lines can be the most significant source of radiation [4] [5]. Efforts have been made to couple a CFD code with radiative solvers to correctly predict the total heat flux on the capsule mainly in the forebody region [6].

In the afterbody region, the plasma can depart from equilibrium owing to the hydrodynamic expansion incurred while flowing from the forebody to the afterbody. This expansion forces quick plasma recombination and departure from chemical equilibrium. The resulting nonequilibrium radiation is not well modeled at the moment [2] [7]. In particular, the uncertainty on the radiation heat flux can be up to 200% for a Mars reentry scenario. Recent results showed that the atomic lines, just as for the forebody, are also the main source of radiation in the afterbody for Earth entry [7], specifically in the VUV region. In order to reduce these uncertainties, experimental data for recombining flows are needed to test the predictions of kinetic and CFD models regarding atomic species production and subsequent radiation.

In a previous publication [8], we presented measurements made in a recombining nitrogen plasma. We measured the temperature profile of the plasma and the resulting molecular radiation. The plasma was found to be in nonequilibrium with an overpopulation of atomic nitrogen and excited states of molecular nitrogen. These overpopulations led to nonequilibrium radiation that was found to be up to  $10^6$  times higher than the equilibrium radiation in the visible region. Here we extend the measurements to the atomic nitrogen radiation to get the population of excited states of atomic nitrogen using optical emission spectroscopy from the vacuum ultraviolet (VUV) to near infrared regions. In the following sections, the temperature profiles of the plasma will be taken from the previous publication [8].

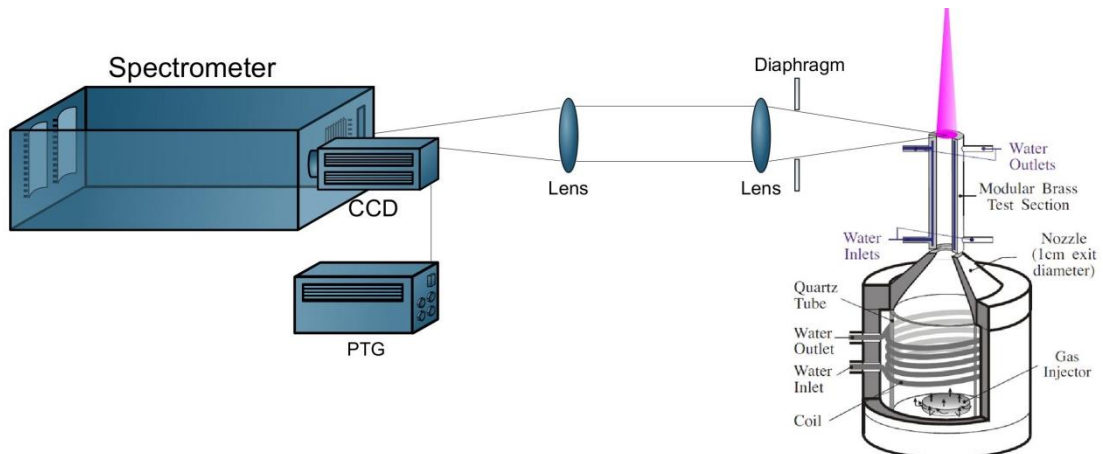
## II. Experimental setup

The plasma torch facility is a TAFE Model 66 inductively coupled plasma (ICP) torch powered by a 120 kVA radio frequency LEPEL Model T-50-3 power supply. The power supply operates at 4 MHz and can supply a maximum of 12 kV DC and 7.5 A to the oscillator plates. Details of the plasma torch facility are provided in previous publications [9] [10] [11]. The plasma produced within the torch exits through a 1-cm diameter nozzle at a speed around 700 m/s in the center. The plasma studied here is comprised of 90 slpm of  $N_2$  premixed with 50 slpm of argon to provide stable operating conditions for the torch. A water-cooled tube made of brass is mounted on top of the plasma torch. Two tubes of length 10 and 15 cm are used in the present work. Emission spectroscopy measurements are performed at the entrance and exit of the modular tube. Instead of creating a strong expansion as with a reentering capsule, the plasma is instead forced to recombine by imposing rapid cooling via the cold wall (maintained at approximately 300 K). This setup is therefore intended as a simplified experimental model of a real atmospheric entry scenario. Details of the experimental setup are presented in figure 1.



**Figure 1: Water-cooled test-section mounted on the top of the plasma torch**

In the visible/ near infrared (NIR), Optical Emission Spectroscopy (OES) was performed with the setup presented in Figure 2. The spectrometer is an Acton Instrument 2500i with 1200 gr/mm gratings. We measured spectra from 400 to 900 nm. All spectra were calibrated from pixel counts to absolute intensity using an Optronics Laboratories tungsten lamp (model OL550) and were Abel-inverted to get spatially resolved local plasma emission. We used several atomic nitrogen lines to get a measurement of the absolute density of the emitting levels. Note that under our conditions the plasma is optically thin in the observed wavelength range. The spectroscopic constants for the observed atomic lines are summarized in Table 1. We performed measurements at three different axial positions: 0, 10 and 15cm.

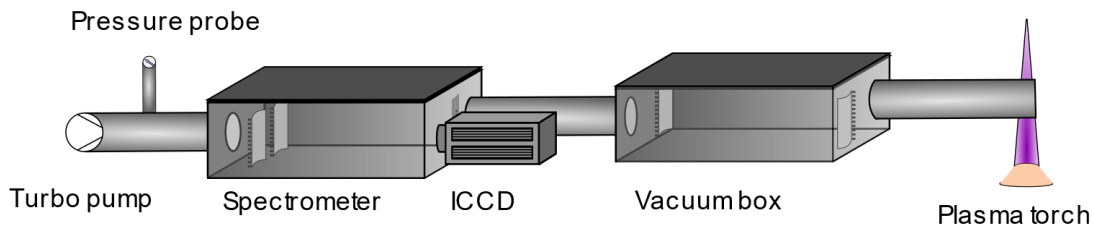


**Figure 2: Experimental setup for emission spectroscopy**

**Table 1: Spectroscopic constants for the observed atomic lines of nitrogen. Data taken from the NIST database.**

Wavelength (nm)	$\epsilon_u(cm^{-1})$	$g_u$	$\epsilon_l(cm^{-1})$	$A_{ul}(10^7 s^{-1})$
746.83	96750.84	4	83364.62	1.96
821.07	95493.69	4	83317.83	0.523
821.63	95532.15	6	83364.62	2.26
822.31	95475.31	2	83317.83	2.62
859.4	97770.18	2	86137.35	2.09
862.92	97805.84	4	86220.51	2.67
871.17	94793.49	4	83317.83	1.26

In addition to the measurements in the visible, we measured atomic lines of nitrogen around 174 nm using a vacuum ultraviolet (VUV) McPherson (Model 218) spectrometer. The setup is detailed in Fig. 3. The grating has 2400 grooves/mm and is blazed at 150 nm. The system is vacuumed using a roughing pump and a turbopump (80L /s pumping speed). The pressure is about  $5 \times 10^{-5}$  Torr in the system for all tests. The spectra are calibrated using an argon mini-arc of radiance traceable to NIST standards [12]. Note that in the VUV the plasma is no longer optically thin. Therefore, we did not Abel-invert the measured spectra. Instead we used the radiation code SPECAIR [13] [14] to analyze our measurements. This analysis will be detailed in the following section.



**Figure 3: Experimental setup for VUV emission spectroscopy**

### III. Determination of the densities of atomic nitrogen excited states

In the visible/NIR, we used the measured nitrogen atomic lines to determine the density of the emitting levels. In the considered range of wavelength, the plasma is optically thin and the measured integrated intensity  $I$  of the transition is related to the density of the upper level  $n_u$  [11] through:

$$I = n_u \frac{A_{ul}}{4\pi} (\epsilon_u - \epsilon_l)$$

where  $A_{ul}$  is the Einstein coefficient,  $\epsilon_u$  and  $\epsilon_l$  are the energies of the upper and lower states of the considered transition, respectively. The measured transitions are listed in Table 1 with the relevant

spectroscopic constants. Note that the measured spectra are Abel-inverted to get volumetric emission of the plasma.

In the VUV, the plasma is no longer optically thin, and this simple relation does not hold anymore. Instead, the measured intensity depends on the densities of both the lower (absorbing) and upper (emitting) states of the transition. Furthermore, the full radiative transport equation must be solved across the plasma profile. Without assumption, a single measurement of a transition cannot be used to get the population of both states in our conditions. The line broadening is dominated by the slit function of the spectrometer in our measurements. In order to determine the density of either the lower or upper state, the other one must be fixed via some appropriate assumption or a secondary measurement. We assume that the lower level of the transition, namely  $N(^2P)$ , is in equilibrium with the ground state at the measured temperature. This was indeed predicted for this configuration in Ref [7]. The density of the lower level  $n_l$  of the transition is given by:

$$\frac{n_l}{g_l} = \frac{n_0}{g_0} \exp\left(-\frac{\epsilon_l}{k_B T}\right)$$

where  $n_0$  is the density of the ground state of atomic nitrogen and where  $g$  and  $g_0$  represent the degeneracies,  $\epsilon_l$  the energy of the considered level,  $k_B$  the Boltzmann constant and  $T$  the plasma temperature.

Note, furthermore, that the ground state of nitrogen was indirectly measured in a previous publication [8] and was found to be highly overpopulated compared to equilibrium conditions. We therefore impose that the density of the lower state of the transition is in equilibrium with the ground state density at the measured temperature. Given this and the measured line intensity, we then used SPECAIR to fit our spectra and determine the upper state density. For each axial position, we measured spectra at three different radial positions:  $r = 0, 1$  and  $2$  mm. We assume that the radial profile of the upper state density of the transition follows a gaussian distribution – however, we impose no limitation on the mean and the standard deviation of this gaussian profile. This assumption is viewed as reasonable because all upper state densities measured in the visible/NIR spectral regions followed a gaussian distribution. Theoretical spectra were obtained with SPECAIR along the line of sight at the different radial positions. Then we estimated the cumulative errors between the computed spectra and the measured ones for each gaussian distribution. The minimum residual gives the best estimate of the population of the upper level. Comparison between the best fit and the measurements will be presented in the following section.

#### IV. Results

The plasma studied here is comprised of 90 slpm of nitrogen and 50 slpm of argon. In a previous publication [8], we showed that this plasma was in equilibrium at 0 cm and in nonequilibrium at 10 and 15 cm. These same axial locations will be studied in this section, namely 0, 10 and 15 cm. The 174 nm doublet of nitrogen was measured. The spectroscopic constants for this doublet are given in Table 2.

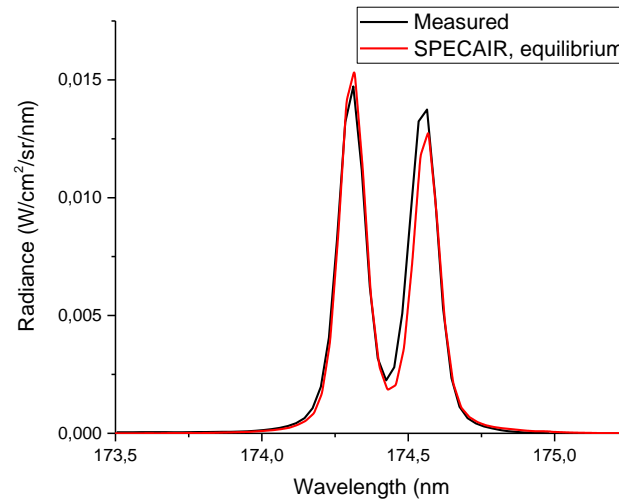
**Table 2: Spectroscopic constants for the 174 nm doublet. Data taken from the NIST database.**

Wavelength (nm)	$\epsilon_u (cm^{-1})$	$g_u$	$\epsilon_l (cm^{-1})$	$g_l$	$A_{ul} (10^7 s^{-1})$
174.27	86220.510	4	28839.306	4	10.5
174.525	86137.35	2	28838.92	2	8.35

## 0 cm

At 0 cm, we previously showed that the visible/NIR emission spectrum was correctly predicted with an equilibrium simulation using SPECAIR [8]. The densities of the excited states of atomic nitrogen can then be found assuming a Boltzmann distribution at the measured temperature.

In the VUV, we measure a spectrum at the center of the tube and compare it with the computed one using SPECAIR along the line of sight assuming the temperature profile previously measured. The results are presented in Fig 4. The good agreement once again supports that the plasma is in equilibrium at 0 cm.

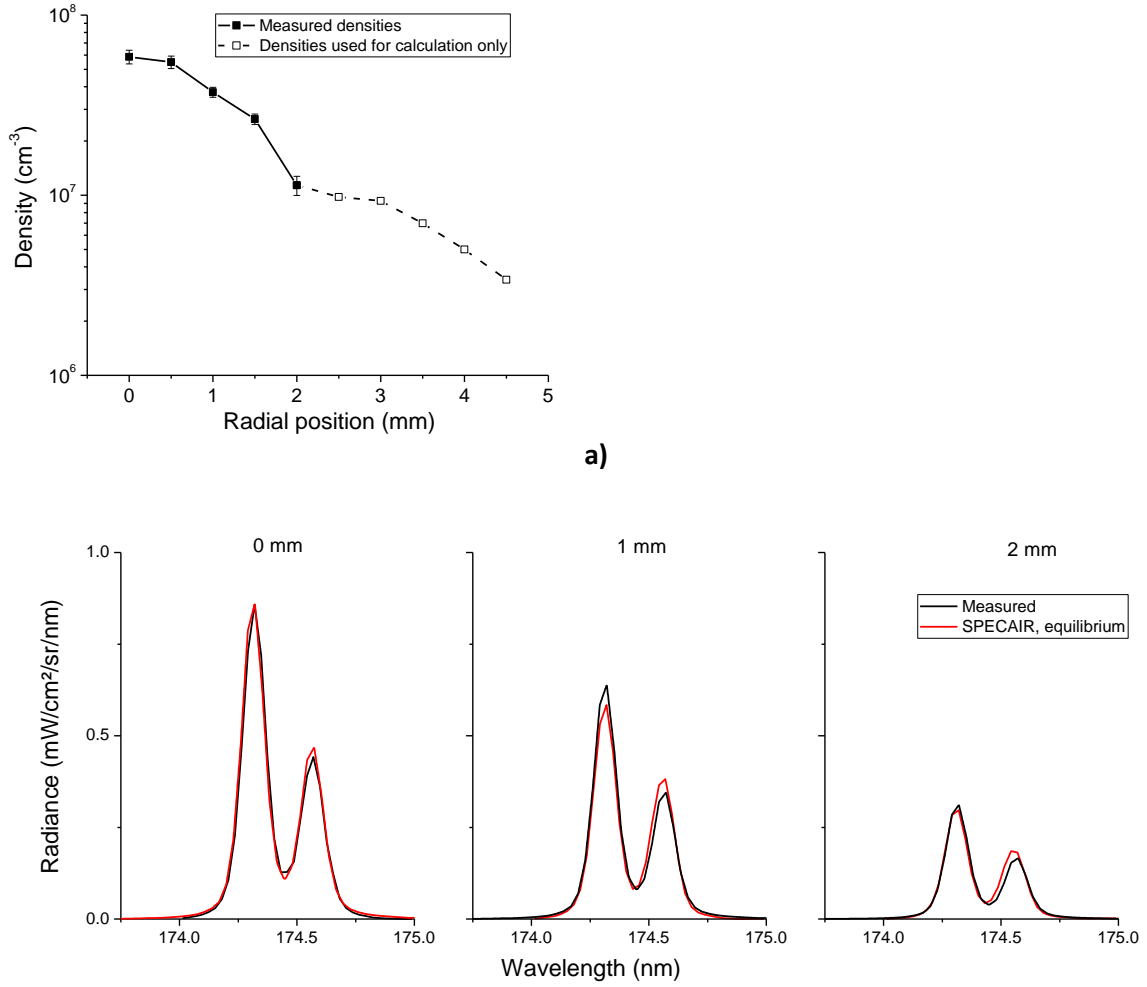


**Figure 4: Measured (black) and computed using SPECAIR (red) spectra at the inlet of the tube ( $x = 0$  cm) at the center of the tube ( $r = 0$  mm).**

## 10 cm

At 10 cm, the plasma is no longer under equilibrium conditions, as shown in Ref. [8]. In the visible/NIR, we integrate the measured intensity of the transitions given in Table 1 to get a measurement of their corresponding upper state densities.

In the VUV, we perform the procedure detailed in the previous section to get the best estimate of the density of the upper state of the transition. Note that the lower state was fixed to be in equilibrium with the ground state of atomic nitrogen at the measured temperature. The radial profile of the upper state and the corresponding measured and computed radial spectra at 0, 1 and 2 mm are presented in Figure 5. The good agreement between computed and measured spectra gives us confidence in the density measurement of the upper level for  $r < 2$ mm. However, for  $r > 2$ mm, due to the lack of measurement, the values suffer from large uncertainties and are reported here for informative purposes of the computed spectra.



b)

**Figure 5: a) radial density profile of the upper state of the measured transition at 10 cm. b) comparison between the computed spectrum using SPECAIR assuming the density profile given in a) and the measured one at three different radial positions (0, 1 and 2 mm).**

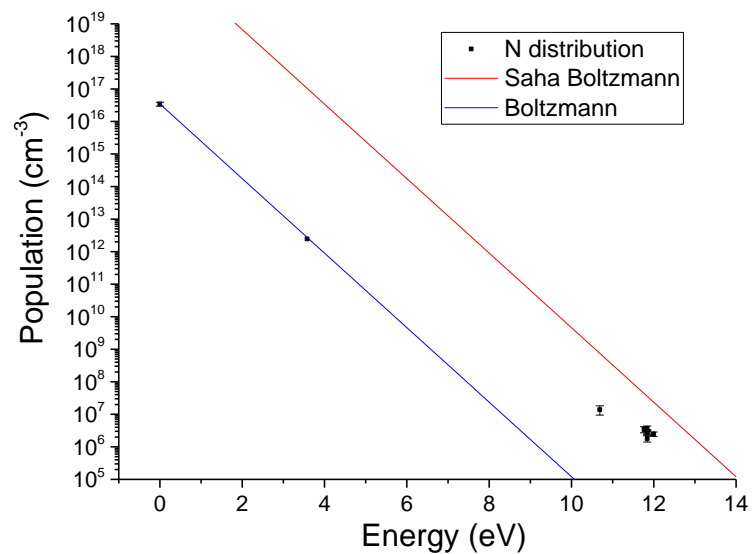
In recombining plasmas, the densities of the excited states  $n_u$  close to the ionization limit often are in Saha-Boltzmann equilibrium with the electron/ion densities, and are given by:

$$\frac{n_u}{g_u} = \frac{n_+ n_e}{2Q_+} \left( \frac{h^2}{2\pi k_B T} \right)^{\frac{3}{2}} \exp\left( \frac{\epsilon_{ion} - \epsilon_u}{k_B T} \right)$$

where  $n_+$  is the density of atomic ions,  $n_e$  the density of electrons,  $Q_+$  the partition function of the atomic ion,  $h$  the Planck constant and  $\epsilon_{ion}$  the ionization energy. Because the electrons/ions do not have time to recombine, the overpopulation of these species results in an overpopulation of the excited state density with respect to the Boltzmann distribution.

In Fig 6, we plot both these distributions along with the measured densities of excited states of nitrogen measured in the VUV, visible and NIR. Note that for the Saha-Boltzmann distribution, the density of electrons was previously measured in Ref. [15] and was found to be equal to  $6.4 \cdot 10^{-13} \text{cm}^{-3}$ . The density of atomic ions was estimated assuming that the overpopulation of  $\text{N}^+$  was equal to the overpopulation of the electrons. The higher energy states of atomic nitrogen are highly overpopulated

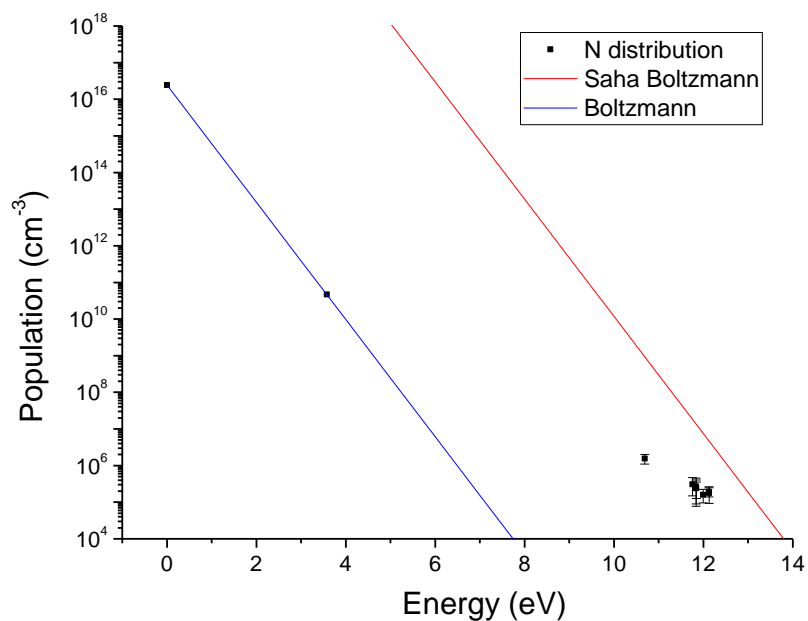
as compared to their equilibrium values and tend to be above the Boltzmann distribution and closer to Saha-Boltzmann. This suggests that the high energy states of nitrogen tend to be at equilibrium with the electrons.



**Figure 6: Measured population of ground and excited states of atomic nitrogen (black) at 10 cm. The Boltzmann (blue) and Saha Boltzmann (red) distributions are also plotted.**

### 15 cm

At 15 cm we perform the same measurements as at 10 cm. The results are presented in Figure 7. We observe the same trend as at 10 cm. The higher energy levels of atomic nitrogen are highly overpopulated relative to the Boltzmann distribution at the measured temperature and tend to be closer to the Saha-Boltzmann distribution.



**Figure 7: Measured population of ground and excited states of atomic nitrogen (black) at 15 cm. The Boltzmann (blue) and Saha Boltzmann (red) distributions are also plotted.**



At both 10 and 15 cm, we observe a departure from the Boltzmann distribution for the densities of the high excited states of atomic nitrogen. The high energy states of nitrogen tend to be in equilibrium with the electrons as they are close to the Saha-Boltzmann distribution. This was previously experimentally observed and predicted in previous works. In compression flows, it was observed in the Fire II flight experiment [16]. Panesi *et al* [17] and Johnston [18] predicted high excited states of atomic nitrogen underpopulated compared to the Boltzmann distribution. In their conditions as it is usually the case in such hypersonic compression flows, the chemistry is partly frozen, and the Saha-Boltzmann distribution curve is lower than the Boltzmann one due to an underpopulation of electrons compared to equilibrium conditions. The density of excited atomic nitrogen tends to the Saha-Boltzmann distribution for high energy levels.

In expanding flows such as in the afterbody region of a reentry capsule, Johnston and Brandis [7] predicted a similar trend. In this case, the Saha-Boltzmann distribution yields higher densities than the Boltzmann distribution as the chemistry is partly frozen. The excited states of atomic nitrogen are overpopulated compared to the Boltzmann distribution and their densities tend to the Saha-Boltzmann one for high energy levels. This model was compared to Fire II [16] and Stardust Echelle experiments [19] and was able to improve the radiation prediction.

The CR models are also used to predict the radiation in shock tubes such as the EAST facility. Previous work [20] showed the importance of correctly predicting the density of atomic nitrogen to avoid uncertainties on the resulting radiation.

Our experiments give another set of data that can be used to validate CR models for atomic nitrogen. Also, the simple configuration of our experiment may be useful to validate the kinetics of nitrogen by coupling CR and CFD models.

## V. Conclusion

Optical emission spectroscopy from VUV to NIR was performed in a recombining nitrogen flow to measure the density of excited states of atomic nitrogen. The plasma is initially under equilibrium conditions at 0 cm and the excited states follow a Boltzmann distribution at the measured temperature.

At 10 and 15 cm, a departure from equilibrium is observed. The lowest energy levels of atomic nitrogen were assumed to be in equilibrium with the measured ground state as this is usually the case for these high enthalpy flows [7] [17]. The highest levels were measured in the VUV and the NIR region and were found to be highly overpopulated relative to the Boltzmann distribution at the measured temperature. Their densities tend to the Saha-Boltzmann distribution. This confirms previous predictions [7] of the Fire II flight experiment and the Stardust reentry for afterbody flows.

These results in a simple configuration experiment intend to serve as validation database for kinetic models coupled with CFD codes. In future work, additional direct measurements of the lower states of the observed transitions will be measured to remove the assumption made of a Boltzmann distribution of these states with the ground state of nitrogen.

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