Experimental characterization of ultraviolet radiation of air in a high enthalpy plasma torch facility

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Abstract

During atmospheric re-entry, a plasma is formed ahead of the surface of the vehicle, and the excited particle present in the plasma produce radiative heating fluxes to the surface of the vehicle. A high-temperature air plasma torch operating at atmospheric pressure was used to experimentally reproduce atmospheric re-entry conditions. A high-resolution and absolute intensity emission spectrum (FWHM: 0.064 nm) was obtained from 200 to 450 nm and then compared with computational results provided by the SPECAIR code [1]. This paper discusses the comparison of the two spectra over this wavelength range in order to confirm the validity of the calculation and provide direction to improve the calculated spectrum.

1. Introduction

The ultraviolet range is responsible for a large fraction of the radiation of high temperature air plasmas, through the NO beta, gamma, delta, epsilon, beta prime, and gamma prime band systems, the N_2 second positive system, the N_2^+ first negative system, the CN Violet bands, the O_2 Schumann-Range system, and the radiation of atomic carbon. For reentry vehicles at velocities from 3 to 8 km/s, these systems represent the dominant fraction of the radiation emitted by the plasma layer located between the bow shock and the surface of the vehicle. Various spectroscopic models have been implemented in radiation codes such as SPECAIR [1] to predict the spectral intensity of these systems and generally good agreement has been obtained with measurements at medium spectral resolution (see Figure 1). However, a close inspection of the spectra shown in Figure 1 shows a few discrepancies between the measurements and the experiments.

The purpose of this work is to compare the predictions of the SPECAIR spectral models against measurements of the various band systems taken at a higher spectral resolution (about 5 times higher) than previously reported in Ref. [1]. To this end, we generated a thermal air plasma with a 50 kW inductively coupled plasma torch operating at atmospheric pressure. This facility provides high enthalpy air plasma at temperatures up to 7500 K, a range well suited for the study of all the systems emitting in the ultraviolet between 200 and 400 nm. Because the plasma is in Local Thermodynamic Equilibrium (LTE), the species concentrations and the population distribution over the internal energy states of atoms and molecules can be modeled with the knowledge of only temperature and pressure. In this work, high resolution measurements were made with an Acton SpectraPro 2750i spectrometer and an intensified CCD camera (PIMAX, Roper Scientific). The SPECAIR model was then compared to the measured data. Section 2 of this paper presents the experimental setup used to obtain the experimental spectra and Section 3 provides the comparison of the experimental and calculated spectra.

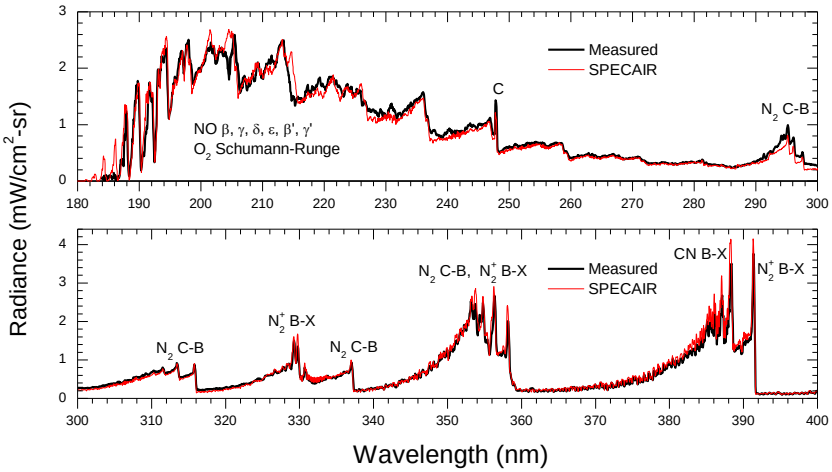

Figure 1: Comparison of measured and predicted spectra of NO emitted by a LTE air plasma at 7500 K [2]

2. Experimental Setup

2.1 High Enthalpy Plasma Torch

The facility used is a radio-frequency inductively coupled plasma torch, TAFA Model 66, powered by a radio frequency LEPEL Model T-50-3 power supply operating at 4MHz (120 kVA) [5]. During the torch starting phase, argon is used because its ionization potential helps initiate the high temperatures required to sustain the plasma. The argon is then slowly switched to air. The gas flow is injected in axisymmetric fashion, with both axial and swirl injection. Swirl injection provides gas recirculation and torch stabilization. A converging copper nozzle is placed at the exit of the torch. In this study, the nozzle diameter is 5 cm. All measurements presented in this paper were conducted 16 mm above the nozzle exit. At this location, the maximum velocity was approximately 20 m/s and the measured maximum temperature was 7500 K, corresponding to a density, ρ, of 3.16×10^{-2} kg/m^3 and a dynamic viscosity, η, 1.99×10^{-4} kg/m/s [4]. With these values, the Reynolds number at the measurement point was about 181. The plasma jet is therefore laminar in the measurement region. It remains so downstream of the nozzle over a few centimetres [5].

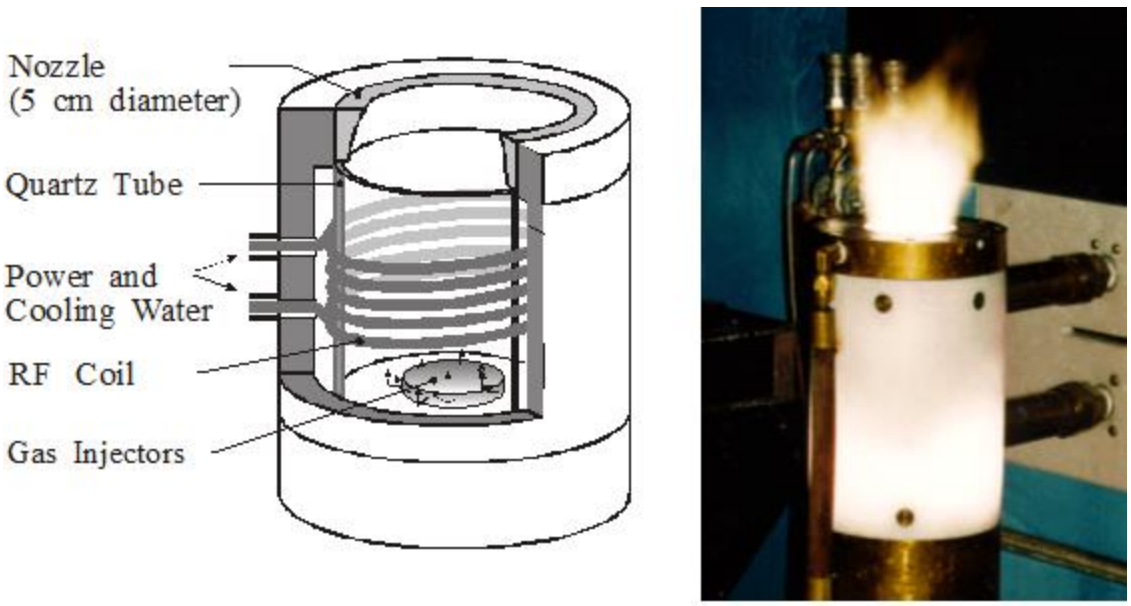

Figure 2: Torch head schematic and photograph of the air plasma plume. All measurements were performed 16 mm downstream of the torch exit nozzle [5]

2.2 Optics Schematic

A photograph of the optical train and spectrometer is shown in Figure 3, and Figure 4 presents the optical schematic. Vertical and lateral scanning capabilities are possible with a set of two off-axis parabolic and five flat mirrors on horizontal and vertical translation stages [5].

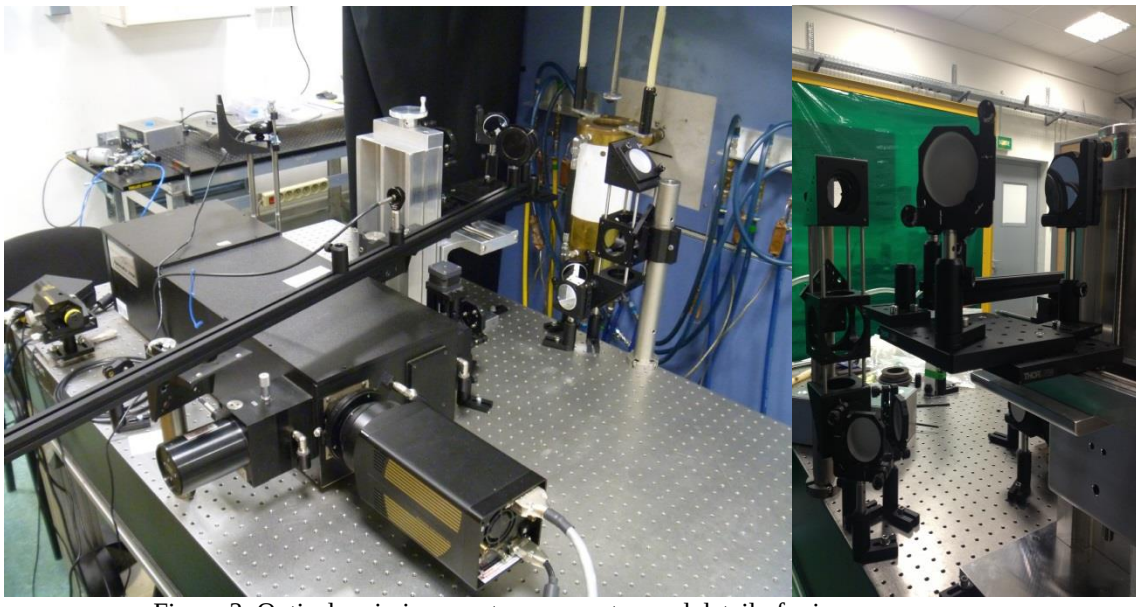

Figure 3: Optical emission spectroscopy setup and detail of mirrors

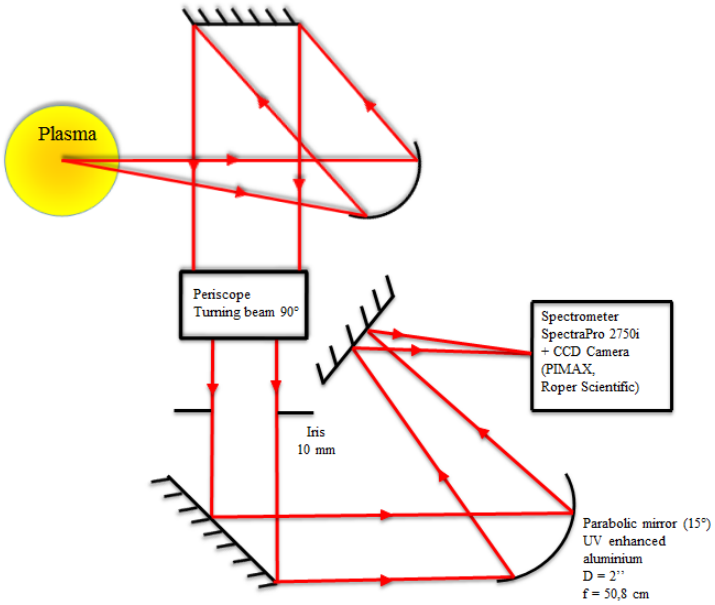

Figure 4: Schematic of the optical system with the SpectraPro spectrometer

2.3 Spectrometer

The measurements conducted in this work used an Acton Research SpectraPro SP2750i spectrograph of focal length 750 mm. The grating has 1200 g/mm and is blazed at 300 nm, providing a wavelength range of approximately 15 nm in one image.

The spectrometer is equipped with a VUV-enhanced intensified CCD camera (Princeton Instruments PI-MAX 1024RB-UV-18mm with 1024 x 256 pixels) allowing acquisition of emission from the VUV to the near-IR (140 - 930 nm). A HeNe laser is used to align the spectrometer with the torch, and the measurements were taken with a

spectrometer entrance slit width of 25 microns. With this setup, the spectral resolution of the spectrometer is approximately 0.064 nm (FWHM). The spectrometer is used to measure the emission spectrum of the air plasma between 200 and 450 nm and to determine the radial temperature profile of the plasma using the oxygen triplet at 777 nm.

Calibration Lamps

The measured spectrum was calibrated in absolute intensity using two calibration sources. An OL-550 tungsten ribbon lamp was used to calibrate the range 350 – 450 nm and an argon mini-arc [5] was used from 200 to 400 nm. The OL-550 lamp and the argon mini-arc are both radiance standards with calibration traceable to NIST standards. The experimental spectrum of the air plasma was calibrated in absolute intensity over the range 200 – 340 nm and 340 – 450 nm by dividing the measured spectrum by the argon mini-arc spectral response and by the OL-550 tungsten lamp response, respectively.

3. Results

3.1 Temperature Profile

The SPECAIR calculations required the equilibrium radial temperature and the width of a slab in order to compute a spectrum. The slab is the width in centimeter that separate two temperature measures. Then we added all the spectra computed with the temperature profile, a spectrum is associated with one temperature, in order to finally model the spectrum with the intensity measured at the center of the plasma.

The radial temperature of the plasma flow was determined by measuring the emission from the atomic oxygen triplet around 777 nm. The line intensity of a specific transition is given by Equation 1 [3].

$$I\left[\frac{W}{cm^3.sr}\right] = \frac{n_u.(\varepsilon_u - \varepsilon_l).A_{ul}}{4\pi} \quad (1)$$

The intensity measured along the plasma centerline is the superposition of the contributions along the plasma radius. In order to determine the radial temperature profile in the plasma, the Abel inversion technique was applied (Equation 2). The method used is presented in greater detail by Laux [3].

$$e(r) = -\frac{1}{\pi}.\int_r^R \frac{dI/dx}{\sqrt{x^2-r^2}}\,dx \quad (2)$$

A_{ul}, ε_u, and ε_l are tabulated [6], and for this transition are given in Table 1.

Table1: Characteristics of the atomic oxygen transition at 777 nm [6]

A_{ul} [sec^{-1}]	$3.69x10^7$ (Uncertainty ≤ 3%)
ε_u [cm^{-1}]	86629
ε_l [cm^{-1}]	73768
g_u [~]	15

This leaves n_u as the final unknown quantity in Equation 1. In an LTE plasma, this quantity is related to the temperature and pressure as given in Equation 3.

$$\frac{n_u}{g_u} = X_O(T_{LTE})\frac{P}{kT_{LTE}}\frac{1}{Q_{el}(T_{LTE})}\exp\left(-\frac{\varepsilon_u}{kT_{LTE}}\right) \quad (3)$$

Here, $X_O(T_{LTE})$ is the mole fraction of atomic oxygen in air at P and T_{LTE}, which is tabulated as a function of temperature, P is the pressure at which the measurement was carried out (here P = 1 atm), k is the Boltzmann constant, and $Q_{el}(T_{LTE})$ is the electronic partition function of atomic oxygen at T_{LTE}, which for the temperatures considered here is equal to 9. This leaves T_{LTE} as the only unknown, and through an iterative process, the temperature of the plasma can be determined, provided the absolute intensity, I, of the line is measured.
The temperature profile obtained is presented in Figure 5.
As explained by Laux [3], by taking the logarithm of Equation 3 and differentiating, we obtain:

$$\left|\frac{\Delta^{(n_u/g_u)}}{n_u/g_u}\right| = \left|\frac{\Delta X_O}{X_O}\right| + \left|\frac{\Delta T_{LTE}}{T_{LTE}}\right| + \left|\frac{\Delta Q_{el}}{Q_{el}}\right| + \frac{\varepsilon_u}{kT_{LTE}}\left|\frac{\Delta T_{LTE}}{T_{LTE}}\right| \quad (4)$$

Then, the uncertainty at an average temperature of 7000 K, neglecting $\left|\frac{\Delta Q_{el}}{Q_{el}}\right|$, $\left|\frac{\Delta(\Delta E_{ul})}{\Delta E_{ul}}\right|$ and $\left|\frac{\Delta X_O}{X_O}\right|$, which are less than 1% according to [3], is given by:

$$\left|\frac{\Delta T_{LTE}}{T_{LTE}}\right| = (1 + \frac{\varepsilon_u}{kT_{LTE}})^{-1}\left(\left|\frac{\Delta I}{I}\right| + \left|\frac{\Delta A_{ul}}{A_{ul}}\right|\right) \quad (5)$$

By considering the uncertainty of the spontaneous Einstein A coefficient to be about 3% [6], we obtain the approximate relation at 7000K:

$$\left|\frac{\Delta T_{LTE}}{T_{LTE}}\right| \cong 0.05\left(\left|\frac{\Delta I}{I}\right| + 0.03\right) \quad (6)$$

As the measured intensities of the oxygen line at 777.3 nm were obtained with an accuracy better than 20%, the uncertainty on the measured temperature is about 3%.

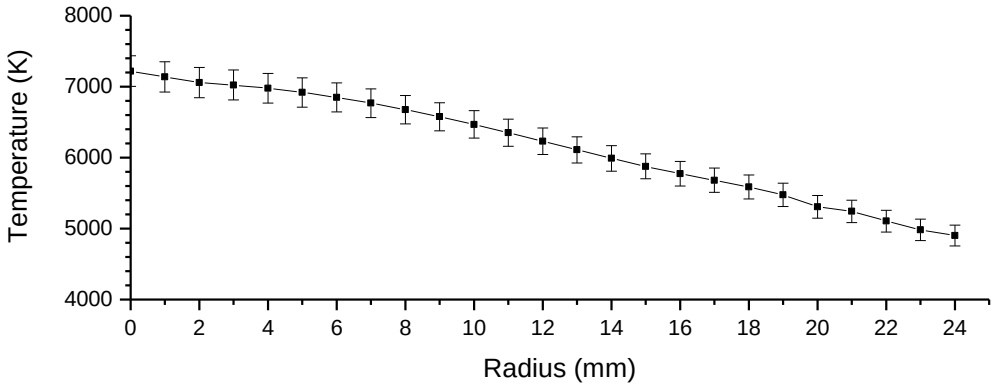

Figure 5: Radial temperature profile at 16 mm downstream of the plasma torch exit, with a nozzle of 5 cm diameter, P=1 atm.

We can examine the sensitivity of several features that appear in the measured spectra to the temperature. The C line (at 247 nm), N_2 (at 337 nm), and N_2^+ (at 391 nm) transitions are very sensitive to the uncertainty in temperature: an error of ±3% on the temperature modifies the maximum intensity of the spectrum of these species by 60%, 27%, and 52%, respectively. The rest of the spectrum is affected by about 10% (for example, the NO transitions). Figure 6 shows the effect of a ±3% variation of the temperature on the C, N_2, N_2^+ and NO transitions. Because the intensity of the N_2^+ first negative spectrum at 392 was better predicted by the upper bound temperature profile, all SPECAIR simulations presented in this article will be based on this upper bound temperature profile. This is further justified by the fact that self-absorption effects on the intensity of the O line are likely to increase the measured temperature, by about 100 – 200 K, according to Ref [3].

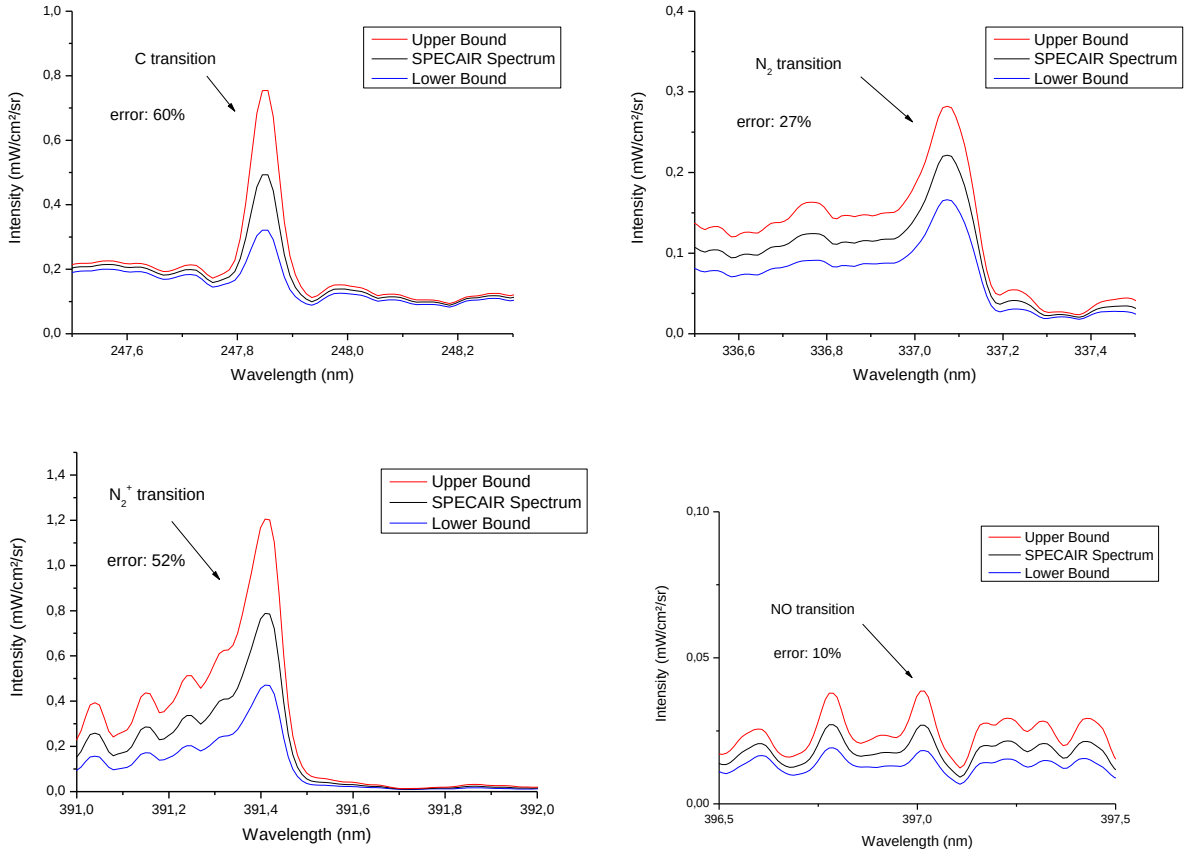

Figure 6: Variations of the C, N_2, N_2^+ and NO transition intensities for a ±3% variation of upper and lower bound of the temperature

3.2 SPECAIR calculations

SPECAIR is a line-by-line radiation code for computing, manipulating and fitting spectra based on several detailed models of the transitions of air species. These models are described in several references: UV and VUV systems of NO and N_2 [7], infrared systems [8], N_2 and N_2^+ spectra [9,10], OH spectra [11], CN and C_2 spectra [12].
The plasma torch spectrum was modeled for slabs of width 1 mm, each assumed to have homogeneous temperatures and species concentrations corresponding to those at the measured radial temperatures shown in Figure 5. The SPECAIR calculations were made between 200 and 450 nm, with a pressure of 1 atm, by generating the mole fractions and considering self-absorption. The species and transitions considered for this spectral range are: C, N, O, CN violet, N_2 (2+), N_2^+ (1-), NO Beta, NO Beta Prime, NO Delta, NO Epsilon, NO Gamma, NO Gamma Prime, and O_2 (Schumann-Runge).

The slit function was measured using a HeNe laser at 632.8 nm. Figure 7 shows the slit function measured with the SpectraPro spectrometer. The equivalent slit width at half maximum is about 0.064 nm, which is about 5 times narrower than the 0.29 nm FWHM of the spectra of Ref. [1] presented in Figure 1. The slit function was convolved by SPECAIR with the predicted emission spectra so that the resulting spectra could be directly compared to the measured spectra.

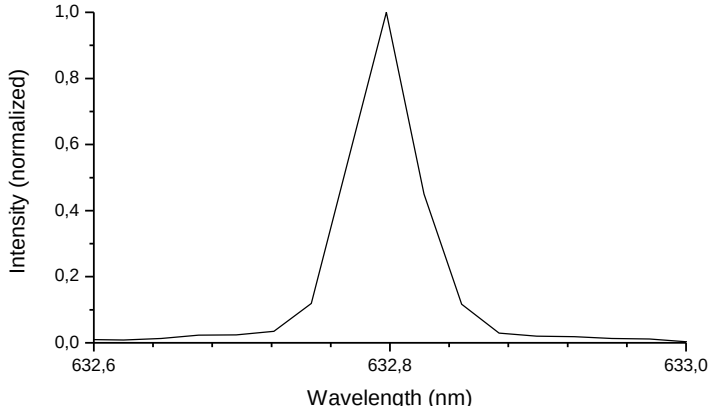

Figure 7: Spectra Pro spectrometer slit function

3.3 Experimental Spectra

Spectra were obtained over two wavelength ranges: 340 - 450 nm (calibrated with the OL-550 lamp) and 200 – 400 nm (calibrated with the argon mini-arc lamp). It can be seen in Figures 8 and 9 that the two spectra in absolute intensity overlap with good agreement between 340 and 400 nm, which is the range common to both radiance standards. For all results presented in this paper, we use the experimental spectrum calibrated with the OL 550 lamp for the comparison between 340 - 450 nm (Figures 11, 12, 13, 14, 15) and the experimental spectrum calibrated with the argon mini-arc for the comparison between 200 – 340 nm (Figures 16, 17, 18 ,19 20, 21, 22). The calibration with the OL-550 is considered to be more accurate in the overlapping region than the calibration with argon mini-arc.

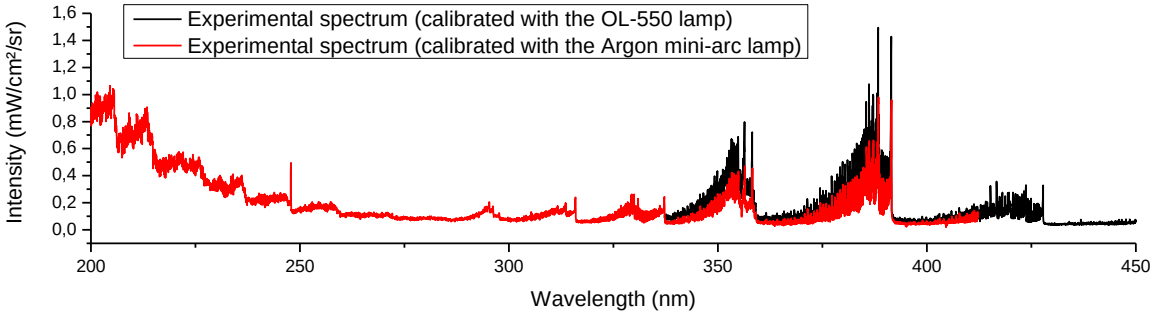

Figure 8: Absolute measured spectra calibrated with OL-550 and argon mini-arc between 200 and 450 nm

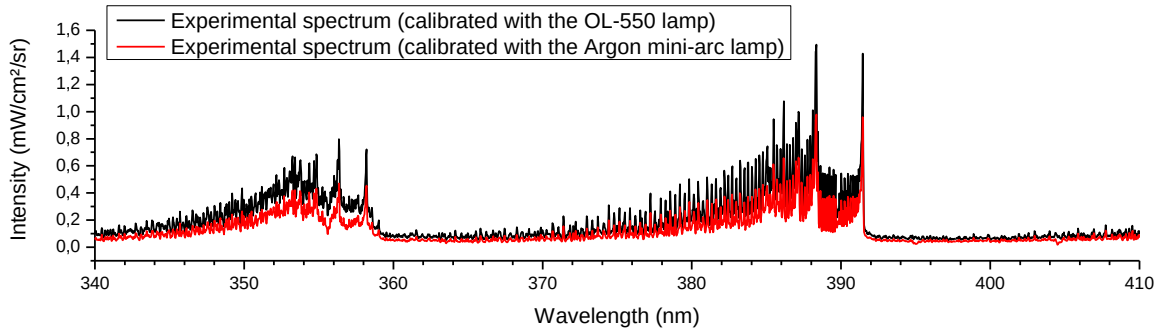

Figure 9: Absolute measured spectra calibrated with OL-550 and argon mini-arc between 350 nm and 400nm

3.4 Comparison of experiment and SPECAIR calculations

The comparison between the experiment and SPECAIR from 200 to 400 nm is presented in Figure 10. The overall agreement is seen to be very good, both in terms of the shape and the absolute intensity of the various features. There are however some differences, which we will examine over smaller spectral range regions.

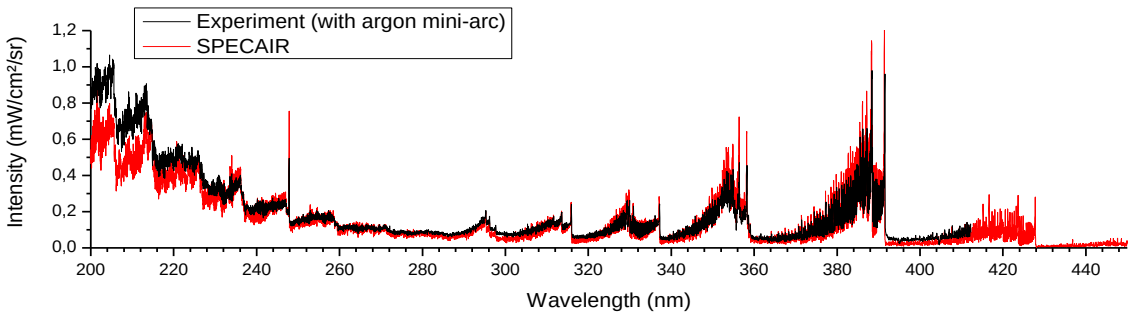

Figure 10: Comparison between the experimental spectrum and the SPECAIR calculated spectrum in absolute intensity between 200 and 400nm

3.5 Species Identification

This spectrum shown in Figure 11 contains different electronic band systems, namely N_2^+ (1-), N_2(2+), O_2 Schumann-Runge, NO (beta, gamma, delta, epsilon, beta prime, gamma prime), and CN violet, as well as one atomic line of carbon. At the considered temperatures (7400K), NO Beta, NO Delta, NO Epsilon, and NO Gamma, emit from 200 to 280 nm, and N_2^+, CN, and N_2 are responsible for most of the radiation between 280 and 400 nm. The atomic line of carbon is at 248 nm. We now consider smaller ranges for a more detailed comparison.

Between 392 and 425 nm SPECAIR closely reproduces the measured spectrum.

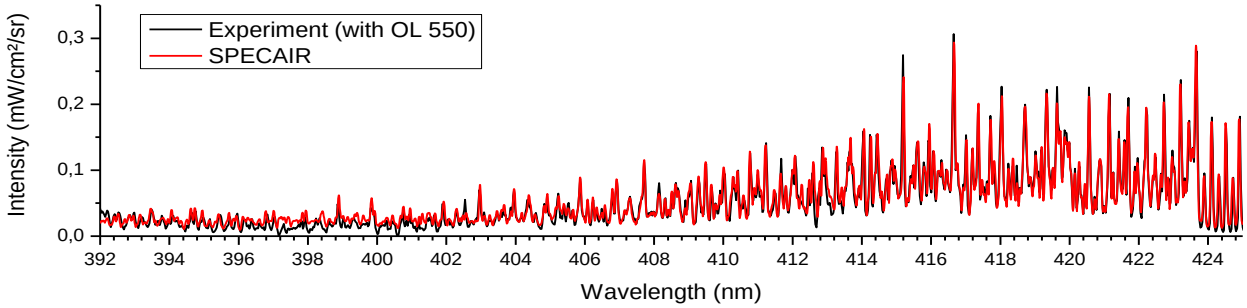

Figure 11: Measured and SPECAIR spectra between 392 and 425 nm

Between 384 and 392 nm, as can be seen from Figure 12, SPECAIR closely reproduces the measured spectrum, in particular the band systems of CN Violet and N_2^+ (1-). It is also remarkable to note the attenuation of the intensity at approximately 389.8 nm, which is due to perturbations of the N_2^+ (1-) transition, modeled in SPECAIR as described in Ref. [9] .

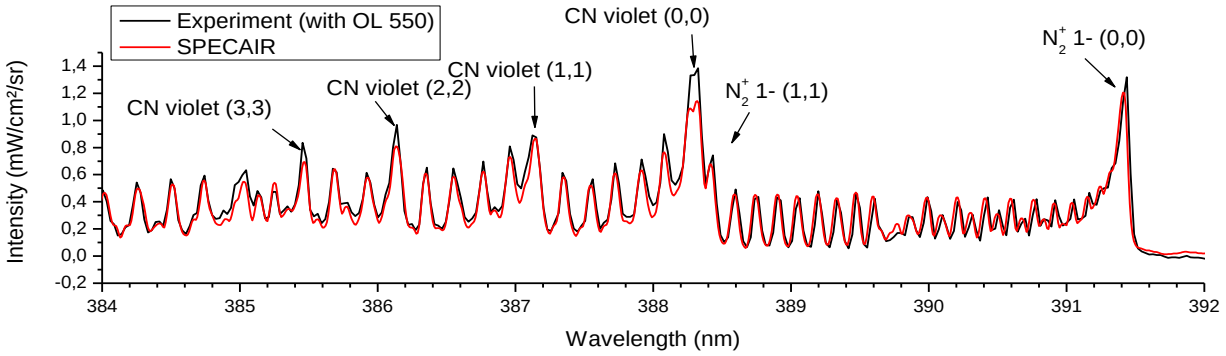

Figure 12: Measured and SPECAIR spectra between 384 and 392 nm

The measured and simulated spectra in the range 370 – 384 nm and 359 – 370 nm are shown in Figures 13 and 14. This range corresponds to the emission of N_2^+ (1-), CN Violet, and N_2 (2+). The agreement is again very good.

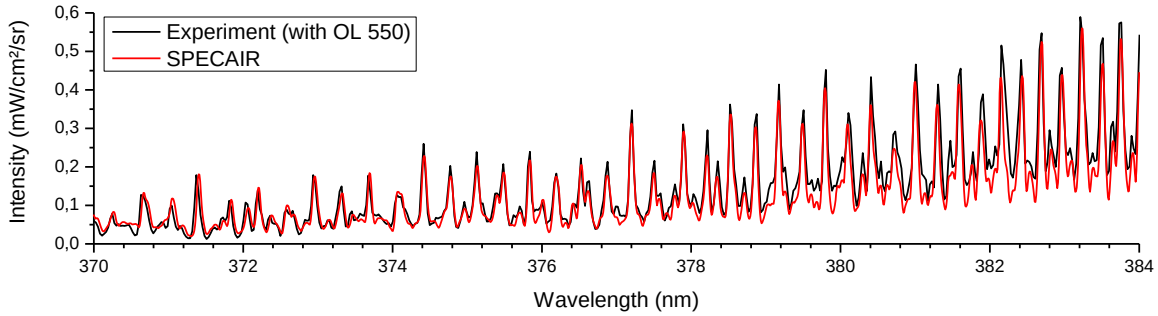

Figure 13: Measured and SPECAIR spectra between 370 and 384 nm

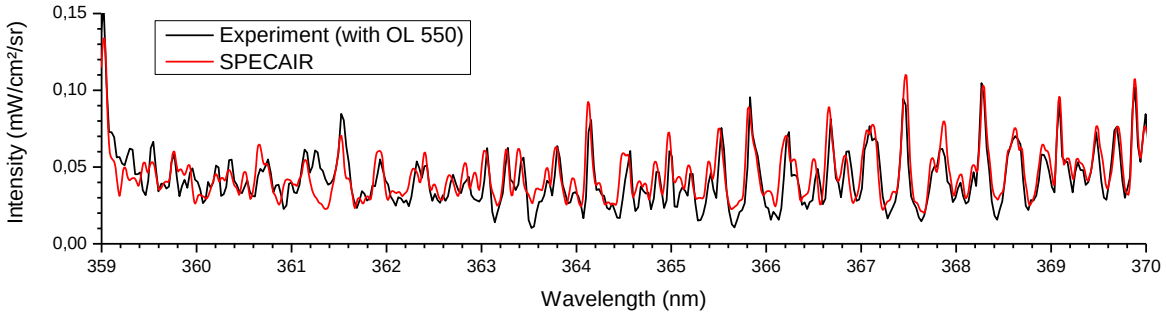

Figure 14: Measured and SPECAIR spectra between 359 and 370 nm

Between 346 and 360 nm (Figure 15), the Δv=1 bands of N_2^+ (1-) , CN Violet, and N_2 (2+) are present in the measured spectrum. Every transition of the measured spectrum is explained by the SPECAIR calculation.

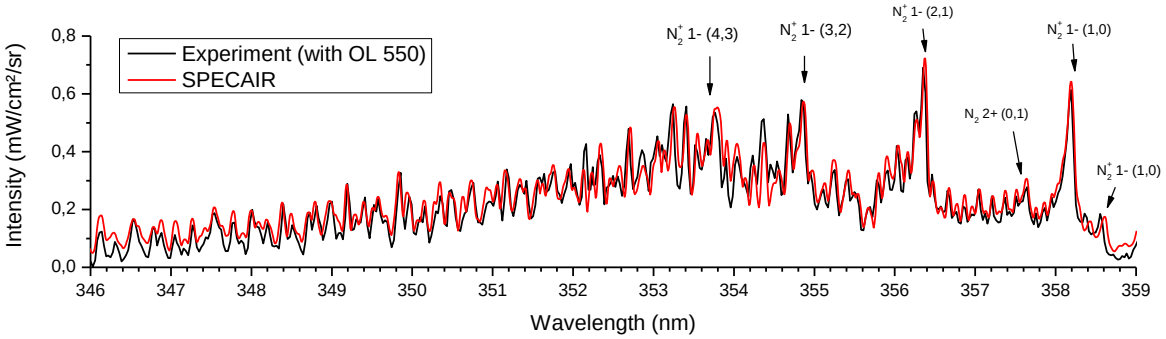

Figure 15: Measured and SPECAIR spectra between 346 and 360 nm

In Figure 16, the head of the (0,0) band of N_2 (2+) at 337 nm and a background of NO Beta, NO Gamma, and N_2^+ (1-) can be recognized from the measured spectrum between 332 and 338 nm. Every transition in the measured spectrum is well predicted by SPECAIR.

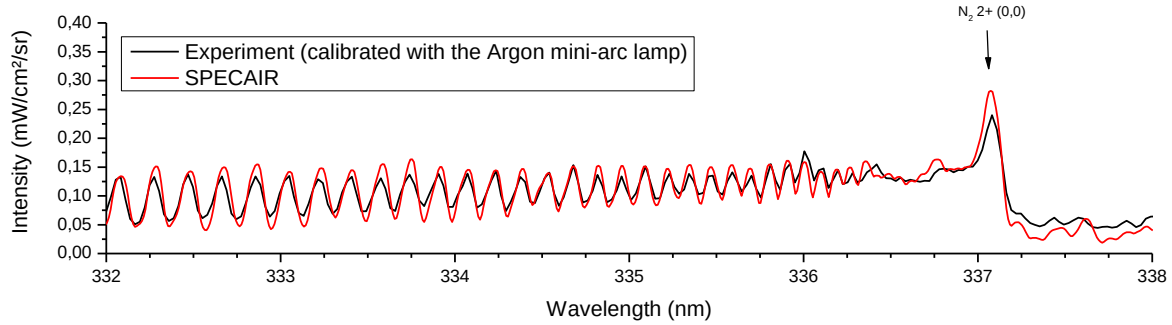

Figure 16: Measured and SPECAIR spectra between 332 and 338 nm

Between 318 and 332 nm (Figure 17) the measured spectrum includes N_2 (2+) and N_2^+ (1-) bands as well as a background of NO Beta and Gamma. The experimental spectrum is again well reproduced by the simulations.

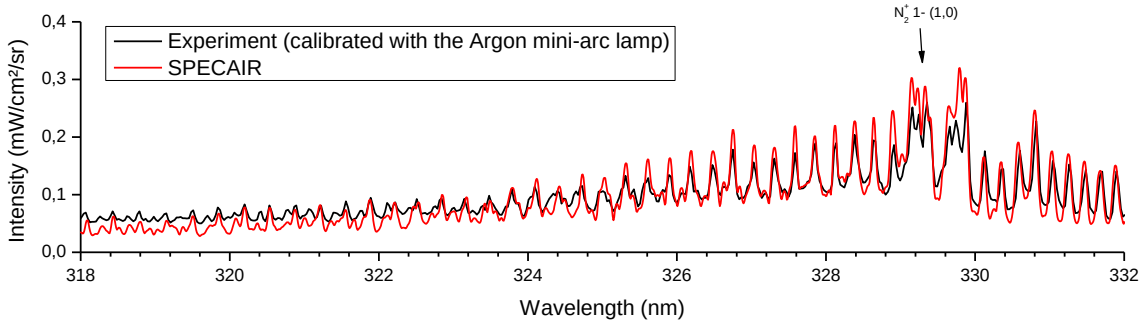

Figure 17: Measured and SPECAIR spectra between 318 and 332 nm

Between 273 and 280 nm (Figure 18), the spectrum mostly comprises the bands of NO Gamma, NO Beta, NO Delta, NO Epsilon, O_2 Schumman-Range, and N_2 (2+). Again good agreement is obtained, even though the signal-to-noise ratio is lower in this spectral range.

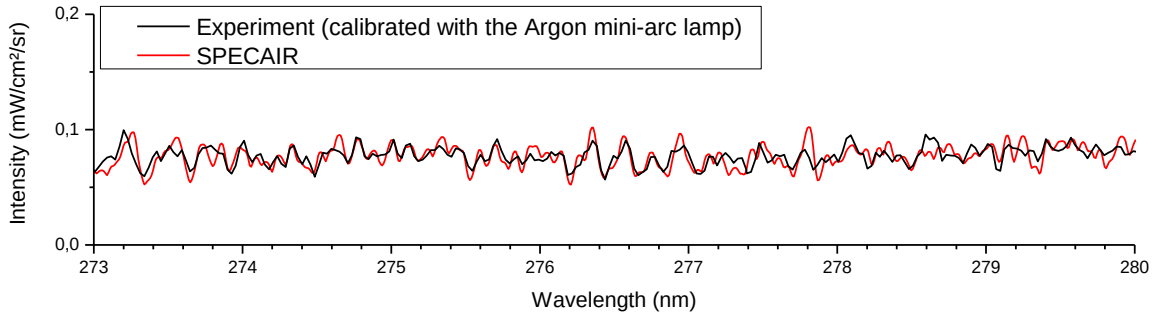

Figure 18: Measured and SPECAIR spectra between 273 and 280 nm

Figure 19 shows spectra between 245 and 250 nm, a range where the carbon line is present along with an underlying background of NO Gamma. A possible reason for the discrepancy could be a calibration uncertainty. However, it should also be noted that this difference may also be due to the uncertainty on the measured temperature (as mentioned earlier, a 3 % change of the temperature causes a 60% change in the intensity of the carbon line).

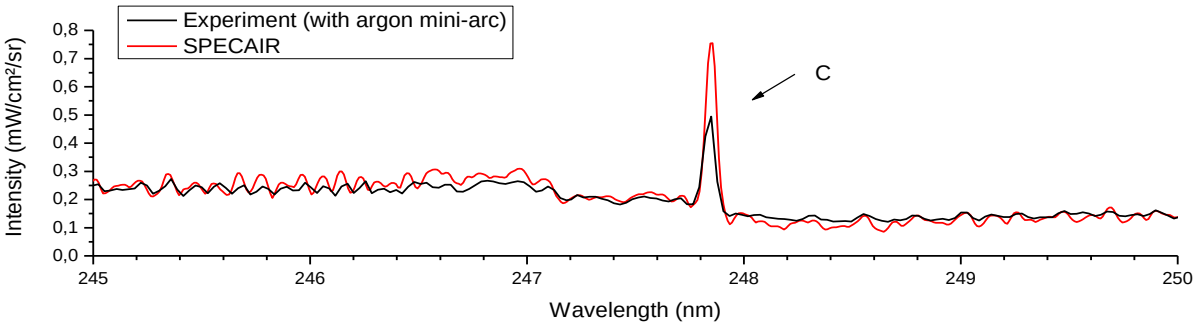

Figure 19: Measured and SPECAIR spectra between 245 and 250 nm

Figure 20 shows the spectrum from 239 to 245 nm. Here the spectrum is dominated by NO Gamma emission bands, which appear to be well modelled.

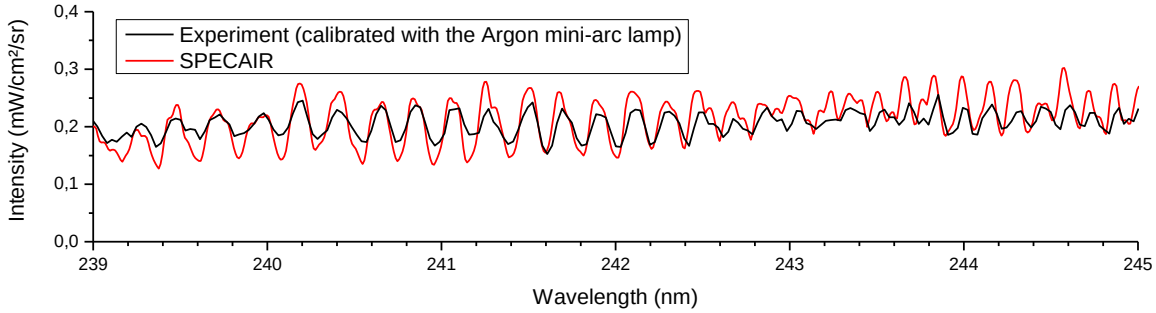

Figure 20: Measured and SPECAIR spectra between 239 and 245 nm

From 224 nm to 238 nm, seen in Figure 21, the two spectra are generally in good agreement. Here, the spectrum is dominated by NO Gamma emission bands, with contributions of NO Beta, Delta, and Epsilon bands that start being significant. The range between 224 nm and 227 nm shows some discrepancies that may be explained by the calibration as, over this wavelength range, the argon mini-arc does not emit strongly and the signal after background subtraction is very weak (only 22% of the measured signal, with the remaining 78% being straylight and electronic noise). This has a major influence on the spectrum. Further work could be done to improve calibration over this wavelength range.

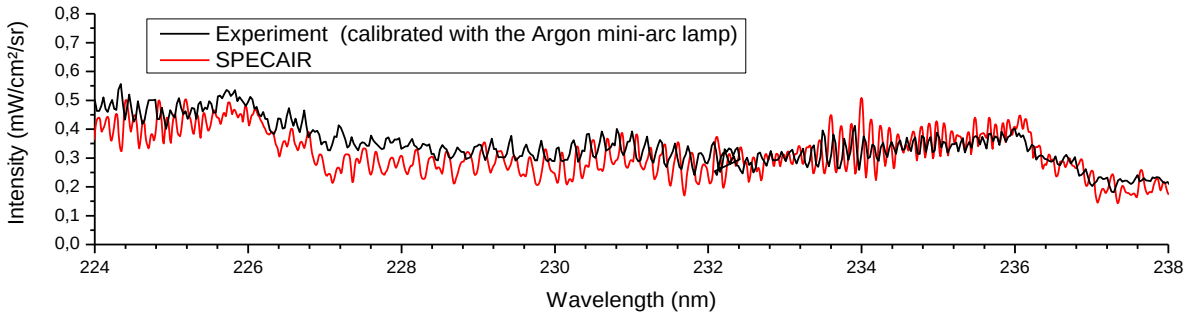

Figure 21: Measured and SPECAIR spectra between 224 and 238 nm

In the last range, shown in Figure 22, the measured spectrum is about 50% higher than the simulation, but the overall shape is well reproduced. The difference may be from the calibration as explained earlier. Also the high resolution spectrometer confirms the presence of either a potential missing additional species or uncertainties in the positions of some of the NO systems. Nevertheless, as can be seen by comparison with Figure 23, the NO Delta band system in particular appears to compare well with the experiments. The previously unexplained peak of SPECAIR at approximately 204.5 nm (Figure 1) is now better predicted but there still appears to be a difference at about 214.5 nm These spectra thus provide valuable information to test future models of the strongly perturbed NO transitions in the range 200 – 230 nm.

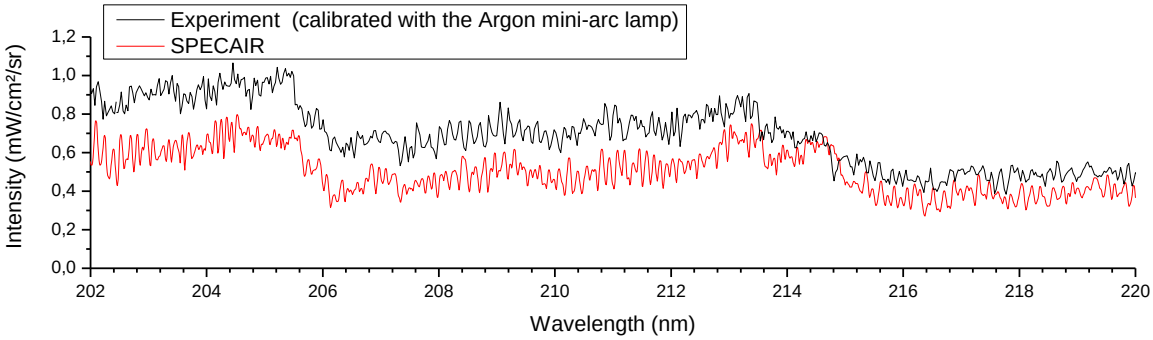

Figure 22: Measured and SPECAIR spectra between 202 and 220 nm

Figure 23 shows the shape and the intensity of each NO emission band. This permits further analysis of the previous spectra. It shows which species emit where. It could be useful to identify species that might not be well modelled in order to best fit the experimental spectra and the SPECAIR one over the range 200 – 300 nm. It is possible that some transitions are missing, and therefore further analysis is needed to improve our knowledge over this wavelength range.

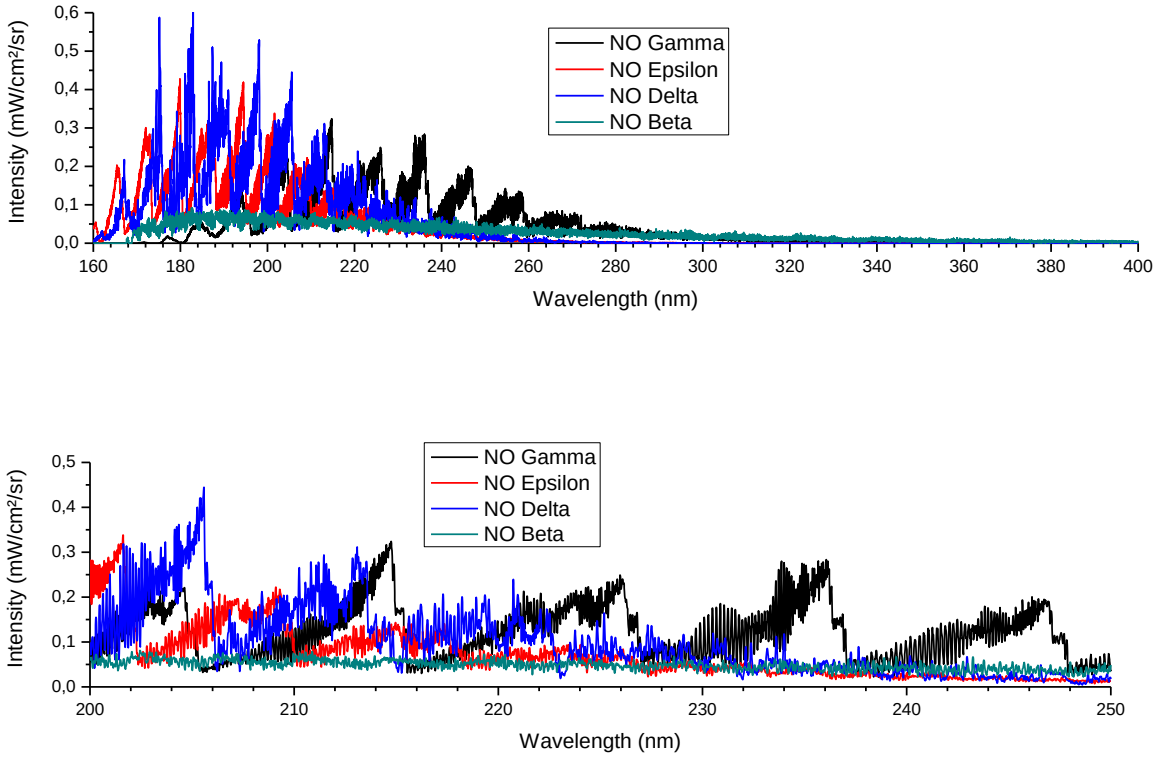

Figure 23: Contribution of different NO emission bands

4. Conclusion

High resolution emission spectra of an air plasma at a temperature of approximately 7500K have been measured and calibrated in absolute intensity. These spectra were compared with simulations of the SPECAIR code. Most transitions observed in the experimental spectra are very well reproduced, both in shape and magnitude, by SPECAIR. In particular, the spectra of the N_2 (2+), N_2^+ (1-), and CN Violet agree very closely with the experiment. A few differences are noted over narrow spectral ranges such as 202 – 220 nm, 224 – 238 nm, and 248 nm. Differences are mainly in the range 200 – 220 nm where the measured spectra correspond to the superposition of at least six band systems of NO: the beta, gamma, delta, epsilon, beta prime, and gamma prime transitions. Some of these bands are strongly perturbed by interactions between the various electronic levels. The SPECAIR model takes into account these perturbations by incorporating the coupled 138x138 hamiltonian of the B, C, L, K, and Q stated of Gallusser and Dressler [13]. Other models will be examined in the future to improve spectral predictions, as some transitions might not be known and implemented in SPECAIR. The present measurements will provide a useful guide to test these models.

5. References

[1] Laux, C.O., Spence T.G., Kruger, C.H., and Zare, R.N., 2003, "Optical Diagnostics of Atmospheric Pressure Air Plasmas," Plasma Sources Science and Technology, Vol. 12, pp. 125-138. www.spectralfit.com.

[2] Laux, C.O., 2007, "Spectroscopic Challenges in the Modelling and Diagnostics of High Temperature Air Plasma Radiation for Aerospace Applications". In: *Atomic and Molecular Data and Their Applications, 5th International Conference*.

[3] Laux, C.O, 1993, "Optical and radiative emission of high temperature air plasmas", Ph.D. thesis, Stanford University.

[4] NASA RP-1311, PART I, 1994 AND NASA RP-1311, PART II, 1996

[5] Mac Donald, M.E., C.M., Jacobs, C.O., Laux, 2013. "Radiation Shapes Thermal Protection Investigations for High Speed Earth Re-entry", ESA Report D4.2: Ablation-radiation studies.

[6] ASD Database, NIST, 2013.

[7] Laux,C.O., "Optical Diagnostics and Collisional-Radiative Models", VKI Course on Hypersonic Entry and Cruise Vehicles; Stanford University, June 30 – July 3, 2008.

[8] Packan, D.M., Laux, C.O., Gessman, R.J., Pierrot, L., and Kruger, C.H., 2003, "Measurement and Modeling of OH, NO, and CO_2 Infrared Radiation," Journal of Thermophysics and Heat Transfer, Vol. 17, No. 4, pp. 450-456.

[9] Laux, C.O., Gessman, R.J., Kruger, C.H., Roux, F., Michaud, F., and Davis, S.P., 2001, "Rotational Temperature Measurements in Air and Nitrogen Plasmas Using the First Negative System of N_2^+," Journal of Quantitative Spectroscopy and Radiative Transfer, Vol. 68, No. 4, pp. 473-482.

[10] Michaud, F., Roux, F., Davis, S.P., Nguyen, A.-D., and Laux, C.O., 2000, "High-Resolution Fourier Spectrometry of the N2+ ion," Journal of Molecular Spectroscopy, Vol. 203, pp. 1-8.

[11] Levin, D.A., Laux, C.O, and Kruger, C.H., 1991, "A General Model for the Spectral Calculation of OH Radiation in the Ultraviolet," Journal of Quantitative Spectroscopy and Radiative Transfer, Vol. 61, No. 3, pp. 377-392.

[12] Caillault, L., Andreasson, J., Risberg, J., and Laux, C.O., 2006, "Modeling the spectral radiation of a methane / nitrogen plasma. Test Case 4," 2nd International Workshop on Radiation of High Temperature Gases in Atmospheric Entry, Rome, Italy, September 5-8 2006.

[13] Gallusser, R. and Dressler, K., "Multistate vibronic coupling between the excited $^2\Pi$ stated of the NO molecule", *J. Chem. Phys.*, vol. 79, pp. 4311-4327, 1982.