Kinetics of a fast gas heating and $N_2(A^3\Sigma_u^+)$ measurements

E.I. Mintoussov\textsuperscript{1}, S.J. Pendleton\textsuperscript{2}, G.D. Stancu\textsuperscript{3}, D.A. Lacoste\textsuperscript{3}, C.O. Laux\textsuperscript{3}, N.A. Popov\textsuperscript{4}, S.M. Starikovskai\textsuperscript{a}

\textsuperscript{1} – LPP - CNRS UMR 7648, Ecole Polytechnique, France
\textsuperscript{2} – University of Southern California, USA
\textsuperscript{3} – EM2C – CNRS UPR 288, Ecole Centrale Paris, Chatenay-Malabry, France
\textsuperscript{4} – Scobeltsyn Institute of Nuclear Physics, Moscow State University, Russia

Abstract: In the present work a set of experiments combining (i) electrical measurements of current, voltage, reduced electric field and electron density in a nanosecond time scale, (ii) gas temperature and densities of $N_2(A^3\Sigma_u^+)$ metastable in a microsecond time scale has been done. Experiments to determine the temperature in the discharge and in the discharge afterglow were performed using emission spectroscopy methods, and $N_2(A^3\Sigma_u^+)$ concentration was measured using CRDS technique.

Keywords: nanosecond discharge, kinetic scheme, fast gas heating, cavity ring-down spectroscopy

1. Introduction

So-called fast gas heating, which is the energy release in the discharge or post-discharge due to the relaxation of energy stored in the electronic degrees of freedom, has been investigated for decades.

The detailed kinetic mechanism has been developed and verified for $N_2:O_2$ mixtures for $E/N$ less than 250 Td [1]. The experimental evidence of a fast energy release has been demonstrated in pulsed discharges, in particular, in a surface nanosecond barrier discharge used for flow control in aerodynamics [2] and in pulsed nanosecond pin-to-pin discharge in heated air [3].

Nevertheless there is no any predictive model describing such energy relaxation at high electric fields. The aim of our study is to develop experimental approach allowing key for fast gas heating parameters and species to be measured.

2. Experimental setup

To study fast energy release in the afterglow of a pulsed discharge at high electric field, we suggested a high-voltage nanosecond discharge in the form of a fast ionization wave (FIW) to be used. The main reasons for that are the discharge spatial uniformity, high values of reduced electric field during the discharge development (100-1000 Td), short duration of the discharge allowing separation of the discharge action and of the relaxation process and the fact that FIWs are well studied [4].

The following parameters are suggested to be measured during the discharge propagation: current and voltage waveforms, energy input, reduced electric field vs time and electron density. These experiments must be combined with two-photon absorption laser induced fluorescence (TALIF) measurements for O-atoms density and cavity ring-down (CRDS) measurements for $N_2(A^3\Sigma_u^+)$ density. At present preliminary results for temperature increase and $N_2(A^3\Sigma_u^+)$ behaviour in the discharge afterglow have been obtained.

The discharge setup (fig.1) has been developed and tested. The discharge cell consists of a quartz tube with hollow metal electrodes (the diameter of the orifice in each electrode is 5 mm), the distance between electrodes is 20 cm, and the tube diameter is 9 mm. High-voltage pulses of 10 kV amplitude and 30 ns duration with pulse repetition rate 2 Hz were used. The tube is connected to two glass side tubes with valves to pump and fill the system with a
gas. The end plates of the glass tubes are the CRDS mirrors separated by 78 cm. The working pressures were 3-9 mbar. All the experiments were performed with a gas flow (synthetic air or pure nitrogen) with typical flow rate 10-30 sccm, enough for mixture renewing between high-voltage pulses. The uniformity of the discharge between the electrodes has been checked, it was proved that the discharge is mainly confined between the electrodes without significant penetration to the side tubes.

3. Temperature measurements

To measure the temperature, we used auxiliary high-voltage pulses. These pulses were the pulses reflected from the end of the additional cables, 100 m and 200 m length, which were connected to the low-voltage electrode of the discharge cell.

The excitation of the N₂ molecules takes place via direct electron impact, so the temperature obtained from rotational structure was equal to the gas temperature divided by the ratio of rotational constants B of the excited and ground states. Using this technique, we were able to get the values of temperature 1.5 and 2.5 μs after the discharge.

The results of the temperature measurements in air for the pressure of 6 mbar are presented in fig. 2. It is clearly seen a significant (40-60 K) temperature increase during first 1-1.5 μs and then additional heating of approximately 20 K in a 1 μs. The calculations show a reasonable agreement with the experiment.

4. N₂(A³Σ⁺) measurements by cavity ring-down spectroscopy

N₂(A³Σ⁺) concentration was measured by CRDS technique. Absorption in the 2-0 vibrational transition of first positive system of molecular nitrogen was used to get the population of 0, 1 and 2 vibrational levels of N₂(A³Σ⁺). In order to achieve high sensitivity the choice of the absorption transition is important. The CRD signal was measured at the maximum of the unresolved Q₁(22)
and $Q_3(16)$ lines structure at the wavelength of 769.945 nm, as shown in ref [6].

The typical CRD experimental signals with and without discharge are presented in fig. 3. When $N_2(A^3\Sigma_u^+)$ metastable was present in the system, the CRD signal decay occurs faster giving the information about the concentration of the absorber.

Experimental setup allowed us to vary the delay between high-voltage pulse, where production of $N_2(A^3\Sigma_u^+)$ occurs, and laser pulse, so the concentration could be measured in a wide region (hundreds of $\mu$s). The typical decay time of the empty cavity was about 2 $\mu$s. The metastable density was considered constant during the decay time. The fast processes of $N_2(A^3\Sigma_u^+)$ formation (during the discharge pulse and shortly after) were beyond the limits of our investigations. The discharge emission and the EM noise have constrained the CRDS experiments. The measurements were performed starting about 3 $\mu$s after the discharge pulse.

The absorption line FWHM was determined mainly by the Doppler broadening, which is in our case 1.8-2.1 pm (depending on the temperature in the afterglow). These values are significantly higher than the collisional broadening, which is 0.1-0.4 pm for the working pressures of 3-9 mbar.

The laser FWHM was 6 pm, which is greater than the absorption line width. In this case the classical mono-exponential approximation of CRD signal

$$I(t) = I_0 \cdot \exp \left[ -\frac{c}{L} \left( (1-R) + k(v_0)I_{\text{abs}} \right) t \right]$$

is not applicable, because we need to take into account not only the absorption coefficient $k(v_0)$ at the line intensity maximum, but all the coefficients along the line profile, each with its own laser intensity $I(v)$ (detailed information about this aspect of CRDS could be found, for example, in [7]). Thus, CRD signal should be described by the following equation:

$$I(t) = \sum_m I_0(v) \cdot \exp \left[ -\frac{c}{L} \left( (1-R) + k(v)I_{\text{abs}} \right) t \right]$$

where the summation goes over all cavity modes.

The free spectral range of cavity in our case is approximately 0.4 pm, so we have enough longitudinal modes under the absorption line profile to replace summation by integration. Moreover, because of signal averaging, we have a displacement of mode positions from pulse to pulse because of mirrors microvibration, so we can treat modes pattern as a quasicontinuum.

A numerical code was used to simulate this multi-exponential approximation, the results are presented in fig 4. Good correlation between experimental and simulated spectrum was obtained in a wide range of absorption coefficients.

The preliminary results for the effective absorption coefficient $K$, which is directly proportional to the $N_2(A^3\Sigma_u^+, v=0)$ concentration are shown in fig. 5 (synthetic air) and fig. 6 (pure nitrogen). The decay in air occurs significantly faster (with typical times 10-20 $\mu$s, depending on the pressure) than in
nitrogen because of the effective \( \text{N}_2(^3\Sigma_u^+) \) quenching on the oxygen molecules. The decay rate increases with the pressure increase.

**Figure 5.** Absorption coefficient for \( \text{N}_2(^3\Sigma_u^+, v=0) \) in air

In nitrogen, the absorption coefficient decays slower (40 \( \mu \)s) and depends weakly on the pressure. If this is plotted in \((1/K, t)\) coordinates, the dependence becomes linear. This fact indicates that the main channel of \( \text{N}_2(^3\Sigma_u^+) \) quenching in pure nitrogen is the reaction between two \( \text{N}_2(^3\Sigma_u^+) \) molecules: \( \text{N}_2(^3\Sigma_u^+)+\text{N}_2(^3\Sigma_u^+) = \text{N}_2(B^3\Pi_g,C^3\Pi_u)+\text{N}_2(X^1\Sigma_g^+)\).

**Figure 6.** Absorption coefficient for \( \text{N}_2(^3\Sigma_u^+, v=0) \) in nitrogen

### 5. Conclusions

In the present work, an experimental scheme has been proposed to validate the kinetic mechanism of fast gas heating. Experiments were performed in air and nitrogen at 3-9 mbar. A temperature increase was measured by emission spectroscopy in the afterglow of a pulsed nanosecond discharge, at delay times of 1.5 and 2.5 \( \mu \)s. The typical values of heating for this timescale are 60-80 K. The calculations are in reasonable agreement with experiments. Preliminary CRDS experiments have been performed. They demonstrate that \( \text{N}_2(^3\Sigma_u^+) \) concentration decay in nitrogen occurs significantly slower than in air because of the different kinetic mechanisms.

### 6. Acknowledgements

This work was partially supported by ANR (RAMPE and PREPA Projects) and EOARD AFOSR, grant FA8655-09-1-3077.

### References


