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Foreword

The International Conference Series on Gas Discharges and Their Applications was formed in London by the Institute of Electrical Engineers in 1970. Overall control of the Series now rests with the Executive Management Committee. They can take advice on various matters from the International Scientific Committee, which is newly formed for each conference from leaders in the various scientific disciplines. Conferences have been taken to many universities in several countries where there are scientific communities involved in research into the behaviour of electricity in gases. Individual conferences are run by the Local Organizing Committee which is formed for each event.

The format of these conferences is now well established. Abstracts for suggested papers are submitted to the organizers of each conference for refereeing by a panel of internationally recognized experts formed by the Local Organizing Committee. These are reviewed for their scientific content and accuracy as well as their relevance to the conference topics listed in the conference literature. Authors of the accepted abstracts are then invited to write cognate papers which are again refereed by the scientific panel prior to their publication in the proceedings. It is a conference tradition for all accepted papers to be orally presented at each conference.

This publication contains the written versions of the accepted papers for the Cardiff conference. They are published in the order of the listed scientific topics and their presentation at the conference sessions, starting with the written versions of the plenary lectures given at the conference. The responsibility for the content of the papers rests entirely with their authors. An electronic version of these proceedings is also available.

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MEASUREMENTS OF $N_2(A)$ BY PULSED CAVITY RINGDOWN SPECTROSCOPY IN REPETITIVELY PULSED NANOSECOND DISCHARGES

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ABSTRACT

We present here results from the measurements of metastable $N_2(A)$ species in nanosecond repetitively pulsed (NRPP) discharge in N_2 and air, obtained by using pulsed cavity ring-down spectroscopy (CRDS) at 770 nm.

Despite the small plasma volume (1-10 mm^3) and short lifetime of studied plasma species, CRDS enabled us to obtain spectra with spectral resolution down to 1 pm. The temporal variations of $N_2(A)$ concentration in a nitrogen plasma were measured and absolute concentrations of $N_2(A)$ in nitrogen and air plasmas were determined. These results were then used to validate a kinetic model, which already showed good agreement with the spatial and temporal evolution of other excited state species such as $N_2(B)$, $N_2(C)$, $N_2^+(B)$, NO(A) and O(3p ^{5}P) measured using intensity calibrated emission spectroscopy.

1. INTRODUCTION

Atmospheric pressure air plasmas are widely used for surface treatment and activation, exhaust gas control, aerodynamic flow control, plasma assisted combustion, and destruction of toxic compounds [1]. The efficiency of these processes strongly depends on the energy requirements of the plasma sources employed. Here, we used a nanosecond repetitively pulsed (NRPP) discharge generated in a pin-to-pin electrode configuration in atmospheric pressure air, characterized by energy requirements of a few W/cm^3 for average electron densities of about 10^{12} cm^{-3}. Corona, diffuse and filamentary regimes of NRPP discharges have been observed, depending on the applied voltage (5-8 kV), repetition frequency (2-30 kHz), gas temperature (300-1000 K), air flow rate (1-17 m/s), and gap distance (1-7 mm) [2,3].

Among numerous possible applications of NRPP, plasma-assisted combustion was studied recently [4], showing significant effects on flame stability for lean mixtures. Based on results of the kinetic model presented in [3], it was assumed that the stabilization effects may be related to the high concentration of O atoms generated by the NRPP. Consequently, we have investigated the production of atomic O via dissociative quenching of O_2 by excited nitrogen species N_2(A, B, C) [3]. The spatial and temporal evolution of the excited state species $N_2(B)$, $N_2(C)$, $N_2^+(B)$, NO(A) and O(3p ^{5}P) were obtained using intensity-calibrated emission spectroscopy [3].

We are currently testing laser diagnostic techniques to detect the other important species that are not accessible via emission spectroscopy. Two promising techniques are Two-Photon Laser Induced Fluorescence (TALIF) at 225 nm with fluorescence at 844 nm [5] for detection of atomic oxygen in the ground state, and pulsed Cavity Ring-Down Spectroscopy (CRDS) at 770 nm for the detection of $N_2(A)$ species.

CRDS has become a widely used method owing to its superior sensitivity which makes it particularly well suited for the measurement of minor species in plasmas. Detailed descriptions of the technique have been presented elsewhere [6]. The technique may be summarized as follows [7]. A laser beam is coupled into a high finesse optical cavity containing an absorbing sample. The light inside the cavity decays (ring down) owing to cavity loss (primarily mirror losses) and sample absorptive loss. A photodetector is used to measure the ring-down signal. The signal profile as a function of time is fitted to yield the absorptive loss. Spectra may be obtained by scanning the laser wavelength.

2. EXPERIMENTAL SETUP

The experiments were carried out in atmospheric pressure air and nitrogen flows preheated to 1000 K, with axial flow rate up to 30 l/min. The plasma discharge was produced by applying nanosecond high-voltage pulses between two refractory metallic electrodes. The distance between these electrodes in point-to-point configuration was 3 mm.

The pulses are produced by a solid-state generator (FID Technologies FPG 10-30MS) that produces pulses with rise/fall times of 5 ns, a flat top of 10 ns at greater than 90% of the maximum voltage, and amplitudes up to +10 kV into an open circuit. The discharge voltage was measured with a high voltage probe (LeCroy PMK-14kVAC) and the discharge current was measured using a current probe Pearson Electronics 2877, connected to a 350 MHz digitizing oscilloscope LeCroy Waverunner 434 (maximum sampling frequency 2GS/s).

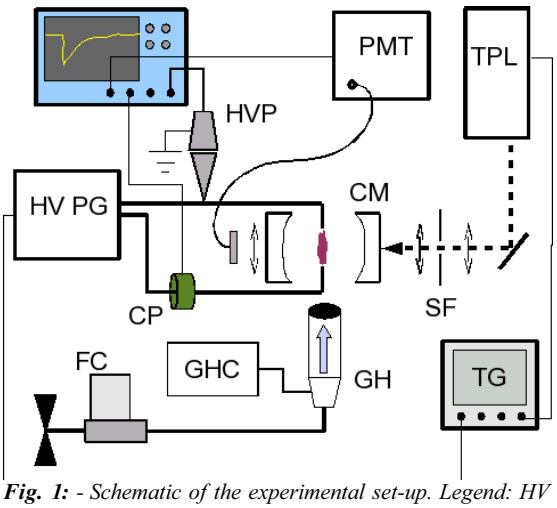

Fig. 1: *- Schematic of the experimental set-up. Legend: HV PG – high voltage pulse generator, HVP – high voltage probe, CP – current probe, PMT – photomultiplier tube, CM – cavity mirror, FC – flow controller, GH – gas heater, GHC – gas heater control box, TG – trigger generator, TPL – tunable pulsed laser.*

The cavity consists of two concave mirror with reflectivity $R > 99.97\%$ (0°, 770 nm). The distance between mirrors is 70 cm. The cavity is fed by a tunable pulsed dye laser Continuum ND 6000 pumped with a Continuum Precision PL 8010 Nd:YAG laser, with energy per pulse up to 40 mJ at 770 nm, and repetition frequency 10 Hz. The light collected after the second cavity mirror is detected with a photomultiplier tube (PMT) with 1.4-ns rise time (Hamamatsu H9305-3) and recorded on the oscilloscope. The collected light is filtered by two bandpass interference filters (CVI) with peak transmittance at 770 nm. Synchronization is controlled with a Berkeley Nucleonics (BNC) 555 four-channel pulse delay generator, which triggers all devices. The experimental set-up is depicted in Fig. 1.

3. RESULTS AND DISCUSSION

In the present study, we examine a discharge operating in gas preheated at 1000 K. The discharge repetition frequency is 10 kHz, and the average current is 60-70 mA. For these conditions, the discharge is filamentary,

characterized by a high conduction current. Fig. 2 shows typical current and voltage waveforms measured in this discharge regime.

Figure 3 shows a sample recorded CRDS signal, where three distinct regions can be defined. The first region is the part of the signal before the HV pulse, thus before the creation of the $N_2(A)$ species. The slope of the signal during this period defines the empty cavity decay time.

The second region corresponds to a narrow temporal interval around 160 ns, where large spikes due to the electromagnetic (EM) noise generated by the 10 ns high voltage pulse (ringing effects) can be observed. The PMT detection system being sensitive to EM fields, no data points can be used for extracting the decay time during this period.

The third region, starting immediately after the spikes, lasts for about 5 µs. Actually, there is another short period with EM noise during this third region, corresponding to a secondary voltage pulse. However, this second spike is not very significant and the decay signal can be interpolated after extracting this noise.

The difference between the measured off-resonance signal in region 3 and the extrapolated signal of region 1 (corresponding to an empty cavity with negligible absorption loss) gives information about the total absorbance of the plasma, including effects related to absorption by $N_2(A)$ species, but also to beam-steering and scattering losses inside the cavity. Beam-steering is caused by the large temperature difference between ambient air and the plasma (whose temperature can be higher than 1000 K), which causes changes in the refractive index of air. As result the laser beam tends to "walk-away" from the cavity and faster decay is measured. We verified that these effects have negligible wavelength dependence for the spectral region scanned (769.82 - 770.75 nm), whereas absorption is

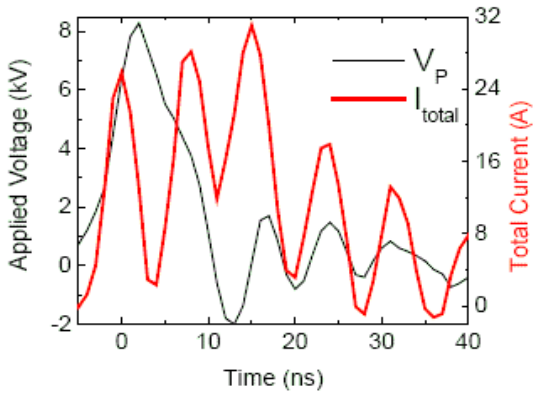

Fig. 2: *- Typical measured current (thick) and voltage (thin) waveforms of filamentary discharge regime [3].*

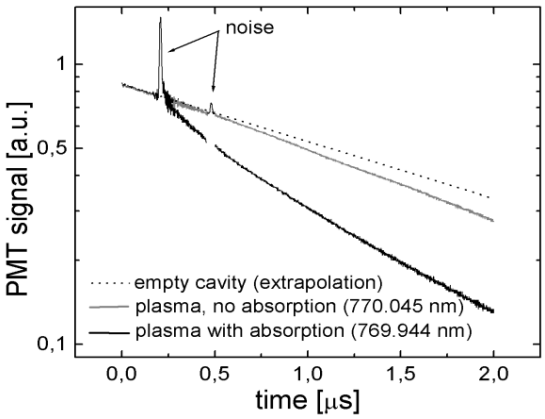

Fig. 3: *- Typical CRDS signal in N_2 plasma.*

present only if the laser is tuned to a specific $N_2(A)$? $N_2(B)$ transition. Fig. 4 shows a comparison of absorption coefficients measured by CRDS and simulated with Specair [8] for translational, rotational and vibrational temperatures equal to 1000 K and for an absorption path length of 1 mm.

When the laser is tuned to 770.045 nm (an off-resonance wavelength with no absorption), the plasma-laser interactions reduce the decay time from 2 µs to approximately 1.75 µs, but the signal in logarithmic scale remains linear (Fig. 3). This means that the intensity of these effects does not change significantly during at least 2 µs. On the other hand, when the laser wavelength is set to 769.944 nm (strongest absorption), the resulting signal is no longer linear. Moreover, the slope of this signal decreases in time. This means that the absorption decreases owing to the decreasing concentration of $N_2(A)$ species (Fig. 5).

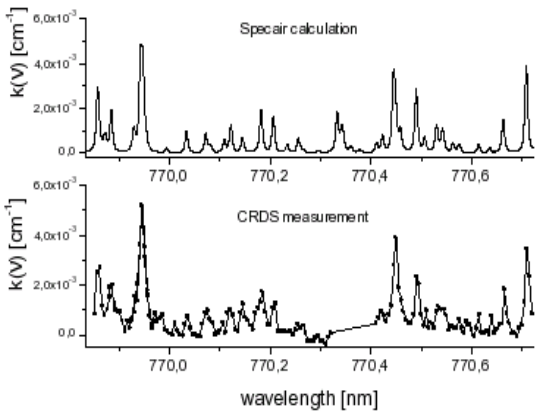

Fig. 4: *- Comparison of the CRDS measured (down) and Specair (up) calculated absorption coefficients (N_2 B-A transition).*

According to to the measured CRDS decay times, the maximum concentration of $N_2(A)$ for the NRPP filamentary regime in nitrogen with a mean current 60 mA was approximately 80 ppm and the life time of $N_2(A)$ is around 900 ns. This result is in good agreement with modeling, which predicted the life time of $N_2(A)$ in the nitrogen plasma to be ~1 μs.

The situation is different in the air plasma, where the intensity of the CRDS signal decreases due to absorption mostly during the region with strong EM noise. It was therefore impossible to observe changes of $N_2(A)$ concentration in time. This is also in good agreement with modeling, which predicts the lifetime of $N_2(A)$ in air plasma to be 200 ns. Thus, we could only estimate that for NRPP filamentary regime with mean current 70 mA, the average $N_2(A)$ concentration during the EM noise period was around 26 ppm.

4. CONCLUSIONS

A nanosecond repetitively pulsed discharge with repetition frequency of 10 kHz in atmospheric pressure air and nitrogen has proven to be very efficient for the generation of reactive species. High concentrations of $N_2(A)$ in nitrogen (~80 ppm) and air (~26 ppm) were observed. According to our preliminary results, we achieved a detection limit below 1 ppm.

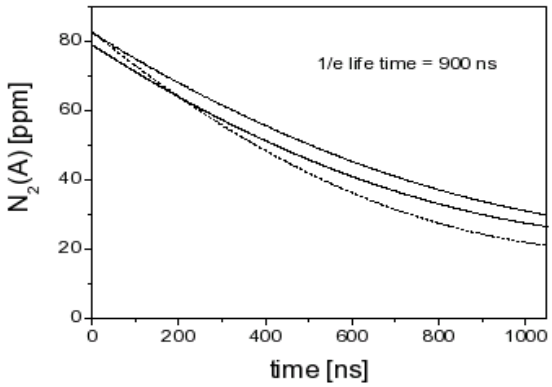

Fig. 5: *- Time evolution of concentration of $N_2(A)$ species in N_2 discharge with average current 60 mA.*

In the next step, parametric studies of the efficiency of different discharge regimes for the generation of $N_2(A)$ are planned in order to determine the concentration of atomic oxygen produced by the discharge via dissociative quenching of O_2 by $N_2(A)$. A further reduction of the electromagnetic noise level after the HV pulse is however required to obtain the time evolution of $N_2(A)$ in air.

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