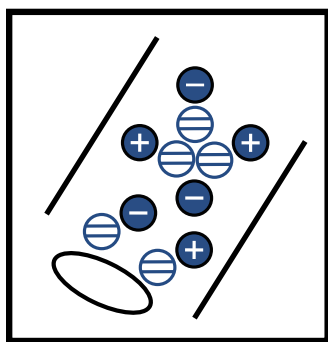




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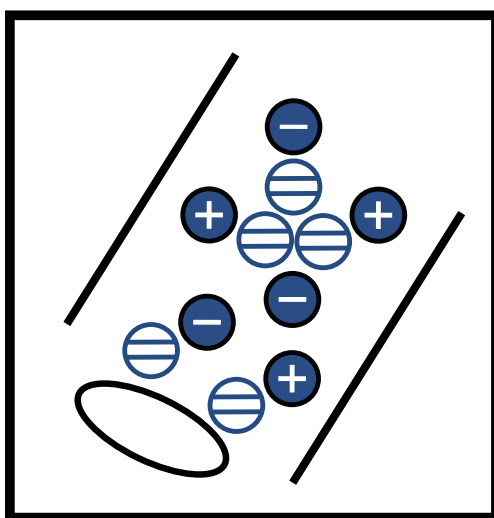
FLTPD XIV

Frontiers in Low Temperature Plasma Diagnostics

1 - 5 May, 2022
Levico Terme, Italy

BOOK OF ABSTRACTS

Book of Abstracts



FLTPD-XIV

Frontiers in Low Temperature Plasma Diagnostics

1 – 5 May 2022

Levico Terme – Italy

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Dear colleagues.

We warmly welcome the participants of the 14th Workshop on Frontiers in Low Temperature Plasma Diagnostics (FLTPD) in Levico Terme from Sunday 1st to Thursday 5th of May 2022, at the Hotel Bellavista.

This edition is the continuation of a biennial series that began in 1995 at Les Houches (France). This year the FLTPD XIV is organized by the Atomic and Molecular Physics Group - Department of Physics of the Trento University - and the Institute for Plasma Science and Technology of the Consiglio Nazionale delle Ricerche. For the second time, FLTPD has been organized in Italy, after the fifth edition in 2003, in Villaggio Cardigliano, Specchia (Lecce).

The workshop offers the opportunity to present recent results on plasma diagnostics and aims to bring together experts in low-temperature plasma diagnostics. It is an important and fruitful opportunity for the new generation of plasma scientists to share and discuss the knowledge of these diagnostics with the leading scientists in the field.

The program consists of expert presentations from 9 invited speakers, 16 topical speakers, and posters in line with the previous meetings.

The number of researchers participating at the FLTPD XIV is close to 60, from 16 different countries.

This edition is shifted by one year with respect to the usual biennial cadence in odd years since we had to postpone the 2021 natural date due to the unfortunate occurrence of the Covid-19 pandemic. Although the virus has not yet been completely defeated, there are sufficient safety conditions to allow the workshop to take place this time. We have been especially pushed by our compelling will to meet again in our warm, friendly atmosphere and restart our activity in favor of the young generations.

These are difficult times. Above all, by the time we are writing this foreword, a horrible war is afflicting Europe, touching also many friends of our community. We nevertheless believe that, from our small world, we can contribute to raising the brotherhood and collaboration message of science, the flag of intellect against the stupidity of blind violence. Our thoughts are with all war victims, especially our colleagues and friends in both countries, whose work and

life have been affected by the recent senseless escalation of violence and oppression.

Finally, we express our gratitude to the members of the International Scientific Committee for the organization of the scientific program and the Local Organizing Committee members for all their efforts in preparing and organizing the meeting. Also, we acknowledge the Department of Physics - University of Trento, the ISTP-CNR, and the Sponsors for financial support.

On behalf of the ISC and LOC, we wish the participants an interesting scientific meeting, fruitful discussions, new scientific ideas, and a pleasant stay in Levico. We want to thank all the participants in this event, whose contribution is crucial for the progress of science in our field.

Nader Sadeghi and Uwe Czarnetzki
chair and vice-chair of the
International Scientific Committee

Paolo Tosi, Giorgio Dilecce, and Luca Matteo Martini
chair and co-chairs of the
Local Organizing Committee

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The 14th Frontiers in Low Temperature Plasma Diagnostics workshop is organized by the Atomic and Molecular Physics Laboratory (University of Trento) and the Institute for Plasma Physics and Technology (CNR-Bari).



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General Information

Oral presentations

For participants with a Topical lecture presentation, please prepare presentations of 20 minutes (+ 5 minutes of discussion). Invited presentations have a length of 40 minutes (+ 10 minutes of discussion). Please bring a USB stick with your presentation before your session starts. Of course, your presentation will be deleted directly after the end of your session, so that your data are handled safely.

The detailed program can be found on the workshop homepage:

<https://event.unitn.it/fltpd2022/>



Poster presentations

The suggested size for the poster is A0 in portrait orientation. The poster sessions are planned after the dinner on Monday and Tuesday. Please make sure that your poster is put on the board well before the beginning of the poster session. We have assigned a number to each poster (see on page **30** and **48**). Please use the board with the corresponding number to hang your poster.

The excursion

The excursion is planned for Wednesday May 4th right after lunch. The excursion will be a scenic walk.

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Invited lectures

New diagnostics for microparticles in plasmas

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1. Microparticles in plasmas

A typical dusty plasma consists of micrometer-sized particles embedded in a low temperature and low pressure plasma. Thus, its properties are determined by the electrons, ions and microparticles which charge up to several thousand elementary charges. In such a system new and interesting structural and dynamical processes can be observed.

The research field of dusty plasmas emerged about 30 years ago. Since then the field made considerable progress and contributed to many fields beyond plasma physics. Strongly coupled systems, soft matter, and statistical mechanics are just a few examples. One of the reasons for this success was that powerful diagnostics have been developed to measure the characteristic parameter of a dusty plasma. Two key parameters are size and charge of the microparticles. This contribution will introduce novel diagnostic techniques to determine size and charge and show how these diagnostics can be utilized to address new research areas.

2. Particle size diagnostic

Using commercially available monodisperse particle samples one can achieve reasonable control on particle size. However, as soon as reactive gases are used to grow particles a size diagnostic is needed. In addition, a number of experiments demonstrated that even in an argon discharge some particle materials are subject of sputtering/etching processes which caused changes of particle size.

An unknown or not well known particle size is a severe problem because the particle size determines the absolute value of all forces acting on the particles. Further, these forces scale to different power with the particle size. Thus, the force balance in a dusty plasma sensitively depends on particle size and thus structure and dynamics will be directly affected. Thus, a precise knowledge of size is the foundation of any precise measurement in a dusty plasma.

The first part of this talk will introduce a Mie-scattering technique, where the light scattered by a microparticle is used to determine the particle size with high precision in-situ, i.e. during plasma operation [1]. While Mie-scattering is a well known

technique, our approach includes a number of new features which allow to reach precision in the order of nanometer and determine other optical properties of the particle as well. It is shown that this new diagnostic enables us to study plasma surface interaction as well as dynamical processes.

3. Particle charge diagnostic

The second very important parameter is the particle charge. Charge measurement techniques have been developed and improved since the early days of dusty plasmas. The most commonly used methods are resonance methods for few particles or wave dispersion measurements for many particle systems. However, these methods are so far limited to monodisperse particle systems. The second half of this contribution will therefore aim at charge measurement in binary systems, i.e. systems that consist of two particle species with different size and thus charge [2]. The results of two complementary approaches are compared. The first is the so-called configurational temperature method [3,4]. The second is a wave dispersion measurement [5]. Finally, the impact of these diagnostics on the newly emerging field of binary systems is briefly discussed.

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Probing Species Fluxes, Plasma Instabilities and Boundary Layers in Atmospheric Pressure Plasmas

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1. Introduction

The unique non-equilibrium conditions of low temperature plasmas enable the delivery of highly reactive plasma species to surfaces. Advances in the generation and control of atmospheric pressure plasmas enabled the extension of their use in the treatment of a broader range of heat sensitive substrates including polymers, tissues, plants, and wounds. This technological advancement not only led to the emergence of many new applications but also to several new scientific questions and major challenges for diagnostics. While plasma-surface interactions at low pressure are often due to energetic ion etching, at atmospheric pressure, the ion mean free path is much smaller than the sheath thickness resulting in insufficient ion energies. Hence, at atmospheric pressure, neutral reactive species and radicals often play an important role in plasma-surface reactions. In addition, sheaths and boundary layers at atmospheric pressure occur on length scales of 100 μm or less. Furthermore, plasma instabilities readily occur at atmospheric pressure introducing high temporal changes in the plasma dynamics. We will discuss these unique challenges in the context of species flux measurements including boundary layer effects to quantify plasma surface interactions and the measurement of highly dynamic changes in plasma kinetics triggered by plasma instabilities.

2. Results

2.1. Molecular beam mass spectrometry (MBMS)

MBMS enables directly linking the obtained fluxes with plasma-surface interaction studies, owing to its ability to detect a large range of different species and to measure species fluxes or densities at a substrate. We have recently extended the capability of MBMS for plasma diagnostics by developing detection and calibration approaches for the absolute measurement of singlet oxygen, vibrationally-excited nitrogen $\text{N}_2(\text{v})$ as well as the absolute density of positive ions in the effluent of atmospheric pressure plasma jets. We will show that these extended capabilities enable us to develop strategies to control reactive species fluxes to a substrate in the effluent of an RF-driven Ar-O_2 plasma jet as a tool to study plasma-surface interactions [1].

2.2. Boundary layers, droplets and bubbles

While MBMS is an excellent tool to study plasma surface interactions, it cannot be applied to the study of interaction of plasmas with liquids. We will show examples of spatially resolved OH radical density measurements near liquid surfaces including droplets and bubbles [2]. These measurements enable the quantitative study of plasma-liquid interactions. Accessing sheath and boundary layer length scales of $\sim 100 \mu\text{m}$ remains a huge challenge for diagnostics. Resolving such near surface gradients is particularly important for the so-called surface discharges that propagate along the substrate, a common phenomenon at atmospheric pressure. We will report OH radical species density measurements with a spatial resolution of $\sim 10 \mu\text{m}$ perpendicular to a substrate in a surface streamer discharge. This microscopic OH LIF measurement allowed to accurately determine OH radical fluxes to interfaces and even sticking coefficients.

2.3. Tracking and measuring electron kinetics to understand kinetics and plasma instabilities

Diffuse discharges at atmospheric pressure often have electron densities below 10^{19} m^{-3} and higher density plasmas typically exhibit contraction and large spatial gradients requiring a temporal and spatially resolved diagnostic, such as Thomson scattering, to investigate plasma kinetics [3]. We will show examples of such measurements including tracking the electron kinetics during the contraction of a discharge channel and highlighting the impact on radical production.

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Diagnosing H₂/D₂ helicon plasmas in RAID

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1. Introduction

In the Resonant Antenna Ion Device (RAID) [1] at the Swiss Plasma Center, we investigate high-power (up to 20kW), steady-state helicon plasmas for a variety of applications, ranging from negative ion production for beam generation to plasma thrusters. A schematic of RAID is shown in Fig 1. RAID is a linear device (1.4 m length, 20 cm radius) in which magnetized plasma columns are created by helicon wave excitation using twin resonant antennas at 10 kW each in a birdcage-geometry operating at 13.56 MHz. A set of 6 independently powered copper coils produce a magnetic field up to 800 G on-axis. Many gases can be used such as H₂, D₂, Ar and He, and typical electron densities of 10¹⁸ m⁻³ and 10¹⁹ m⁻³, respectively, in hydrogen and argon can be attained with electron temperature in the range of a few eV. A number of diagnostics have been implemented to advance the physics understanding of H₂/D₂ helicon plasmas. Here, we will discuss their technical implementation on RAID and the experimental challenges in a high-power helicon plasma environment as well as selected physics results.

2. Diagnostics for H₂/D₂ plasmas on RAID

Optical emission spectroscopy (OES) measurements in RAID interpreted using a collisional radiative model (CRM) [2] first suggested a complex chemistry of H₂ and D₂ plasmas. In particular, a radial shell was observed where negative ions (H⁻/D⁻) are produced by dissociative attachment with significant absolute densities. Recently, numerical simulations confirmed the richness of H₂/D₂ plasma chemistry [3]. This experimental and numerical evidences stimulated the development of two direct diagnostic techniques for H⁻/D⁻, which have rarely been employed combined to probe helicon plasmas: Cavity-Ring Down Spectroscopy (CRDS) and Langmuir probe Laser Photodetachment (LPLP) [4].

In H⁻ or D⁻ ions, the extra electron is weakly bound to the atom (binding energy=0.75 eV). In RAID, we employ a pulsed Nd:YAG laser with pulse duration ~5 ns whose photon energy (=1.2 eV, corresponding to a wavelength of $\lambda = 1064$ nm) is large enough to strip the weakly bound electron of H⁻ by photodetachment ($\nu + \text{H}^- \rightarrow \text{H} + e$). In CRDS, the laser beam is injected inside an optical cavity made of two high reflectivity mirrors between which the light undergoes multiple reflections, thus multiplying the

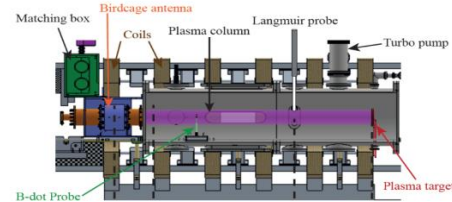


Fig. 1: A schematic of RAID with main ancillaries.

interaction length of the light with the absorbing medium. The laser signal leaking through the cavity consists of an exponentially time-decaying signal, whose time constant is inversely proportional to the line-integrated negative ions density. However, CRDS at a fixed measurement position is not sufficient to determine the local negative ion density since its signal results from the interaction of the laser beam along the whole path intersecting the plasma. Using the laser pulse combined with a Langmuir probe in electron saturation mode allows the detection of the photodetached electrons. Moving through the plasma column the LP and the laser beam allows measuring spatially resolved negative ion density profiles. We combine these two measurements to obtain absolute negative ion density radial profiles for different values of injected power and magnetic fields and compare with predictions from OES and CRM.

Finally, we will discuss the recent development and first data from a Two-Photon Absorption Laser Induced Fluorescence (TALIF) based on a picosecond pulsed laser at 205 nm to excite ground state atomic H. The excited states decay by emitting fluorescence at the H α line, which is detected by an ICCD camera allowing spatially resolved measurements of the density and temperature of atomic H. The width of the fluorescence emission line, mostly determined by Doppler broadening, is related to the neutral temperature. These measurements are of upmost importance to improve CRMs of H₂/D₂ plasmas.

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RF probes for plasma diagnostics and their modifications

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The work describes the use of planar RF probe working in the frequency range of 100 kHz – 1 MHz for time resolved measurements of plasma parameters during a reactive deposition process. In this case, the surface of the probe is very often coated with a dielectric layer and classical DC methods cannot be easily used. The RF planar probe [1] presented in this contribution is an extension and modification of the RF Sobolewski probe [2] and the floating harmonic probe [3]. The RF signal is applied on this planar probe, merged with reactive plasma and RF current and voltage waveforms are measured and digitally stored for further numerical processing. Obtained RF current and voltage waveforms detected are numerically processed and in this way plasma parameters like ion flux, electron temperature and ion density can be determined with time resolution. The key procedure to get the real current flowing through the probe is the calculation of capacitive current flowing through the probe and the voltage dependence of a parallel connection of parasitic probe capacitance and dynamical sheath capacitance. This capacitive current can be then numerically subtracted from the total RF current on the probe. The RF probe can easily handle the coating of the surface of the probe because it only works with RF signals and the RF voltage drop on this dielectric layer can be neglected if this layer is not too thick. The RF probe also provides the possibility to measure plasma parameters with time resolution, where the minimum time step is the period of the applied RF signal on the probe. The RF probe was used for plasma parameter measurements during the growth of semiconductor oxide thin films like CuFeO_x , WO_3 and Fe_2O_3 in reactive pulsed sputtering. The surface of the probe was coated with a semiconductor layer, which was utilized in the second configuration of this diagnostic technique. This version of the probe has the same hardware but a modified electrical unit supplying the probe. A small RF voltage signal was applied to this planar probe, with a variable frequency in the range of 1 Hz – 1 MHz. The impedance spectrum of the system plasma-semiconductor thin film was detected with a suitable electronic circuit.

The proper electrical model for this impedance spectrum was found and it was shown that the plasma space charge sheath at the surface of the probe can be

approximately modelled by the parallel connection of the capacitor and resistor. Besides this, the semiconductor film behaves as a parallel connection of capacitor and resistor. Analysis of these impedance spectra in complex Gauss planes resolve the impedance spectrum of space charge sheath and the impedance spectrum of deposited semiconductor films. The use of a plasma impedance

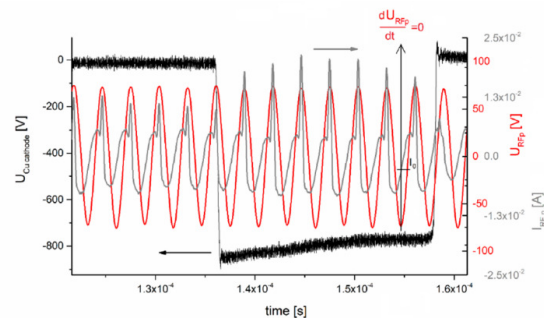


Fig 1 Measured RF current and voltage waveforms on the probe in pulsed reactive sputtering process.

spectroscopy method was demonstrated on the deposition process of selected oxide films like Fe_2O_3 , TiO_2 and WO_3 , where the complex dielectric constants (relative permittivity and conductivity) of these films were determined during the growth process. Accuracy, limits and possibilities to use this method in practical pulsed reactive deposition systems are further discussed.

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LIBS applied to agro-environmental and forensic solutions.

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Abstract

Laser Induced Breakdown Spectroscopy (LIBS) is an atomic emission technique that analyses the plasma generated by the incidence of a high intensity pulsed laser in the sample[1]. Overall, it is a relatively simple and easy handle technique, which allows performing a simultaneous multivariate analysis. The operational advantages are low relative cost per measurement, fast analysis, no need of labored sample treatment and the possibility for in situ and real time measurements [2,3]. One of the most limiting problems of the technique arrives from the matrix effect, compromising quantitative analysis. However, the huge amount of information contained in just one LIBS spectrum opens up several possibilities, like overturning the matrix effects in some cases [4,5] and the possibility of multivariate analysis associated with machine learning algorithms [6–9]. Due to the technological advances in electronics and optical components in the recent decades, LIBS has become more popular and nowadays it is present in many laboratories around the world, with a growing presence in developing countries. Specifically for Brazil, LIBS is quite attractive because of the possibilities of immediate application in some specific issues.

In this talk, it will be presented some applications of the LIBS technique developed in our research group in Brazil. Because Brazil and the state of Mato Grosso do Sul (MS) is an agricultural hub, with a large production of soybeans and corn, it will be presented how the "LIBS signature" can be used to identify seeds with greater germination capacity [9], contributing to increase the agricultural productivity. A study dealing with fertilizers and soils elemental quantification [4,5] and the identification of heavy metals in environmental samples [10,11] will also be addressed. For the last, it will be discussed the recent study that has been developing in partnership with the public security secretary and the department of civil police in Brazil with the aim to develop a protocol to detect gunshot residue (GSR) in hands and clothes in a rapid and accurate way using LIBS and machine learning algorithms.

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Optical properties of plasma surfaces in high-resolution spectroscopy of backscattered and sputtered atoms

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High-resolution spectroscopy is an important tool to provide insights into the physics of low-temperature plasmas at plasma surface interaction. It includes for instance measurements of the density of ion species, of the electric field or of the energy distribution of sputtered atoms [1]. We demonstrate in this work that the optical properties of plasma surface components have a significant impact on the spectra of backscattered or sputtered atoms, independently of the question if this effect is used for a further analysis or not. The experimental data were obtained using the linear plasma device PSI-2 described elsewhere [2].

1. Spectra of backscattered H and D atoms

We show that the spectra of backscattered D or H atoms in low density ArH or KrH gas discharges can be efficiently used for *in-situ* measurements of the optical properties of the surfaces. The excitation channel of H atoms due to atom-atom collisions with Ar or Kr atoms exceeds the conventional excitation by electron impact by 2-3 orders of magnitude in the same gas discharge. This admixture (Ar, Kr) significantly improves the signal-to-noise ratio of the obtained spectra. A negative potential in the order of about 100 V must be applied to the surface to distinguish the emission of backscattered H or D atoms from thermalized ones. The DSRM (Doppler-Shifted Reflectance Measurements) approach allows to measure the optical properties of the surface in the absence of additional calibration of the light source. The total reflectance is obtained directly from the ratio of reflected (red-shifted) and direct (blue-shifted) signal emitted by atoms separated due to the Doppler Effect [3] for different target materials (see Fig.1). Moreover, neither modelling nor simulations are required to obtain the value of reflectance at the angles of 0-45° with respect to the surface normal. For larger angles, both signals start to overlap significantly so that the modelling of emission is required. Nevertheless, the polarization properties of the plasma surface could be still extracted at the angles close to the pseudo Brewster one.

Finally, the quantitative monitoring of the cleaning or the removal of the substrate by Ar ion sputtering could be performed using the DSRM diagnostic. An

example of removing W from Ag substrate is presented here.

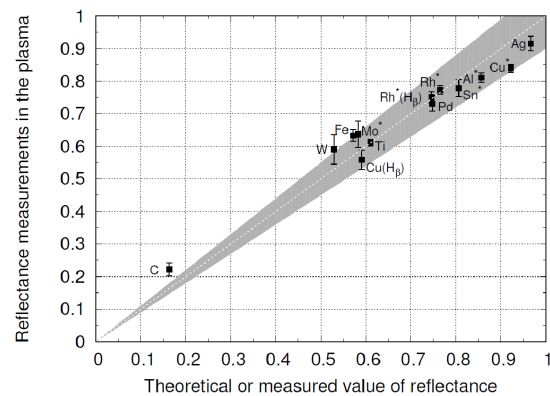


Fig. 1. Comparison between the DSRM in the plasma and the theoretical or measured values in the laboratory for different elements [4].

2. Spectra of sputtered Al and W atoms

Also, the spectra of sputtered atoms experience a strong impact of reflection in case of line-of-sights terminated at the target's surface. Moreover, the degradation of the optical properties due to sputtering leads to the dynamics in the line shapes of sputtered atoms. This effect is clearly demonstrated in the case of Al targets. The red-shifted part of emission reduces considerably during minutes of plasma operation as observed by the high-resolution instrument. Due to the low energy of sputtered atoms in comparison to the backscattered H atoms as well as the higher mass the separation between the red and the blue-shifted signal is hardly possible without Doppler-shifted modelling of emission. At the same time, in contrast to the spectra of backscattered H atoms, the spectra of sputtered atoms provide accurate and spatially resolved information on the magnetic field strength using the technique of *optical isolation* of spectral lines, similar to the one applied in laser physics. It is exemplified in the emission spectra of W atoms sputtered by Ar or Ne ions impact.

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Physics of atmospheric pressure dual RF-LF frequency DBD's

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Due to their large potential of applications, homogeneous DBD's were largely studied for frequencies in the range from kHz to hundreds of kHz (LF) and thus for radiofrequency (RF). More recently dual RF-LF DBD's were considered [1-3].

This presentation will be centered on 50 kHz - 5 MHz DBD's in a penning mixture of Ar/NH₃. From experimental and fluid model results, the physics of the different discharge regimes will be described and the role of Ar₂ formation and VUV photoemission will be shown.

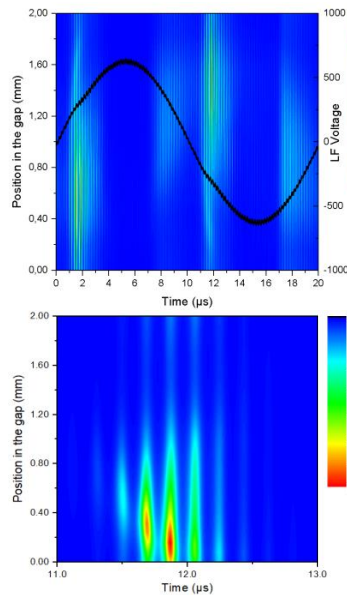


Fig 1: Emission of a RF-LF α - γ DBD for LF voltage (650 V and 50 kHz) and RF voltage (195 V and 5 MHz) a) emission of the continuum at 500 nm over the gap and the LF cycle b) emission of Ar 750 nm over the gap for 10 RF cycles, illustrating the γ mode.

The key point for experiments is to realize measurements on a LF cycle (ms) with a resolution at the RF scale (ns). For emission spectroscopy, it is reached with multichannel photons counter while a specific optical set-up allows a 100 μ m resolution of the 2 mm gap. This leads to results like those of Fig. 1 [3]. The discharge current and power as well as the density of Ar metastable are also measured.

Results obtained as a function of the amplitude of the LF [3] and the RF voltages [4] will be presented

showing that ohmic/ α or α - γ RF DBD regimes are obtained. The physics of the α - γ regime will be detailed on the basis of numerical model results [5]. At first [2], the fluid model only includes Ar, e, Ar⁺, Ar₂⁺ and Ar*. However, to well described the metastable density, Ar₂* and its VUV emission have to be added (Fig. 2) showing the importance of photoemission [6].

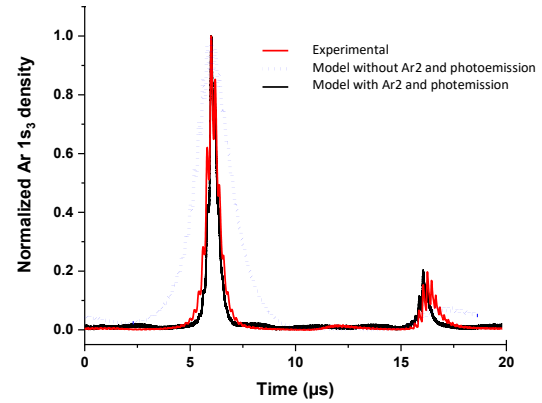


Fig 2: Comparison of the measured and calculated time variation of the Ar 1s₃ density over a LF cycle of a 5 MHz - 50 KHz DBD in the α - γ mode. Data are taken close to one of the electrodes which is the LF cathode during the first half cycle. Model results with and without Ar₂ and photoemission are compared.

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Electron impact excitation and dissociative excitation processes to plasma relevant molecules

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The electron induced fluorescence technique (EIF) is an experimental method that can be used to study the interactions of low energy electrons with the atoms and molecules. This technique can provide new insight into the collisional processes of electrons resulting in excitation of atoms and molecules, such as electronic excitation, dissociative excitation, and ionization associated with excitation.

In this contribution EIF technique developed at Comenius University in Bratislava will be presented [1,2,3] including the new experimental developments. In the EIF technique the beam of free electrons is colliding with the target particles in low pressure environment. The mean free path of the electrons substantially exceeds the dimensions of the experiment. Typically, pressure are in order $\sim 10^{-5}$ mbar.

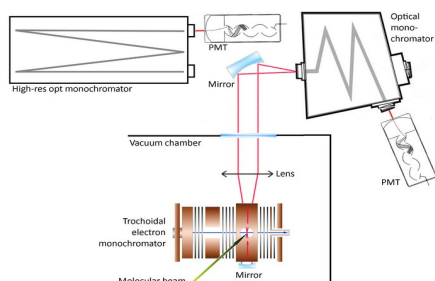


Fig. 1: Schematic view of the EIF set-up.

The EIF technique allows to study the emissions spectra of the atoms and molecules in the spectral range 180 – 1100 nm and in the electron energy range from 0 up to 2000 eV. Using this technique the electron energy resolved emission spectra of the atoms or molecules can be recorded in collision free environment. The spectra are not influenced by quenching reactions. Moreover, this technique allows to measure the cross sections for the electron impact excitation processes of the atoms and molecules as function of the electron energy.

The recent achievements in the field of EIF will be presented, including the electron molecule interactions to plasma relevant molecules such as H₂, O₂, N₂O and others.

Besides the data regarding electronic excitation and emission of the molecules, we are going to present data about dissociative excitation processes leading to cleavage of the excited atoms and functional groups.

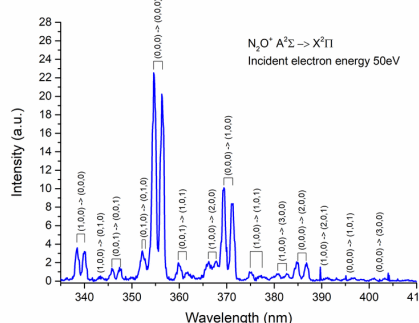


Fig. 1: Emission spectrum of nitrous oxide induced by 50 eV electron impact.

Special attention will be paid to the formation of continuum radiation arising from molecules initiated by dissociative excitation process and to the formation of the excited ionic species (molecular ions and ionic fragments) and their radiation.

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Advances in IEDF Measurements by Lockin-Detection

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1. Motivation

The distribution functions of the particles in a plasma provide the most complete description of its properties. Often the distribution of the electrons is targeted, but the distribution of the ions can also supply valuable information [1]. However, this requires the precise determination of the ion velocity distribution function (IVDF). Here we demonstrate a method to improve the dynamic range of the IVDF measurements with a commercially available retarding field energy analyzer (RFEA) in a combination with a lock-in amplifier technique. The method is applied to the investigation of the plasma pressure.

2. Experimental

The ion distribution function is related to the first derivative of the current-voltage characteristics of the RFEA [2]. The derivative is obtained through a modulation of the retarding voltage U_0 by a sinusoidal signal $\hat{U}$ and detecting the amplitude A_1 of the first harmonic by a lock-in amplifier (Fig. 1(a)). The choice of the amplitude of the modulating voltage is restricted by the requirement for negligible contribution of the higher order derivatives to A_1 and by the desired energy resolution through the transfer function. The contribution of the displacement current between the last two grids of the RFEA due to the oscillating voltage is compensated by mixing a signal $U_{\text{zero-line}}$ with a suitable amplitude and phase.

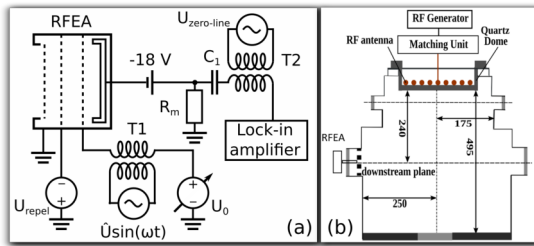


Fig. 1: (a) Realisation of the lock-in technique for the RFEA. (b) Discharge setup for the measurements.

The system is applied to an inductively coupled plasma source (Fig. 1(b)), operated in argon at pressures in the range of 1 Pa.

3. Results

The obtained ion distribution functions (inset of Fig. 2) show the typical single peak shape, observed also previously [1]. The acquisition through a lock-in

amplifier provides an increase in the dynamic range to about four orders of magnitude. This allows better estimation of the plasma potential Φ_0 on the axis of the discharge from the highest energy of the ions, while the energy $\varepsilon_{\text{peak}}$ of the peak allows an evaluation of the electron temperature near the sheath edge: $\varepsilon_{\text{peak}} \approx [0.8 + 0.5 \ln(M/2\pi m_e)]kT_e$, M and m_e – ion and electron mass, respectively. For the IVDF in Fig. 2 the values are $\Phi_0 \approx 24$ V and $T_e \approx 3$ eV versus $\Phi_0 = 22.6$ V and $T_e = 3.1$ eV from probe measurements. The large dynamic range of the IVDF allows also other plasma parameters to be determined [1, 2].

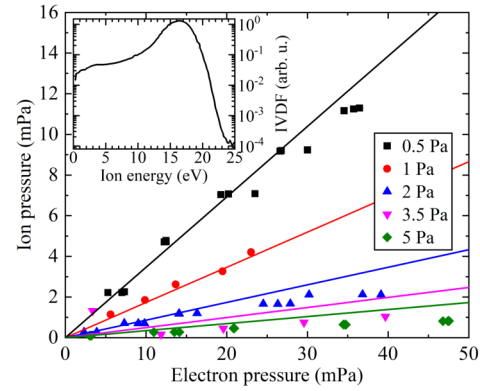


Fig. 2: Ion pressure at the wall versus electron pressure in the centre for different neutral gas pressures and input power levels. The lines are theoretical estimations. The inset shows an example IVDF at 0.5 Pa and 600 W.

From the IVDFs the ion pressure at the walls $p_i = \langle n M v^2 \rangle$ (n – wall ion density) is calculated and compared to the electron pressure on the axis $p_e = n_0 k T_e$, n_0 – density on the axis (Fig. 2). According to the theoretical expectations [3], the ion pressure at the wall is only a fraction of p_e . The lines in Fig. 2 give the theoretical dependence that scales inversely proportionally to the gas pressure. Excellent agreement between theory and experiment is observed.

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Topical lectures

Combining pulsed plasma and FTIR spectroscopy for detailed study of complex chemistry in CO₂-CH₄ plasma

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Dry Reformation Methane (DRM) assisted by plasma, which uses plasma discharges to turn CO₂ and CH₄ into value-added products such as synthetic gas has gain much interest over the last years, first because of its potential to play a key role in the energy crisis, second because of the possibility to use non-thermal characteristics to promote specific chemistries [1]. In particular, the radio-frequency discharges, which have demonstrated good conversion efficiency in pure CO₂ plasmas, could be of significant interest in this process. However, though CO₂ plasmas have been widely studied over the last decades [2], the chemistry of CO₂-CH₄ remains challenging to understand because of the many possible reactions taking place, leaving the gas-phase chemistry largely unknown. The goal of this work is to obtain a detailed description of the main reactions taking place in the gas phase of a CO₂-CH₄ RF plasma thanks to experimental data, which will also be used for the validation for a kinetic model.

However, obtaining such description of the reaction pathways requires the ability to perform time-resolved measurement of a large number of species at once. To this aim, we have developed a new approach combining IR absorption by FTIR spectroscopy with a pulsed RF discharge at low pressure (2 to 5 torr) in a closed reactor (without any gas flows). In this approach, a power supply allowing fast switching on/off of the RF voltage to perform plasma pulses of few ms (with a stabilisation time of the RF voltage within few tens of microseconds) is used to ignite a plasma in a pyrex reactor with trains of pulses (typically 10 pulses of 5ms with 10ms off between pulses), each train separated by approximately 2 seconds, as described in [3]. At the end of each train, after relaxation of the mixture, an FTIR measurement of the gas is taken. The densities of IR active species in the plasma are followed by fitting the IR spectras, thus giving an evolution of the composition as a function of the number of pulses (or of the plasma-on time) from start to steady-state. The typical evolution of a 50/50 CO₂-CH₄ mixture at 2 torr is given in figure 1, where we can see that CO and H₂ (measured indirectly) are the main products, while C₂H₆ seems to be an important intermediary product, disappearing under long plasma exposure.

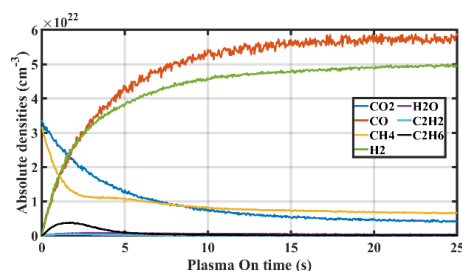


Fig. 1: Evolution of the composition of 1:1 CO₂-CH₄ mixture at 2torr

The parameters of the measurements such as the duty cycle of the pulses, the duration of the pulses or the total number of pulses are varied to single specific reactions selected by their characteristic time. These data will be compared with time-resolved measurements of the vibrational temperatures of CO₂ and CO measured by fitting of infrared spectras (as described in [4]) in a CO₂-CH₄ glow discharge in similar conditions of pressure and initial compositions, which bring information on the vibrational characteristic time and allow comparison.

The reaction pathways are also explored through the use of isotopical C¹³O₂ in the initial mixture. The C¹³O₂ specific IR absorption makes it traceable (as well as its by-product C¹³O) and distinguishable from C¹²O₂, which allows for unique probing of the reaction pathways, giving insights on the atomic exchange processes.

This original approach coupling a pulsed RF plasma in a closed reactor and FTIR measurements provide a unique set of parameters in a single plasma configuration to investigate CO₂-CH₄ plasma kinetics and in particular to develop kinetic models of complex mixtures coupling electron, vibrational and chemical kinetic processes.

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Absolute oxygen atom density in a DC discharge in pure O₂ :

1: Cavity Ringdown Spectroscopy

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1. Introduction

Accurate measurements of the absolute density of reactive atoms are essential for the validation of models of plasmas in molecular gases. However, even for such a ubiquitous species as oxygen atoms, the results of different measurement techniques are rarely compared. Other than optical emission spectroscopy (widely employed, but of questionable absolute accuracy), Two-photon absorption Laser-induced Fluorescence (TALIF), with the calibration scheme proposed by Niemi et al. [1], is widely used. However, high absolute accuracy is difficult to achieve due to the many complex steps in the calibration, the fluctuations inherent in a non-linear technique, and the fact that the O/Xe two-photon cross-section ratio has been measured only once.

Recently, Peverall et al. [2] measured the oxygen atom density in a low-pressure inductively-coupled plasmas by cavity ringdown spectroscopy of the forbidden $^3P_2 \rightarrow ^1D_2$ transition at 630nm. The absolute accuracy of absorption techniques depends only on the accuracy with which the line-strength is known ($\pm 7\%$ in this case [3]), provided that the density profile along the absorption path is either uniform or well-characterized. We have measured the oxygen atom density by CRDS in the uniform positive column of a DC glow discharge in pure oxygen, and will compare the results to TALIF in a separate paper.

2. Experimental and results

The DC discharge is sustained in a borosilicate glass tube (id 20mm, length 68cm) between cylindrical electrodes located in side tubes (separated by 53cm). High-reflectivity concave dielectric mirrors are located at the tube ends. One mirror is scanned by a piezo-actuator to achieve longitudinal mode matching. The first-order beam from an acousto-optic modulator is injected into the cavity, stopped when the optical intensity in the cavity passes a threshold value, and the ringdown time measured.

Since it is not possible to make CRDS and TALIF measurements at the same time, it was necessary to evaluate the reproducibility of the atom density in the DC tube using CRDS. The gas pressure (0.85 Torr) discharge current (40mA) and wall temperature (20°C) are all stabilized.

Measurements were made every three minutes on three consecutive days. The density when the discharge is first lit (after overnight vacuum) was always around $5.2 \pm 0.3 \times 10^{15} \text{cm}^{-3}$, dropping quickly to about $3.5 \times 10^{15} \text{cm}^{-3}$ in the first hour, then decreasing much more slowly. On the last day the discharge was allowed to run all night, and the density had stabilized at about $3.0 \times 10^{15} \text{cm}^{-3}$. Since all external parameters are stabilized, this effect can only be attributed to a (reversible) increase in the recombination probability on the borosilicate glass tube walls.

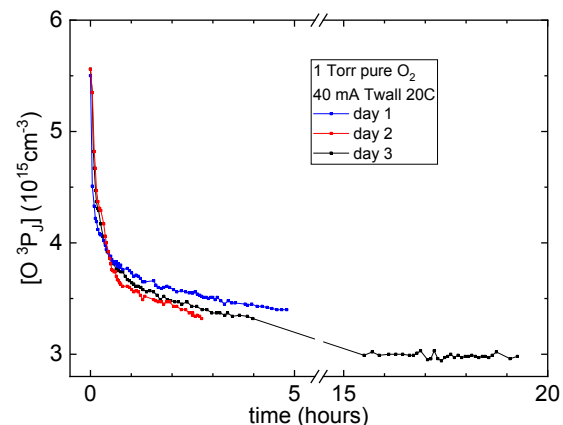


Figure 1. Oxygen atom density versus time after discharge initiation on three consecutive days.

The TALIF measurements (see abstract by Shu et al.) were taken 2-6 hours after discharge initiation. The CRDS measurements indicate a line-averaged O atom density of $3.3 \pm 0.2 \times 10^{15} \text{cm}^{-3}$ at 2 hours, decreasing by about $0.1 \times 10^{15} \text{cm}^{-3}$ per hour.

Acknowledgements

We thank Fédération de Recherche PLAS@PAR and China Scholarship Council for financial support.

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Investigation of microwave surfatron plasma source during plasma-assisted atomic layer deposition process

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Microwave surface wave sustained plasma source was used as a source of oxygen/nitrogen ions and radicals in plasma-assisted atomic layer deposition (ALD) system. The surface wave launcher—surfatron was modified to be compatible with a UHV chamber and equipped with the RF electrode ensuring fast and stable ignition of the surface wave discharge even in the pure molecular gas. The knowledge of plasma parameters and their spatial homogeneity is crucial for optimization of thin film deposition on large substrates. Therefore, Langmuir probe was employed to measure plasma density N_e , electron effective temperature T_{eff} , plasma V_{pl} and floating V_{fl} potential in the radial distance of 0–50 mm and in the axial distance of 27–57 mm from the surfatron nozzle outlet. Moreover, optical emission spectroscopy (OES) was employed to investigate emission lines nearby the surface wave launcher.

Four different gas mixtures were investigated at the constant pressure 6 Pa, namely (i) 50 sccm of Ar, (ii) 56 sccm of Ar and 3 sccm of N_2 , (iii) 49 sccm of Ar and 21 sccm of N_2 , and (iv) 54 sccm of Ar and 6 sccm of O_2 . An example of spatial distribution of the measured plasma density in the case (iv) can be seen in Fig. 1. Obtained results proved that N_e and the plasma potential are homogeneous in the radial direction and their magnitudes rapidly decrease with the axial distance from the surfatron nozzle outlet. On the contrary, T_{eff} is more or less constant in the studied spatial region with an average magnitude of ≈ 2 eV. The addition of nitrogen to the gas mixture led to a decrease in the studied plasma parameters in the axial direction with the exception of V_{pl} and V_{fl} , which increased. On the other hand, the radial homogeneity of the plasma parameters decreased again with the exception of V_{pl} . Adding 10% of oxygen into the total mass flow rate through the surfatron demonstrates a gradual decrease in N_e in the radial and axial directions with a magnitude roughly similar to the one measured in the pure argon plasma. On the other hand, T_{eff} attains in average of 2 eV in the investigated region with the maximum near the nozzle outlet.

Optical emission spectra revealed many Ar I lines of neutral atoms with only a few Ar II ions' lines. The gradual addition of nitrogen causes a systematic decrease in the Ar I line intensity. We expect that excited nitrogen molecules are produced by the

inelastic collisions with electrons and by the collisional quenching of metastable Ar(4s) states. Oxygen atom and ion lines are detected when oxygen was mixed with argon. Moreover, we observed plenty of emission lines coming from the oxygen atoms and ions. Then, we believe that the surfatron plasma is a source of significant fraction of active oxygen atoms and ions, which can play an important role in the plasma-assisted ALD process.

Since the ALD process runs at pressure circa 100 Pa and pure oxygen flows through the plasma nozzle using the Langmuir probe is complicated due to charged-neutral particles collisions in space charge sheath and permanent probe's surface contamination. Therefore, the Sobolewski probe [1] and OES were employed to investigate ion flux, tail electron temperature and optical emission spectra in pure oxygen surface wave sustained plasma *in-situ* during ALD process. Obtained results showed that the plasma parameters are strongly influenced by microwave power delivered into the plasma, which is the main parameter having impact on growth of thin film.

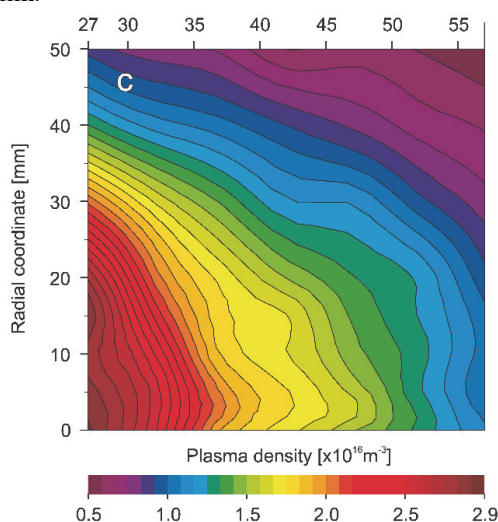


Fig. 1: The plasma density measured in Ar/ O_2 mixture.

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Vibrational excitation measurements by coherent anti-Stokes Raman scattering in a nitrogen ns-pulsed plasma jet

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1. Introduction

In this work the vibrational excitation dynamics of nitrogen molecules (electronic ground state) are investigated by coherent anti-Stokes Raman scattering (CARS). It was shown previously [1,2], that a homogeneous discharge can be ignited at (sub-)atmospheric pressure by applying high voltage pulses of several kV to plane-parallel electrodes with a duration of more than 100 ns. After a transient ignition phase, this kind of discharge exhibits a quasi-DC phase – similar to a traditional DC glow discharge. At 200 mbar in pure nitrogen, the plasma bulk during the quasi-DC phase makes up about 80% of the discharge volume. Furthermore, the electron density and the electric field in the bulk are essentially constant in time and in space. These constant conditions make the discharge a suitable tool for fundamental research of plasma processes.

The CARS measurements are supported by field measurements via E-FISH (electric field induced second harmonic generation) and a Particle-in-Cell simulation with Monte Carlo collisions (PIC/MCC).

2. Results

It was shown [2,3] that the discharge pulse results in a non-equilibrium distribution for the vibrational excitation. The repetitive pulsing of the discharge (1 kHz) results in an elevated vibrational temperature in the order of 1000-1500 K compared to the (rotational) gas temperature of approximately 330 K. Additionally, during each discharge pulse some of the molecules are vibrationally excited by electron collisions. The vibrational excitation occurs mostly in the plasma bulk. Therefore, the distribution and the excitation rate of these newly excited molecules depend mainly on the bulk electric field and the bulk electron density.

Common excitation rates [4] for the measured electric field result are in good agreement with the CARS measurements (see figure 1).

Additionally, plasma models were developed with insights provided by the PIC/MCC simulation. With those models the bulk field can be estimated for a given gas mixture. In combination with simple current measurements also the bulk electron density can be determined.

The easily accessible plasma parameters, which in addition are nearly constant in space and time, make the presented plasma source a promising device for benchmarking diagnostics and plasma chemical models.

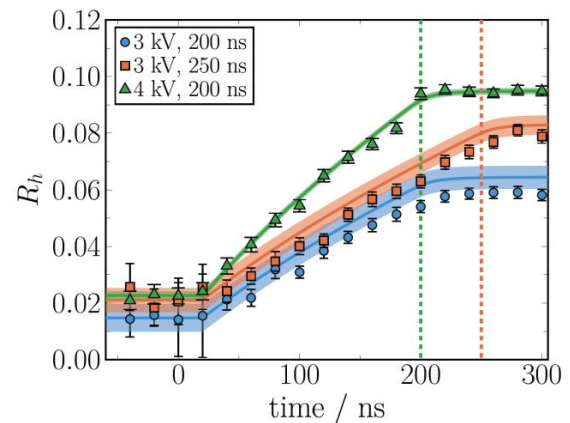


Fig. 1: Fraction of “vibrationally hot molecules”, which correspond roughly to the molecules excited during the current discharge pulse (exact definition given in [2,3]). Depending on the conditions, up to $\approx 10\%$ of the molecules are excited during a discharge pulse. The solid lines are calculated based on excitation rates for the measured field. [3]

Acknowledgements This work is supported by the DFG funded SFB1316 “Transient atmospheric plasmas – from plasmas to liquids to solids” and by Grant K134462 of the National Office for Research, Development and Innovation (Hungary).

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Breakdown and quasi-DC phase of a nanosecond discharge.

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1. Introduction

It was shown recently [1, 2] by picosecond Electric-Field Induced Second Harmonic generation (E-FISH) measurements in a nanosecond Atmospheric Pressure Plasma Jet (ns-APPJ) that the discharge consists of two distinct phases: fast breakdown at high electric fields and a quasi-DC phase at lower permanent electric field and high electron density. It was also demonstrated that the phases are separated and can be controlled independently in order to optimize the production of excited species [2, 3].

In this work the ns-APPJ operated in pure nitrogen and N₂:CO₂ mixtures is studied by spatially and temporally resolved Optical Emission Spectroscopy (OES) and Particle-in-Cell/Monte Carlo Collisions (PIC/MCC) simulations. To study the discharge structure and its development the emission of the First Negative System (FNS) and the Second Positive System (SPS) of nitrogen is considered and compared with the excited species densities which are inferred from the simulation results.

2. Results

An example of the measured emission intensity of the FNS (0-0) transition as a function of time and distance from the cathode is shown in Fig. 1. Waves of FNS emission corresponding to ionization waves traveling between the electrodes are well resolved.

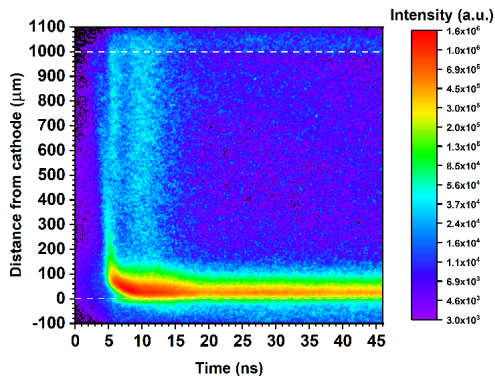


Fig. 1: Emission intensity of the FNS (0-0) transition measured as a function of time and distance from the cathode. Dashed lines denote the positions of the electrodes.

The results of the N₂⁺(B²Σ_u⁺, v = 0) density calculations are presented in Fig. 2. Similar dynamics

of the reconstructed density compared to the experimentally measured emission is clearly seen.

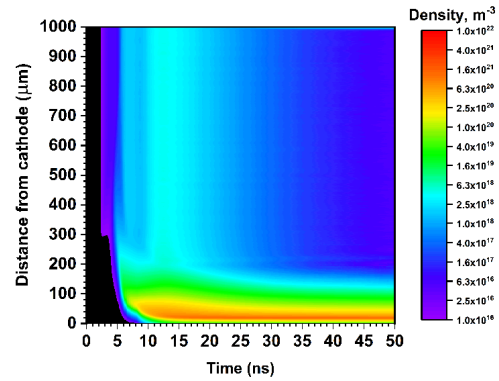


Fig. 2: Density of the N₂⁺(B²Σ_u⁺, v = 0) state obtained using results of the PIC/MCC simulations.

A good agreement between the experimentally measured emission dynamics and the excited species densities reconstructed from PIC/MCC simulations is demonstrated both for the breakdown and for the quasi-DC phase, both for the N₂⁺(B²Σ_u⁺, v = 0) and for the N₂(C³Π_u, v = 2) states. Such agreement serves as a validation of the PIC/MCC simulations in addition to comparison of the numerical results with electrical measurements.

Measurements in N₂:CO₂ mixtures agree well with the conclusions of an analytical model predicting weak dependence of the discharge structure on the gas composition.

Acknowledgements The work is supported by the DFG funded SFB1316 Project "Transient atmospheric plasmas - from plasmas to liquids to solids" and by Grant K134462 of the National Office for Research, Development and Innovation (Hungary).

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Time resolved CO₂ ro-vibrational excitation in a nanosecond discharge measured with laser absorption spectroscopy

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1. Introduction

CO₂ conversion is of growing interest in the context of greenhouse gas abatement and renewable energy exploration. The non-thermal plasma is a promising means for efficient conversion since the unique electron, vibrational, rotational, and gas temperatures in these plasmas allow focusing the discharge energy to the desired channels instead of heating the gas. Specifically, the vibrational excitation, starting with electron-impact-excitation of lower vibrational levels, followed by vibrational–vibrational (V–V) excitation close to the dissociation threshold level, is a more efficient dissociation pathway. This ladder-climbing population to higher vibrational levels occurs due to the small difference (~ 0.3 eV) between the higher vibrational levels and faster V–V relaxation rates compared to vibrational–translational (V–T) transfer.

Temporally resolved measurement of the ro-vibrational excitation is of great importance in non-thermal and transient discharges to gain insight into the excitation and relaxation processes and to validate detailed kinetic models of CO₂ dissociation.

2. Experiment

Measurements are performed in a nanosecond pulsed discharge ignited between two parallel molybdenum electrodes of 20 mm length, 1 mm wide and 1 mm apart. The discharge pressure is 150 mbar with CO₂ (10 %) in He (90 %). The voltage pulses ($V = 2$ kV, $f = 2$ kHz) are about 100 ns long leading to currents of 10 A.

A single-mode, continuous wave quantum cascade laser tunable between 2276 and 2290 cm⁻¹ is used as light source and a fast (65 MHz) detector for measuring the absorption with high temporal resolution. The chosen wavelength range allows for simultaneous measurement of rotational and vibrational temperature for the symmetric as well as the asymmetric modes. High temporal resolution of down to 8 ns is achieved by a special strategy for the laser wavelength tuning and the subsequent data acquisition. In order to obtain the multiple temperatures (T_{rot} , T_{v1} , T_{v2} and T_{v3}) and the concentration of CO₂, spectra as a function of the temperature, calculated beforehand, are fitted to the measured ones [1].

3. Results

A typical absorbance spectrum obtained for a discharge with a voltage of 2.25 kV and a pulse length of 150 ns measured with 16 ns resolution is shown in figure 1 [2]. By fitting calculated absorption spectra to the measured ones, the evolution of the rotational and vibrational temperatures of CO₂ within the nanosecond discharge pulse are obtained temporally resolved (figure 2).

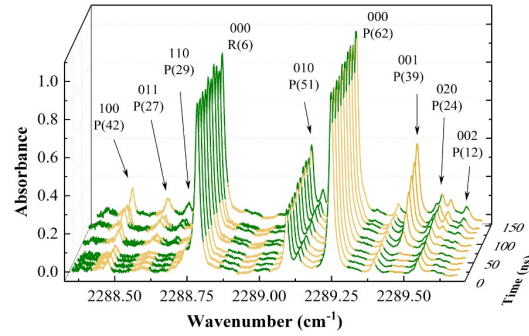


Fig. 1: Temporal development of the absorbance.

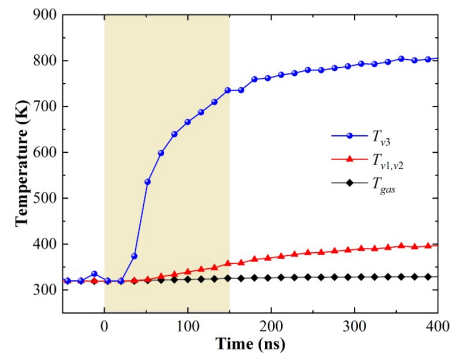


Fig. 2: Temporal evolution of the gas and vibrational temperatures. The duration of the discharge pulse is marked by the shaded area.

Acknowledgment

This work is supported by the DFG (SFB1316) and Y. Du also acknowledges the financial support from the Alexander von Humboldt Foundation.

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Optical measurement of the microparticle charge in plasmas using quantum dots: Statistical model

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Electrical charge is a very important parameter of the dust particles immersed in plasmas. It enters into practically all dust-related quantities. Many phenomena in dusty plasmas are supposed to be the consequence of, e.g. charge fluctuations, spatial charge gradients or delayed charging. In laboratory experiments, the charge is usually measured by means of dynamical methods. Such methods have obvious disadvantages: They require certain assumptions on the forces acting on the particles, their interaction potential or plasma parameters. Also, the temporal resolution of such measurements is limited by the slow dynamics of dust. It is, therefore, very important to develop optical methods of the charge measurements.

There are two ways to address this problem: (i) Use the naturally available charge-dependent spectral features of light scattering by the dust particles (see e.g. [1]) or (ii) design the surface of a dust particle in such a way that it acquires optical sensitivity to the charge [2]. In the former case, the spectral features usually lie in the infrared region. It is, however, very advantageous to perform the optical measurements of dust charge in the visible spectral range since this would allow to measure the charges of the dust particles individually.

Therefore, we propose to use the quantum dots (QDs) deposited on the surface of the microparticles as optical charge sensors [3]. Radiative transitions of the QDs are subject to the so-called quantum-confined Stark effect [4], for which the spectral shift of the photoluminescence wavelength is proportional to the square of the local electric field. Realization of such measurement will allow to use the QD-coated microparticles as floating optical probes which can be manipulated in plasmas by means of optical tweezers [5]. Experiments with quantum dots deposited on large flat plasma-facing surfaces proved the sensitivity of the QDs to the surface charge [6].

The average distance between the electrons sitting on the surface of a microparticle is much larger than the size of a QD. Therefore, the electric field sensed by a QD will be subject to large fluctuations. The radiation lifetime of an excited state of a QD appears to be sufficiently longer compared to the lifetime of a quasi-steady-state electron configuration on the surface of a microparticle. The Stark shift registered over long exposure time will consequently be

equal to the average over all the quasi-steady-state electron configurations. This value is of the order of a fractions of nm and can therefore be easily measured (see Figure 1).

Other issues related to the possible experimental realization of such measurement (heating of microparticles and QDs, sputtering of QDs, surface design) will be discussed.

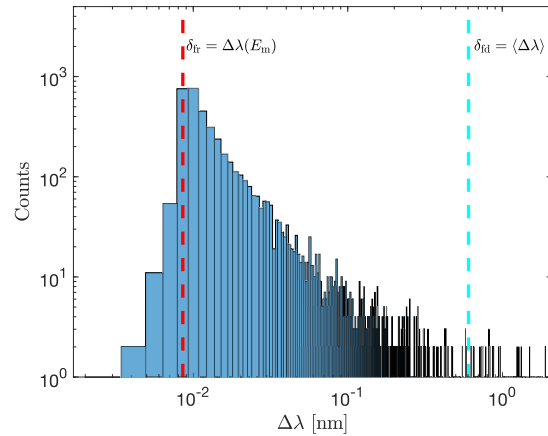


Fig. 1: Statistics of the Stark shift for a CdSe quantum dot of 3.3 nm radius on the surface of a microparticle of 4.6 μm radius and charge of 3×10^4 elementary charges [7].

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Probing N_2^+ ions in an ICP sheath by cavity ringdown spectroscopy

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Inductively coupled plasma (ICP) finds extensive use in surface processing, from plasma enhanced chemical vapour deposition to reactive ion etching[1], and these important applications make it highly desirable to understand the fundamental chemistry and physics of these systems. In particular, plasma etching is controlled by the behavior of ions in the region where the plasma interacts with chamber surfaces (the plasma sheath) and so being able to probe this region selectively is highly desirable.

The sheath is characterized by the loss of quasi-neutrality that exists in the bulk plasma. Highly mobile electrons negatively charge the chamber walls leaving a net positive sheath in which ions are accelerated towards the negative surface. This acceleration is responsible for plasma etching and leads to a depletion in ion density close to the chamber walls[2].

Ion densities in ICP sheaths are typically of the order of 10^8 - 10^9 cm^{-3} . Cavity Ringdown Spectroscopy (CRDS) is a species selective and quantitative technique that is capable of achieving the levels of sensitivity required to detect such low densities (minimum detectable absorption coefficient, $\alpha_{\text{min}} \sim 10^{-10} \text{ cm}^{-1}$)[3, 4].

We present measurements of N_2^+ ($v=0$) in an N_2 ICP by use of CRDS on the $N_2^+(A^2\Pi_u, v=2) \leftarrow N_2^+(X^2\Sigma_g^+, v=0)$ transition. This technique allows measurements, not only of ion densities, but also ion temperatures in the plasma. By use of a height adjustable lower electrode we are able to probe ion densities as a function of position within the plasma sheath adjacent to the electrode. Notably, the spectra also show effects of optical saturation which must be accounted for when extracting number densities.

We find that both ion densities and translational temperatures in the plasma bulk increase with increasing plasma power and also with decreasing plasma pressure from $\sim 10^9 \text{ cm}^{-3}$ and $\sim 600 \text{ K}$ at 100 mTorr and 200 W up to $\sim 10^{10} \text{ cm}^{-3}$ and $\sim 1400 \text{ K}$ at 10 mTorr and 400 W.

Our sheath measurements, some of which are shown in figures 1 and 2 indicate a significant change in ion density (by a factor of ~ 4) within $\sim 1 \text{ cm}$ of the chamber wall (at 100 mTorr 300 W).

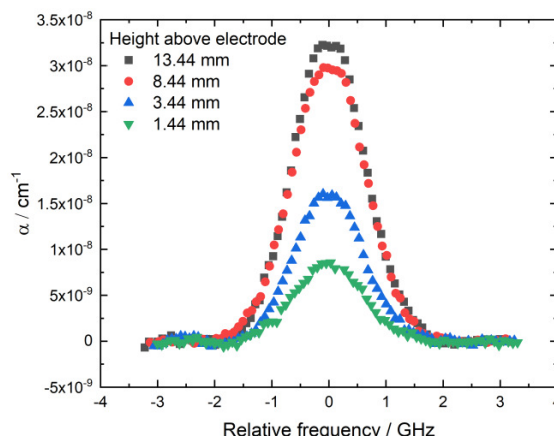


Fig. 1: CRDS spectra of N_2^+ as a function of the height of the probe laser beam above the lower electrode surface. Plasma conditions: 100 mTorr and 300 W.

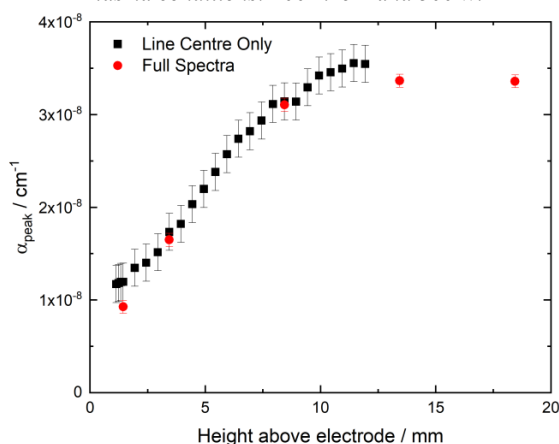


Fig. 2: Peak absorption coefficient as a function of beam height above lower electrode showing a marked depletion in the plasma sheath. The red points are taken from the data in fig. 1 whilst the black points come from a simple on/off resonance measurement.

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Measurement of atom density in Ar*(1s) metastable and resonance states by BBAS and of electron density and temperature from continuum emission in atmospheric pressure argon RF discharge

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1. Introduction

Recently, densities of He*(2³S) and He₂*(a³Σ_u⁺) metastables species have been measured in a parallel plate (20x20 mm²) RF-driven capacitively coupled glow discharge at atmospheric pressure helium by using broadband absorption spectroscopy (BBAS) [1]. The same technique and experimental device are employed to determine densities of Ar atoms in the metastables (1s₅ and 1s₃) and resonance (1s₄ and 1s₂) states in pure argon and He-Ar plasmas, produced with about 14 W of 13.56 MHz RF power [2].

Also, the electron temperature and density in Ar and He plasmas are deduced by fitting the measured absolute intensity of the continuum emission with the neutral bremsstrahlung radiation model [2].

2. Results

In Fig. 1 is shown the spatial distribution of atom densities within the 2.0 mm gap of electrodes for the four Ar*(1s) states in He+17% Ar plasma. Similar density variation is also obtained in pure argon. The density profiles correlate well with the sheath structure of the RF plasma operating in the α-mode. The larger density of Ar*(1s₂) atoms in the middle of the gap, relative to Ar*(1s₃) and Ar*(1s₅) densities, is attributed to the transport of population from the excitation zone in the sheath edges to the discharge middle via 104.7 nm resonance radiation trapping.

Absolute emission intensity calibration with a tungsten halogen lamp permits to estimate the

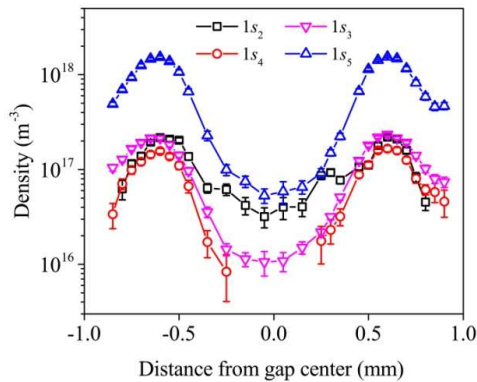


Fig. 1: Ar*(1s) densities in He+17% Ar plasma

density and temperature of electrons from the recorded continuum radiation emission of plasmas, shown in Fig.2. Fitted with the synthetic spectra built for e-atom bremsstrahlung radiation with a non-Maxwellian EEDF [3], the estimated values are $T_e \approx 2.1$ eV and $n_e \approx 1.1 \cdot 10^{19} \text{ m}^{-3}$ in pure Ar plasma and $T_e \approx 3.5$ eV and $n_e \approx 1.2 \cdot 10^{17} \text{ m}^{-3}$ in He plasma.

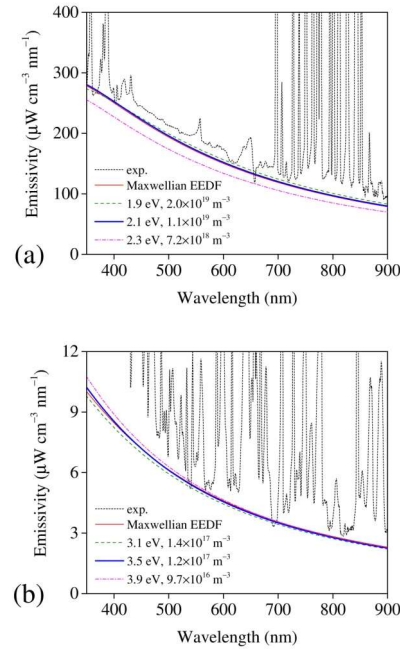


Fig. 2: Absolute intensity of continuum emission spectra (a) in pure Argon and (b) in pure Helium plasmas

Acknowledgement

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A new broadband high-resolution spectrometer based on a mid-infrared frequency comb for plasma diagnostic

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The development of frequency combs as broadband light sources has improved the performance of traditional broadband detection schemes. The two widely used broadband detection schemes for spectroscopy have been Fourier transform spectrometers (FTS) and grating-based spectrometers. Grating-based methods are usually faster, and more compact than FTS, but provide poorer spectral resolution. However, the combination of a grating with another dispersive element, such as a virtually imaged phased array (VIPA), drastically increased the resolution. Such combination is commonly called a VIPA spectrometer [1-3].

Here, we report on a high-resolution comb-based VIPA spectrometer in the mid-infrared (mid-IR) as a new broadband detection technique for plasma diagnostic. The mid-IR comb has a repetition rate, f_{rep} of 250 MHz, a bandwidth of ~ 400 nm, and a centre wavelength of 3200 nm. The VIPA detection system, see **Fig.1(a)**, consists mainly of: (i) a cylindrical lens to line focus the laser light, (ii) a VIPA etalon to vertically disperse the laser frequencies, (iii) a grating to further cross disperse the frequencies that are not dispersed by the VIPA horizontally, and (iv) a detector array to collect the cross-dispersed 2D frames. We used an air-spaced VIPA etalon with free spectral range of 4 GHz and effective resolution of ~ 80 MHz. **Fig.1(b)** presents an example of the measured frames. As the resolution of the VIPA is three times better than the f_{rep} of comb, the modes are clearly resolved vertically.

The capability of the detection system to measure high-resolution spectra have been tested in a plasma nitrocarburizing test reactor. **Fig.1(b)** shows the measured 2D images in the test reactor under low pressure of 1.5 mbar and flow of 20 sccm N_2 + 20 sccm H_2 + 1 sccm CH_4 . The plasma power was 68.1 W ($V_D = 364.2$ V, $I = 0.187$ A). The intensities of the pixels of the 2D images were sampled and the corresponding absorption spectra are obtained as a function of wavenumber. **Fig.1(c)** shows the constructed absorption spectra under plasma being off and on conditions. The final spectra were obtained by interleaving four spectra at different f_{rep} separated by 62 MHz in the optical domain. The resolved spectral lines are part of the congested Q-branch of the ν_4 band of methane, indicating the capability of the new VIPA spectrometer for high-resolution

spectral measurement. It can also be seen in **Fig.1(c)** that the absorption intensities of CH_4 lines reduced by a factor of ~ 2.3 as a result of its consumption in the plasma.

Sensitivity analysis, based on the estimated signal to noise ratio of 175, indicates a minimum absorption coefficient (for 1σ) of $5.7 \times 10^{-5} \text{ cm}^{-1}$ and a minimum detectable density for methane at room temperature of $5.2 \times 10^{12} \text{ molecule cm}^{-3}$.

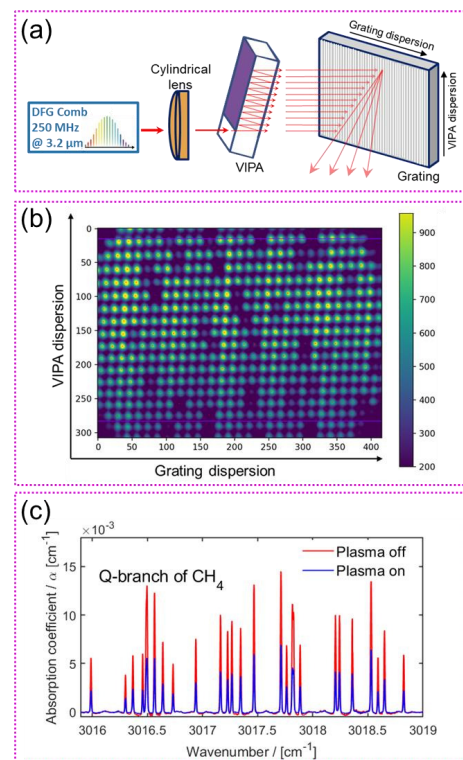


Fig. 1: (a) Schematic of the VIPA technique. (b) Example of a 2D image (1000 averages) measured by the IR detector array with a rate of 20 Hz. (c) the constructed high resolution absorption spectrum under plasma being off (red) and on (blue) scenarios.

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Insights into nanosecond repetitively pulsed CO₂ discharges by time-resolved optical emission spectroscopy

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Nanosecond repetitively pulsed discharges are seen as a promising way to achieve plasma (-catalytic) conversion of CO₂ into value added compounds in a commercially interesting, industrially viable manner; the non-equilibrium conditions unlock thermodynamically unfavourable reaction pathways. It has been demonstrated that a further enhancement of conversion is possible by utilizing a burst pulse pattern rather than a continuous one, under the hypothesis that this further augments the non-equilibrium features of the plasma and provides suitable conditions for vibrational ladder climbing to occur [1]. The short duration nature of these discharges—as well as the fast equilibration times at atmospheric pressure—necessitate fast diagnostics that are capable of probing the time evolution of discharge conditions in a non-invasive manner.

While running in low frequency continuous- or burst-mode, each pulse in the discharge is uncorrelated to the preceding one, i.e. it does not encounter a memory effect and will form its own streamer. Decreasing the pulse separation however means that a pulse will encounter a progressively stronger memory effect from the preceding pulse, altering discharge conditions and even traversing the electrode gap along the same discharge channel [1, 2]. As such, the electrical characteristics of the discharge as well as the emission produced by it change considerably. Time-resolved spectroscopic analysis of specific spectral lines has allowed both the study of transient conditions in the discharge as well as identifying underlying mechanisms or reaction pathways for the emission.

The Stark broadening of the long lasting oxygen $3s\ ^5S_0 \leftarrow 3p\ ^5P^{J=1,2,3}$ triplet allows for an estimate of the electron density n_e along the evolution of the burst discharge pattern, with a maximum corresponding with 5 % ionization, when assuming an electron temperature of 2.0 eV. The ratio of highly excited C⁺ and C⁺⁺ lines that are present confirm this assumption, yielding a rough T_e estimate of 2.0 eV to 2.5 eV.

The addition of a small fraction of N₂ allows the gas temperature T_g to be determined as well when

there is electron impact excitation occurring in the discharge, employing the (0, 0) band of the N₂ second positive system. Other common spectroscopic markers such as the CN Violet system or the C₂ Swan bands have to be discarded as there are chemical pathways that form a rotationally hot population and will thus not represent T_g due to insufficient rotational energy transfer to thermalize the state. A peak temperature of around 2500 K was found, which compares well with earlier results based on LIF thermometry and modelling [3, 4].

The overall picture that arises from these observations is of a highly ionised spark discharge in quasi-LTE, as opposed to the thermal spark encountered in NPG discharges in air-like mixtures [5]. Based on time-resolved spectra, it appears that CO₂ dissociation and O₂ formation are both processes that occur over longer ($\geq 1\ \mu\text{s}$) timescales and only occur after a delay, implying that other processes than electron impact play an important role.

Acknowledgements

This project has received funding from the European Union's Horizon 2020 research and innovation programme under the Marie Skłodowska-Curie grant agreement No. 813393. M.C. acknowledges support by the Progetto Strategico di Ateneo ERICSOL of the University of Trento.

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Diagnostics with an optically trapped microparticle in the sheath of an asymmetric CCP

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1. Motivation

Applications of low-temperature plasmas range from etching processes and coatings of solids to plasma medicine and basic research. Important plasma parameters such as density, temperature or composition of the species are diagnosed using many established methods [1]. However, it is difficult to probe the extremely important sheath region, which is only a few millimeters thick and, thus, not accessible with macroscopic probe methods, as they themselves change the plasma.

In recent years, therefore, microparticles have been qualified as probes for so-called non-conventional plasma diagnostic purposes. Due to their size and their behavior in the plasma, they, in particular, are well suited for increasing the spatial resolution and, thus, providing information in addition to common diagnostics [2].

2. Experiment

In this study SiO₂ microparticles are in an optical trap to manipulate them in the environment of a capacitively coupled asymmetric radio frequency discharge. In contrast to common plasma diagnostic tools (e.g. Langmuir probes, calorimetric probes, mass spectrometers etc.), in the μ PLASMA (microparticles in a discharge with laser assisted manipulation) experiment particles can be regarded as noninvasive single probes [3]. The displacement of the particle in the laser trap is observed to measure a force while it is moving relatively to the plasma, either deeper into the sheath or into the plasma bulk.

3. Measurements

Force profiles at different pressures and rf-powers have been performed in the sheath of an asymmetric capacitively coupled plasma. The force is mainly determined by the particle charge and the electric field in the sheath region. Thus, the measured force

while moving a single particle from the bulk plasma towards the electrode surface show a characteristic profile with a maximum and a decrease close to the electrode (Fig. 1).

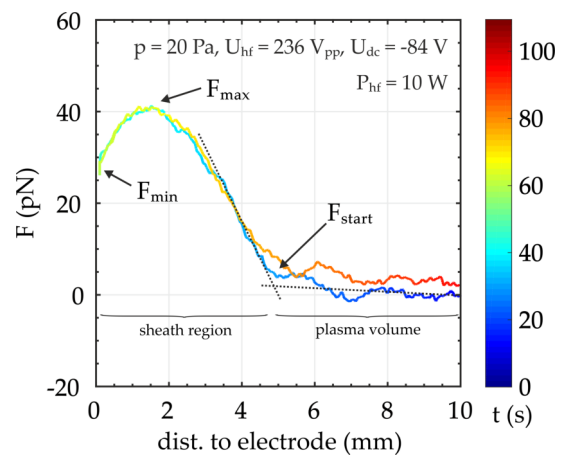


Fig. 1: Measured force profile while moving a single particle in the sheath of a CCP from the bulk plasma towards the electrode surface.

Furthermore, the benefit of the presented technique is the possibility to retain the particle even after the plasma is turned off providing the possibility to perform additional studies, e.g. on the residual particle charge.

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Time&space-resolved ICCD spectrometry and imaging help to uncover the nature of nanosecond discharges in water

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1. Introduction

Clarifying or confirming the driving mechanisms responsible for the formation of nonequilibrium discharges in liquid water and the corresponding plasma parameters is an extremely challenging task. Emission spectroscopy, although essentially a simple non-invasive method, appears to be an essential tool for revealing the basic ionization and energy transfer processes mediated by energetic electrons. In this study, we investigate spatial/spectral distribution of vis-NIR photon sources produced under single nanosecond discharge pulse conditions in DI water.

2. Experiment

We carried out the present experiments using a discharge reactor described in detail in recent works [1-3]. The electrode system is made of the tungsten anode pin and grounded stainless-steel chamber walls. Microdischarges were produced in pre-degassed DI water ($\sim 1 \mu\text{S/cm}$) using short positive HV pulses ($\sim 7 \text{ ns}$ full width at half maximum). Four Channel ICCD imaging system (Stanford Computer Optics, XXRapidFrame) and imaging NIR ICCD spectrometer (Andor, Shamrock 303i) were used to acquire microscopic images and space-resolved spectra with temporal resolution ranging from 500ps to tens of nanoseconds (few micrometers in space). Figure 1 illustrates an approach used to acquire the NIR ICCD spectra from the region of interest and resolved along the vertical (anode) axis.

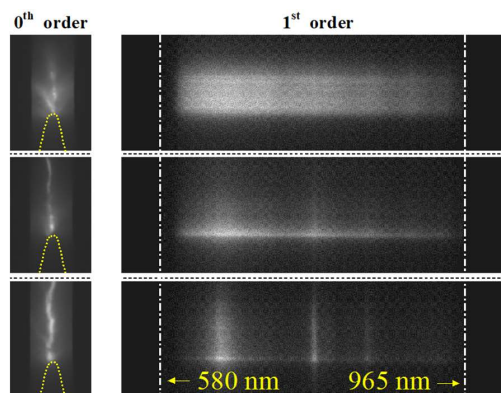


Fig. 1: ICCD images in the 0th order (left) with spectrally-resolved images in the 1st diffraction order (right) acquired during three specific discharge phases

3. Results

Figure 2 illustrates the first NIR emission spectra resolved along the vertical (anode) axis and acquired during two specific phases of a single discharge pulse. These current results make it possible to distinguish the main spectrometric signatures originating from both the anode-liquid interface and the bulk liquid, as well as the features from the initial and subsequent discharge phases with very high (sub)ns resolution.

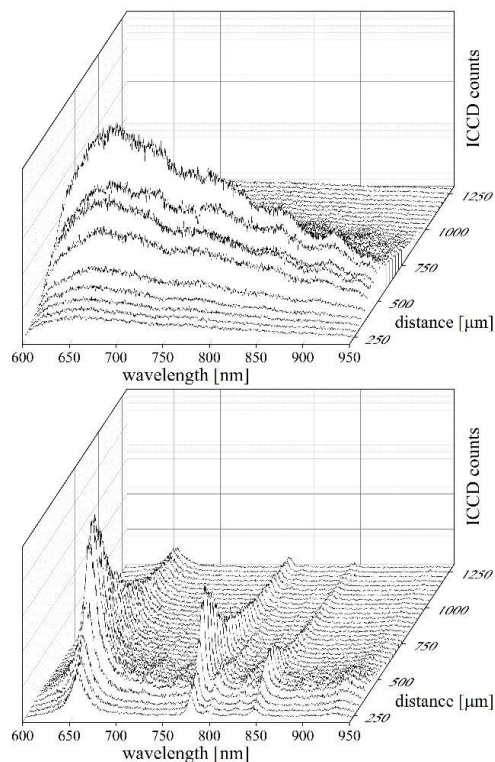


Fig. 2: ICCD spectra resolved along the vertical axis and acquired during two specific discharge phases

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Clarification of the production mechanism of ammonia in a hydrogen plasma with parts per million nitrogen

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1. Introduction

The admixture of nitrogen in hydrogen plasmas often plays a technologically important role, for example in plasma nitriding processes, in impurity seeding for power dissipation in fusion reactors and in thin film deposition processes. In this contribution, the significant role of impurity seeded nitrogen (ppm range) on a pure hydrogen plasma at low pressure is presented. Absolute ground state atomic hydrogen densities were measured utilizing two-photon absorption laser induced fluorescence (TALIF) in a low-pressure electron cyclotron resonance plasma for microwave powers from 100 W to 150 W. For the experiments conducted pressures of 3, 5, and 7 Pa were considered. Absolute ground state densities were derived using established calibration routines with krypton [1].

2. Results and Discussion

At N₂ admixtures of 1500 ppm and higher, two distinct spectral distributions of the fluorescence could be observed manifesting a dominant second production channel of atomic hydrogen. Amorim et al showed that this is due to a separate, nascent contribution arising from the photolysis of ammonia molecules [2].

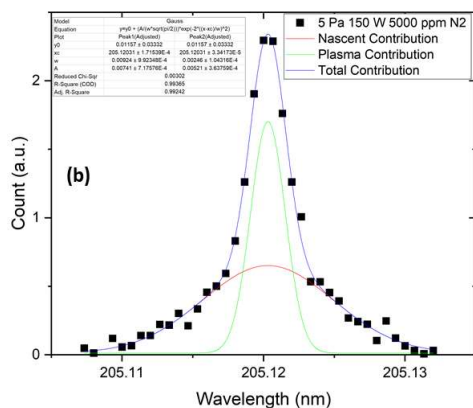


Fig. 1: The spectral distribution of the H_α fluorescence at 656 nm for a pressure of 5 Pa, an applied power of 150 W, and an admixture of 5000 ppm N₂.

At N₂ admixtures of 5000 ppm, the nascent contribution becomes the dominant contribution in all investigated pressures. The formation of NH₃ in H₂-N₂ plasmas is not quantitatively understood; however, it is widely accepted that production is dominated through plasma-surface interactions. To clarify the production mechanism of NH₃, i. e., the type of recombination reactions at the surface, thermal loading experiments were conducted.

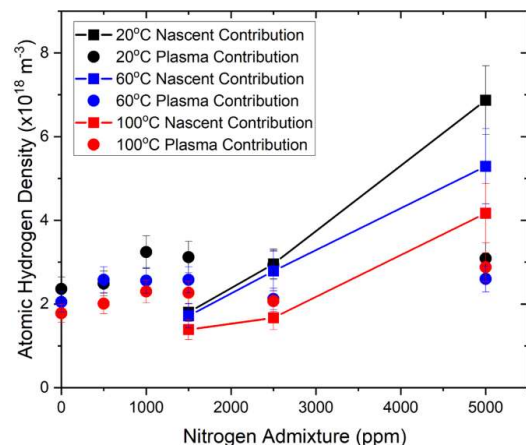


Fig. 1: The plasma and nascent contributions as a function of nitrogen admixture for three different wall temperatures.

From the plasma and nascent contributions of the atomic hydrogen densities as a function of N₂ impurities for different wall temperatures, it can be inferred that the Langmuir-Hinshelwood recombination mechanism is dominant over the Eley-Rideal mechanism [3].

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Terahertz Spectroscopy for Measurements of Electron and Atomic Oxygen Densities

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1. Introduction

Terahertz (THz) spectroscopy is a powerful diagnostic that provides information on numerous plasma parameters. Firstly, the THz spectral region between 0.1 and 10 THz contains energies related to molecular and atomic transitions involving rotational mode changes and fine structure splitting, respectively [1]. THz absorption spectroscopy therefore offers direct access to the corresponding densities. Secondly, THz frequencies lie well above typical values for the plasma frequency. This allows the electron density to be determined from a phase shift of transmitted THz radiation [2]. Knowledge about electron densities as well as atomic and molecular densities in plasmas is crucial for understanding plasma chemical phenomena. Two promising diagnostic techniques for measuring these densities are THz time-domain spectroscopy (THz-TDS) and THz quantum-cascade laser (QCL) absorption spectroscopy (THz-QCLAS).

2. THz Time-Domain Spectroscopy

The principle of THz-TDS is based on the ultrafast generation and detection of broadband THz pulses [3]. These pulses are sampled in the time-domain and subsequently Fourier transformed to obtain spectral information. Measurements therefore provide both amplitude and phase information, rather than only the intensity, meaning that electron densities can be investigated whilst simultaneously yielding information on atomic and molecular densities in the plasma. An example of the (power) spectrum acquired in atmospheric air is shown in figure 1.

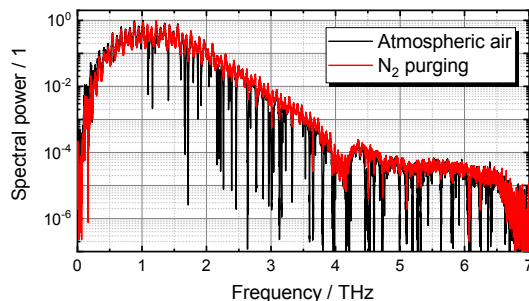


Fig. 1: An example of a THz spectrum acquired in atmospheric air, containing water absorption lines, compared to a spectrum acquired under N_2 purging.

THz-TDS is a non-invasive and pressure-independent technique, as opposed to conventional probe-based methods like Langmuir probes, and therefore especially promising for electron density measurements at atmospheric pressure.

3. THz QCL Absorption Spectroscopy

The spectral resolution of a THz time-domain spectrometer is limited to approximately 1 GHz. THz QCLs operating in continuous-wave mode are therefore better suited for the detection of sharp absorption lines. Although these lasers have a relatively small tuning range, their narrow linewidth (below 10 MHz) makes them an excellent THz source for high-resolution absorption spectroscopy [1, 4]. A QCL operating at 4.75 THz has been used to measure the absolute density of ground state oxygen atoms in a low-pressure radio-frequency plasma. An example of the measured absorption line is given in figure 2.

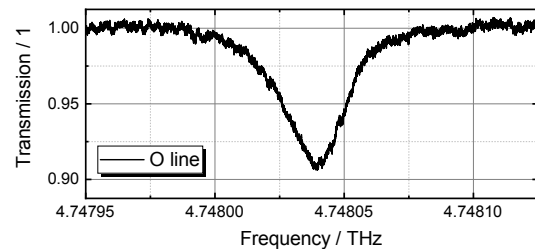


Fig. 2: An example of the absorption line of atomic oxygen measured in an O_2 plasma at a radio frequency of 13.56 MHz, a power of 60 W, and a pressure of 1 mbar.

In this contribution, the possibilities and limitations of both diagnostics will be extensively discussed, and results will be presented on electron and atomic oxygen densities measured with THz-TDS and THz-QCLAS, respectively.

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Investigation of multi-periodic self-trigger plasma in a AC-driven Atmospheric Pressure Plasma Jet

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Abstract

Atmospheric Pressure Plasma Jets (APPJ) have been intensively studied due to their potential application in biological fields. APPJs driven by low frequency AC voltage are known to produce randomly ignited ionization waves [1], often called plasma bullets, that travels over long distances; only few works have reported the propagation of bullets showing a periodic or multi-periodic stability [2].

In the present report, a helium plasma jet driven by 15~18 kHz sinusoidal voltage ignites multi-periodic self-triggered mode or random mode depending on applied voltage, driven frequency and inter-electrode gap distance. Most of the observed multi-periodic bullets operate every 2 or 3 sinusoidal period. An example of discharge current waveform of such kind of multi-periodic plasma is shown in figure 1. Each 3 cycle, a negative bullet is ignited, followed by a positive bullet, which propagates over 11cm. The presence of an outer grounded electrode ring is a key parameter permitting the ignition of multi-periodic bullets; it also enhances the propagation length up to 8 times. Fast imaging reveals that 2~3 self-triggered discharges occur in the gap region prior ignition of the bullet in both positive or negative polarities; this leads to an accumulation of charges beneath the ground electrode, locally enhancing the electric field. Bullet velocities for different polarities and gap distance are compared using optical emission spectrum. Such self-triggered bullets travel typically at a velocity of 25 km/s.

Such multi-periodic plasma jet is used to treat water target under different conditions including helium flow rate, voltage and jet-to-water distance. The production rate of H_2O_2 and NO_2^- in water is measured using colorimetric methods. The results show the H_2O_2 is mainly produced inside the dielectric capillary due to the small impurities, while the NO_2^- is produced in the plasma plume due to mixing with ambient air.

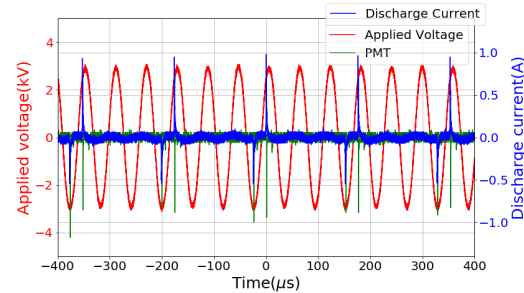


Fig. 1: The discharge current, and light signal measured by photon multiplier tube(PMT) plotted with the applied voltage. The applied voltage amplitude is 3.1 kV. The plasma bullet is ignited every 3 AC cycles.

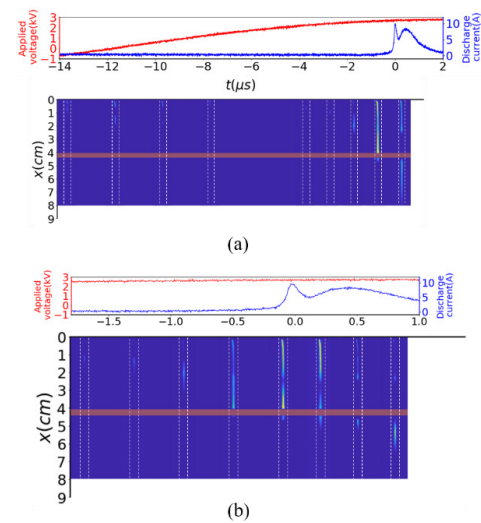


Fig. 2: Waveform and ICCD photos of the propagation dynamics of the positive bullet in different time scales. (a) shows the propagation in a time range of 14 μs with exposure time of 1 μs for each photo; (b) shows the propagation in a time range of 2.5 μs with exposure time of 200 ns for each photo. Note that the horizontal time axis applies for both the waveform and the ICCD photos.

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First Poster session

First Poster session	
Presenter	Stand
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OPO-based LIF Measurements of Atomic Oxygen and Nitric Oxide in Nanosecond Discharges.

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Atmospheric and sub-atmospheric pressure gas discharges recently increased their attractiveness to the biomedical industry, materials processing, and very recently plasma agriculture. The discharges have been mainly investigated considering the effect of the plasma on the treated sample. While many of these sources are in the initial stages of development, all of the applications share an interest in understanding the spatial distribution and temporal evolution of key intermediate plasma species, including ions, neutral metastable, and reactive species. One of the main molecular gases used to process surface as well as biological tissue is oxygen. In noble gas fed discharges O₂ is a minor component that is broken down leading to production of atomic oxygen and ozone.

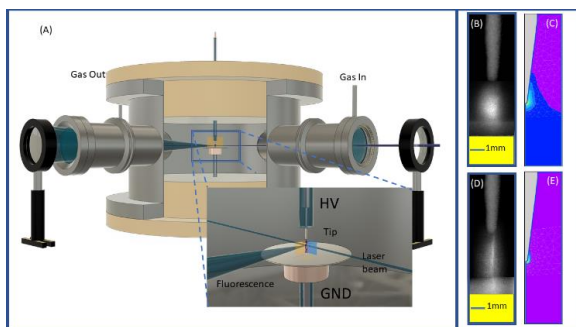


Fig. 1: Experimental apparatus setup with a closeup of the discharge region (A). ICCD images acquire for a single discharge event using a 200 ns gate are reported for 100 Torr (B) and 760 Torr (D). The corresponding DC electric field for the two cases are reported in inset (C) and (E).

In air discharges in which the main components are N₂ and O₂ the chemistry involved is more complex. The atomic oxygen is an important precursor for the ozone and N_xO_y species formation. The most important channels for O atom formation under non-equilibrium low-temperature discharges are dissociation of oxygen molecule by direct electron

impact and by N₂(A) metastable species [1]. The electric discharge used in the present work was obtained by using an asymmetric point to plane geometry based on a tungsten needle (HV) and a brass disk (GND) electrode (Fig. 1). LIF/TALIF was induced by a Nd:YAG pumped optical parametric oscillator laser (OPOTEK Opolette™ 355 LD) equipped with second and third harmonic, delivering up to 500 μJ @225 nm (rep. rate up to 20 Hz, linewidth 5 cm⁻¹, pulse duration < 7 ns). TALIF and LIF were used to study kinetics of atomic oxygen and nitric oxide in the afterglow of an air like mixture (N₂:5%O₂) fed nanosecond diffuse filament discharges. Production of atomic oxygen and nitric oxide was followed in the center of the gap at pressures of 100 and 760 torr. We observed slow increase of O atom production during first microseconds, long plateau of 20-30 μs duration, followed by single exponential decay characterized by time constants 239 μs and 89 μs at 100 and 760 torr, respectively. TALIF signals were detectable for post-discharge times up to 0.9 ms. The process involved in the formation of atomic oxygen and NO will be discussed, together with acquired spectroscopic data.

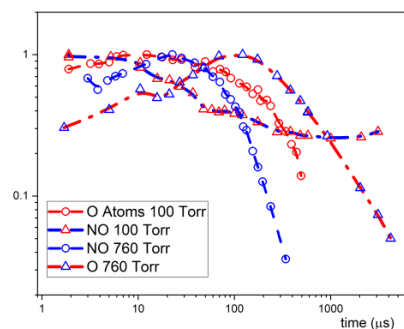


Fig. 2: Time evolution of Oxygen atoms and Nitric oxide at pressure of 100 Torr.

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Low energy electron induced processes on pyridine

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1. Introduction

The pyridine is found, for example, in nicotine or in B vitamins and it is used as a solvent or precursor for agrochemicals in fertilizers [1,2]. Pyridine in B vitamins has also been found on the surface of meteorites [3]. The aim of this work is the experimental study of electron-induced processes such as ionization and dissociative ionization, dissociative excitation, and ionization excitation of pyridine. hand margin on a separate line.

2. Results

Using two experiments, both crossed beams one utilizing quadrupole mass spectrometry and other optical emission spectrometry, we can provide more complex study of electron-induced processes. In the emission spectrum induced by 70 eV electrons the Douglas - Herzberg system of CH⁺, CH⁺ (B-A), C⁺, the hydrogen Balmer series, Deslanders - d'Azambuja system of C₂, Swann system of C₂ and CH (A-X), CH (B-X), CH (C-X), CN (B-X), NH (A-X) transitions are observed. In the mass spectrum the sequential dissociation of C, H and CH from parent of pyridine is observed. In Fig 3. we show an illustrative example of a 2D spectral map, which combines spectrum information and cross-sections into one whole.

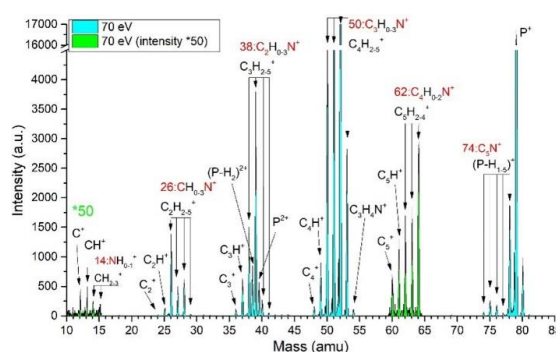


Fig. 2: Mass spectrum of pyridine

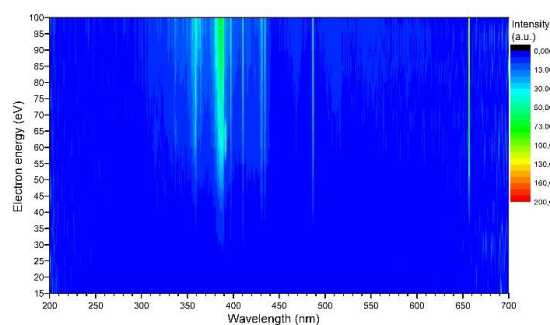


Fig. 3: 2D spectral map of pyridine

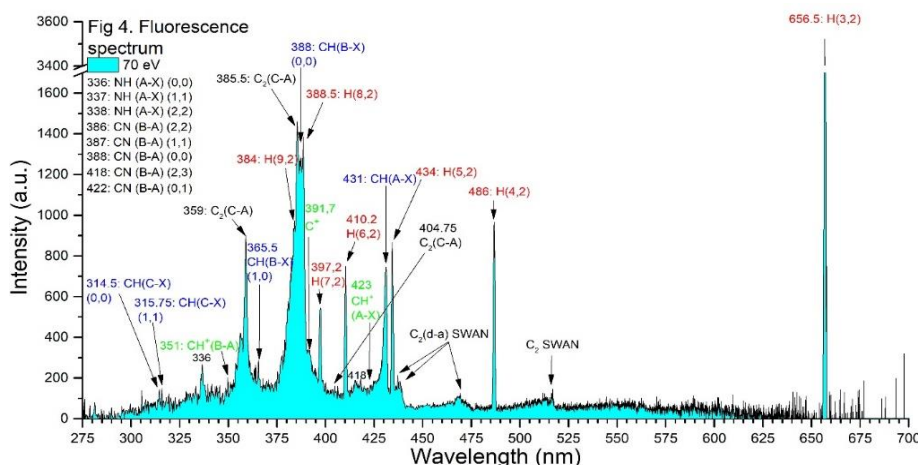


Fig. 1: Fluorescence spectrum of pyridine

This work was partially supported by Slovak Research and Development Agency under projects APVV-19-0386 and APVV SK-RU-RD-21-0020 and project VEGA 1/0489/21.

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Pretty exciting - Calibrated OH laser-induced fluorescence spectroscopy, opportunities and challenges in unravelling the role of H₂O in CO₂ plasma conversion

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1. Introduction

In the course of climate change, interest in CO₂ utilisation techniques like plasma conversion is steadily increasing. Striving for application, it is crucial to not limit the research to ideal laboratory conditions but also to consider the real world situation. In particular, impurities must be considered among which H₂O plays a vital role since it can potentially serve as abundant hydrogen source. To illuminate the role of water in carbon dioxide conversion, laser-induced fluorescence spectroscopy (LIF) is used to detect the hydroxyl radical in a pulsed DC glow discharge consisting of CO₂ with H₂O admixture. OH formed by H₂O dissociation, is of high interest due to its reactivity and can be used to probe the CO₂ conversion process.

2. Results

2.1. Quantitative OH LIF in a CO₂-H₂O glow discharge

A novel technique is introduced to calibrate the OH LIF setup by means of two-photon absorption LIF spectroscopy measurements on a CO-filled gas cell. OH densities in the order of $1 \times 10^{18} \text{ m}^{-3}$ are reported at a pressure of 6.67 mbar with the admixture of 20% of water and a discharge current of 50 mA. Furthermore, the time evolution of the CO₂ conversion is determined from collision energy transfer LIF and validated against literature. The time-dependent rotational temperature of OH is obtained with LIF thermometry [1].

2.3. Spectral crosstalk with excited CO

When decreasing the water content in the used glow discharge, spectral overlap between the 3064 Å system of OH, in particular the excitation of the P₁(3) transition for number density determination and the Q₁₂(1), Q₂(1), Q₁(6), Q₁₂(3) and Q₂(3) transitions for temperature determination, and the third positive system of CO, in particular transitions of the (0,0) and (0,2) bands, becomes apparent. The overlap is dis-

torting excitation and fluorescence spectra, see fig. 1, as well as fluorescence time decays after excitation of the P₁(3) transition of OH. As a consequence, systematic errors are introduced into the determination of temperatures, gas compositions and absolute number densities. A new set of transitions is proposed to circumvent the distortion while still allowing for quantitative measurements due to the availability of non-radiative rate coefficients for these transitions [2].

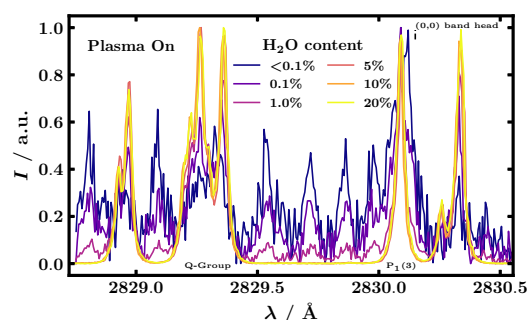


Fig. 1: Distortion of an excitation spectrum of OH for decreasing water content in a CO₂ glow discharge. The yellow line represents a spectrum dominated by OH. Important OH transitions are indicated at the bottom. Here Q-Group abbreviates the Q₁₂(1), Q₂(1), Q₁(6), Q₁₂(3) and Q₂(3) transitions. Appearing lines belong to the (0,0) band of CO, see the band head at 2830.15 Å.

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Measurement and study of surface-produced negative-ion in low pressure H₂ or D₂ plasmas

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1. Introduction

Plasma based negative-ion sources generate negative-ion beams that can be employed to inject particles in accelerators, to sputter or etch surfaces, or to generate neutral beams for fusion or microelectronic applications. Negative-ion (NI) production in plasmas is less studied than positive-ion one. There are basically two main negative ion (NI) formation processes: volume production by dissociative attachment of electrons on molecules and surface production through the electron capture by a neutral atom or an ion when colliding with a surface [1]. Contrary to positive-ions, negative-ions are not easily extracted from an ion source due to the plasma potential profile that confine them inside the plasma, and due to the fact that they have to be separated from the electrons before extraction. In this contribution we study H⁻ and D⁻ surface production and their extraction towards a Retarding Field Energy Analyzer (RFEA) collector.

2. Experimental set-up

Measurements are performed in an ICP reactor. The sample is placed in the middle of the plasma chamber 37 mm away from a NI detector. The sample is negatively biased with respect to the plasma potential. Positive ions from the plasma are attracted toward the sample and form NI, which are then accelerated toward the plasma. A single grid RFEA is placed in front of the sample to collect emitted negative-ions. Thanks to their acceleration in the sample sheath they can overcome the potential barrier due to the sheath in front of the RFEA and are self-extracted from the plasma. In order to prevent electron extraction together with negative-ion one, a magnetic barrier (500 Gauss) is placed in front of the RFEA giving rise to a Magnetised RFEA (MRFEA) depicted on figure 1 [2].

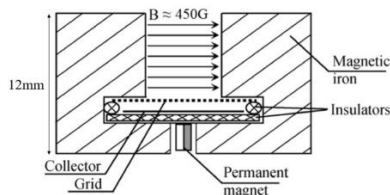


Fig. 1: Sketch of MRFEA device

3. Measurements

The collector potential is scanned and I-V curves are obtained (see figure 2). The negative charge saturation current (positive collector bias) is not used to determine negative-ion flux due to the possible presence of electrons escaping the barrier and secondary electrons generated by positive ion impact on the grounded grid.

Negative-ions are detected in the negative collector region where they start to be electrostatically filtered when the collector reaches and goes below the sample bias value. Their presence is revealed by a step on the ion saturation current. By differentiating the I-V curve, one can get the Negative Ion Velocity Distribution Function as depicted on figure 2.

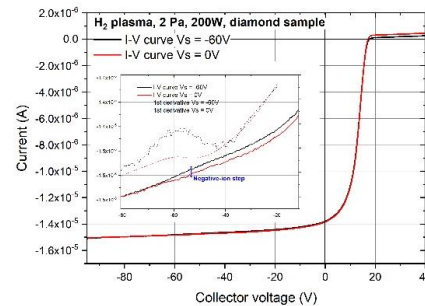


Fig. 2: I-V curve obtained when scanning MRFEA collector bias with sample bias at -60V (black, NI can be collected) and 0V (red, NI cannot be collected). Inset: zoom on I-V currents and their derivatives showing the NI velocity distribution function.

In this contribution the MRFEA diagnostic is studied. In particular attention is paid to the magnetic barrier transmission probability for positive and negative ions and for electrons. An attempt is made to estimate the sample emitted negative-ion flux using the measured negative-ion current at the MRFEA collector. As the negative-ion sample emission is strongly correlated to the sample work-function, a work-function in-situ diagnostic based on Photoemission Yield Spectroscopy (PYS) has also been installed on the experimental device and is employed to study negative-ion surface production together with the MRFEA diagnostic.

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Temperature Dependence of Non-thermal Rate Constants of Quenching and Vibrational Relaxation in the $\text{OH}(\text{A}^2\Sigma^+, v' = 0,1)$ Manifold

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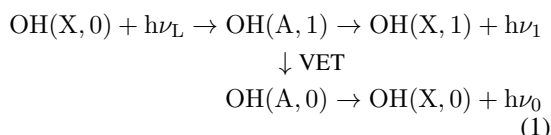
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1. Introduction

Collisional Energy-Transfer Laser-Induced Fluorescence (CET-LIF) has turned out to be an excellent tool for the determination of CO_2 dissociation in ns repetitively pulsed (NRP) discharges [1]. CET-LIF on OH allows for quantification of gas mixtures of known components from a measured OH LIF fluorescence spectrum, provided that the rate coefficients of the involved species are known [2]. The LIF scheme of interest is:



Atmospheric pressure CO_2 NRP discharges are spark events followed by a quick increase of the gas temperature in a few hundreds of ns and subsequent thermalization to room temperature in the far post-discharge [1, 3]. Consequently, the complete application of CET-LIF to these systems requires the knowledge of the temperature dependence of the rate coefficients for the relevant collisional processes.

In this contribution, the temperature dependence of vibrational energy transfers (VET) and non-radiative relaxations (electronic quenching) of $\text{OH}(\text{A},1)$ and $\text{OH}(\text{A},0)$ is presented.

2. Results

The measurements are carried out in a cell, where a constant flow of H_2O_2 is injected as a source of OH in combination with a fixed amount of collider gas at 300K. Photodissociation of H_2O_2 by a pulsed 266 nm laser beam ensures the production of $\text{OH}(\text{X})$, while a second laser beam at about 281 nm allows for LIF detection according to the scheme (1). Nascent OH fragments are translationally very hot. Elastic collisions with the surrounding gas re-equilibrate OH velocity towards a Maxwellian velocity distribution that approaches room temperature after a few hundred ns. By varying the delay τ between the two laser beams, exploration of different kinetic regimes is possible and the energy dependence of the collisional rate constants can be determined.

From a measured rate coefficient $k(T)$, the temperature-averaged cross section $\sigma(T)$ can be inferred as the ratio of $k(T)$ and the average collision

velocity. For illustrative purposes, Fig. 1 shows the temperature evolution of both the rate coefficient and the cross section of total quenching of $\text{OH}(\text{A}, 1)$.

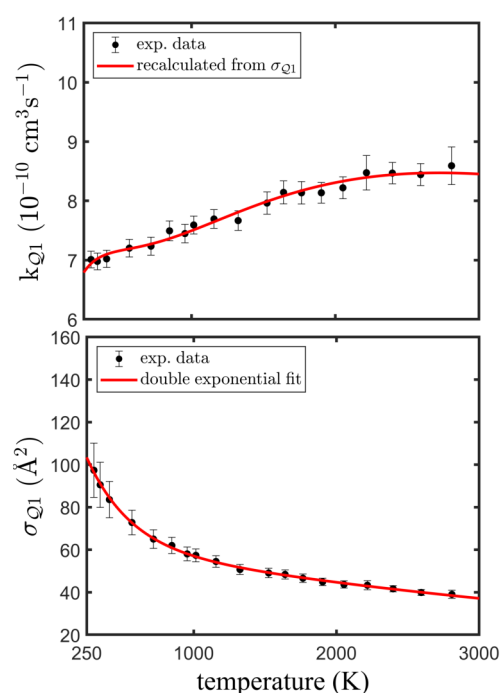


Fig. 1: Temperature evolution of the rate constant k_{Q1} and the cross section σ_{Q1} of total quenching of $\text{OH}(\text{A}, 1)$. Dots: experimental data; blue and green: fits according to [5] and [4]; red: double-exponential fit.

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Determination of electron temperature and electron density of a Plasma Jet Printer using Optical Emission Spectroscopy and line intensity ratio method

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1. Abstract

Flexible printed electronics are generally printed by 2D direct-write printing and produce various types of devices on flexible substrates using appropriate materials in the form of inks and colloidal suspensions. Conventional printing technologies such as Aerosol Jet Printing (AJP) and Inkjet Printing (IJP) only deliver the material onto a surface, requiring thermal or chemical post-processing. It has been shown that a novel micro-plasma based Plasma Jet Printer (PJP) using Dielectric Barrier Discharge (DBD) can print and sinter at the same time [1][2]. However, the relationship between various plasma and print parameters, such as electron density and print density, with parameters such as voltage, frequency and gas flow rate is not completely understood. While a 2D model of a DBD was presented previously [3], this work covers the electron temperature and electron density measurement of the plasma jet printer using optical emission spectroscopy and line intensity ratio method[4][5]. Using this measurement, we provide an understanding on how the micro plasma varies with various parameters such as voltage, frequency and gas rates and its effect on the sintering of micro plasma.

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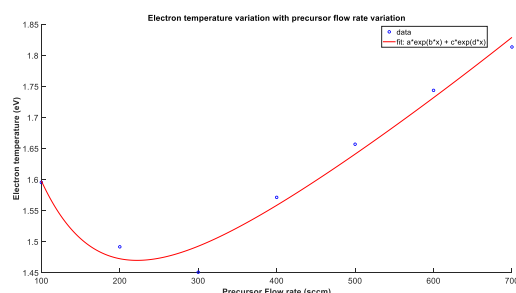


Fig. 1: Electron temperature variation with precursor flow rate variation

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On the determination of the vibrational temperature by Optical Emission Spectroscopy

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The vibrational excitation of molecules in gas discharges is of great interest in the kinetics of gas phase processing. An actual issue is its importance for the dissociation of CO₂, for example. The experimental determination of the vibrational distribution function, VDF, or, more modestly, of the vibrational temperature, is then an attractive issue, even more appealing if done with a simple apparatus with good time resolution, like with optical emission spectroscopy (OES). Still today, a persistent mistake is found in the literature, that is the use of the “vibrational temperature” of an emitting electronic state as somewhat representative of the vibrational temperature of the gas in the discharge. Such a temperature being determined by fitting the spectrum by some software, free or commercial. Besides the misuse of the word *temperature*, the results of such fittings are ambiguously named “vibrational temperature” and sometimes used to argue about the vibrational non-equilibrium and its variation with discharge conditions or time in pulsed discharges. What has this *temperature* to do with the vibrational excitation of the molecules’ ground state - T_v , i.e., of the large majority of gas components? It is well established and should be well known that the vibrational population of the excited state is connected to that of the ground state through the excitation process, quenching and vibrational relaxation in the manifold of the excited state. Nevertheless, this is very often ignored in the literature. In this contribution, we discuss the issue with the example of the “vibrational temperature” of the N₂(C,v) manifold, T_v^C , showing how much all the mentioned parameters can drive to incorrect deductions from an anyway conceptually wrong measurement. The N₂(C,v) manifold is the best characterized one, with decent knowledge of the electron impact excitation cross sections and collisional rate constants [1, 2]. Using the collisional-radiative model of [1], we have calculated how the ratio T_v^C/T_v can change by modifying the collisional conditions (gas pressure or gas composition). Some calculations are shown in figures 1 and 2. Although the N₂(C) state might be peculiar, its example shows the kind of misinterpretations found in the literature. The present paper is, then, mainly a warning against this widespread mistake, and an attempt to define the boundaries and explore the possibilities of a correct

use of OES for the determination of the VDF of the ground state of molecules.

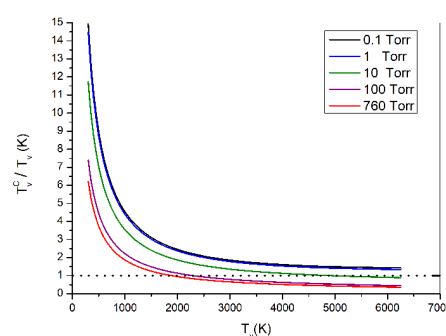


Fig. 1: T_v^C/T_v ratio vs. ground state vibrational temperature at 5 pressures, showing how much the collision processes in the N₂(C,v) manifold influence the population of vibrational levels.

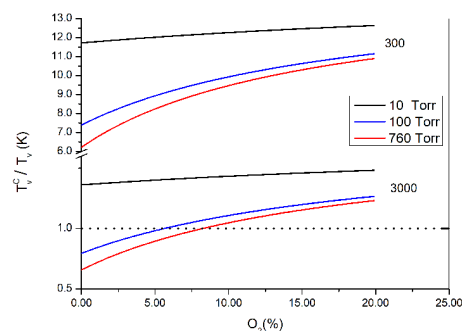


Fig. 2: T_v^C/T_v ratio vs. O₂ percentage in a N₂ – O₂ mixture up to the air composition. T_v is fixed at the two values of 300 K and 3000 K. The change in gas composition can substantially alter the population of the N₂(C,v) levels.

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Spectral investigations of discharge plasma on complex structured cathodes

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1. Introduction

Plasmas can form complex 3D structures hard to investigate in terms of space and time. Frequently the physical access is limited, and the dynamic behaviour (hysteresis or transient phenomena) requires high temporal resolution of the diagnostics [1-4]. Emission spectroscopy of low temperature plasmas inside a vacuum chamber has to be carried out through a glass or plexiglass window (Fig. 1a,b) or by fiber optics inserted into the chamber by feed-throughs (Fig 1c).



Fig. 1: Optical measurements through (a) a window; (b) lens projection on a screen, (c) a fiber optic adaptor.

Fiber optics have the advantage that the light can be collected near the plasma. Drawbacks are possible deposition on the fibre, perturbation of the discharge, and intricate variations of the position. Readings from outside of the chamber are more straightforward (see for instance Fig. 2 [1]).

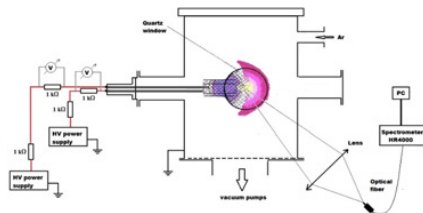


Fig. 2: Typical experimental setup with lens projection.

The so-called eyepiece projection, also used in astrophotography [5], creates an image of the plasma either on a 2D mobile screen with fixed fiber position or on a screen with grid holes where the optical fiber can be shifted.

2. Setup and data

Here we present a setup for measuring electron density and temperature along an axis, similar as in Fig. 2 [1]. The cathode system consisted of two concentric gridded cylinders [4]. Exemplary results are shown in Fig. 3 [4]. The temperature was determined from the average of the Boltzmann plot slope [6]. Electron density profiles have been calculated using relative intensities of neutral atomic lines and singly charged ionic lines according to Saha-Eggert [1].

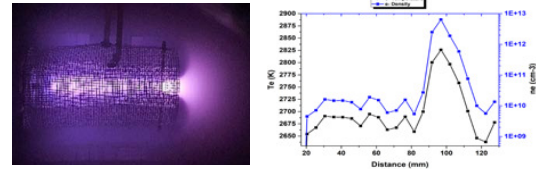


Fig. 3: Axial distribution of electron temperature and density for $V_{int} = -275$ V and $V_{ext} = -100$ V at $1.2 \cdot 10^{-2}$ mbar. Ar: 0 mm corresponds to the closed end of the external cylinder ("ext" - on the LHS), 100 mm corresponds to the open end of the external cylinder (on the RHS).

A problem using a single lens projection was that the plasma image was so small (1:5) that the fiber had to be moved in 1 mm steps. More resolution is required for comfortable readings, wherefore a bigger image is needed. This can be done by using an Ome-gon Telescope AC 80/400 on a tripod (Fig. 4).

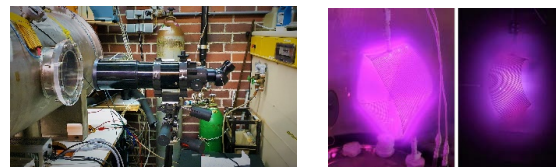


Fig. 4: Typical experimental setup with telescope projection and a Möbius band electrode [4].

The image was larger, however, the focal length of 400 mm required the screen to be placed at 2-3 m. A way to avoid this is to use telescope eyepieces, however, light absorption through multiple lenses is disadvantageous. Another possibility is using an apochromatic astrograph telescope with Extra low Dispersion lenses to avoid chromatic and spherical aberrations, and a laser collimator in order to point the scope at a specific region of the plasma before collecting data.

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Investigation of the dry reforming reaction by plasma-catalysis

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Methane emissions have increased in the past decade, with a substantial contribution from the agriculture, waste, and fossil fuel sectors. The dry reforming of methane (DRM) reaction converts one molecule of CH₄ and one of CO₂ into syngas, a value-added gaseous mixture of carbon monoxide and hydrogen [1]. Although the benefits of DRM are prominent and this process has been studied for decades, it still struggles to become a mature industrial technology, mainly because of its low energy efficiency and catalyst deactivation at high operating temperatures. Among non-thermal plasmas, the pulsed nanosecond discharge is gaining growing attention as one of the most energy-efficient ones to promote chemical reactions, taking advantage of the high electron densities and electron energies that can be reached out of thermal equilibrium.

In recent years, a developing field of interest has been the study of the effects caused by changes in the discharge's pulsing scheme. In the plasma-driven CO₂ splitting, Montesano et al. showed that conversion and efficiency increased by shortening the time between successive discharges for the same total energy [2]. Below 100 μ s, subsequent pulses do not act independently but occur in an environment perturbed by the initial pulse. The same phenomenon was then investigated for the DRM reaction [3]. At the maximum explored SEI (Specific Energy Input), around 6 kJ dm⁻³, the CO₂ conversion doubled, and that of CH₄ increased by almost 50%, by shortening the pulse interval (Tp) from Tp > 833 μ s to Tp < 40 μ s. We suggest that the increased performance observed shortening the pulse separation is due to the progressive modification of the discharge conditions (gas composition, temperature, and load impedance). A "memory effect" is present when closer pulses are coupled in a post-discharge medium not fully relaxed to the initial conditions. The imaging of the discharge supported this finding. When the camera exposure time is large enough to catch three subsequent pulses, a difference in the discharge path is observed at varying inter-pulse times. They

are spatially independent for inter-pulse times greater than 100 μ s, while tending to 40 μ s they converge to follow the same path.

An upgrade of the reactor configuration can be achieved by integrating a catalyst in the discharge reactor. Early results show an improvement in the DRM performance after introducing an alumina monolith cylinder coaxial to the discharge (Figure 1). Further investigations are ongoing.

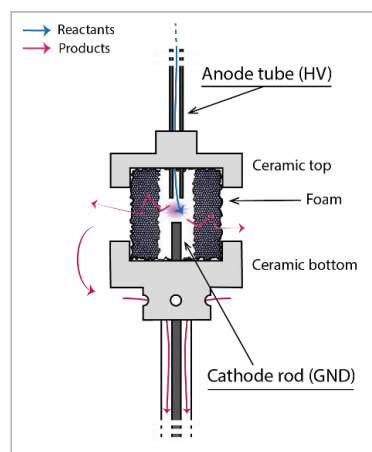


Fig. 1: Configuration of the NRP discharge and monolith foam catalyst.

Acknowledgements

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in situ FTIR transmission experiments through catalytic pellets under CO₂-CH₄ plasma exposure

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1. Introduction

Closing the Carbon cycle can be achieved by converting CO₂ into platform molecules or even short hydrocarbons, by coupling renewable energies and hydrogenated co-reactants (H₂O, CH₄, etc.). Several approaches are being explored and non-thermal plasmas is one of them. The principal advantage is to promote the asymmetrical stretching vibration for CO bond breakage. However, the presence of a catalyst could greatly improve the conversion and selectivity. Although, the complexity of the interaction of plasma with a surface brings the necessity to study the underlying mechanisms occurring on the catalyst as a function of time and under different conditions.

Glow discharge plasma reactor at low pressures are a good benchmark for studying plasma kinetics and especially plasma surface interaction. *in situ* studies of surface reactions under plasma exposure for CO₂ conversion and especially for Dry Reforming of Methane Reaction (DRMR) are scarcely reported. [1,2]

In this study, *in situ* FTIR transmission experiments were performed to elucidate the plasma-catalytic surface reactions in detail on cerium oxide (CeO₂) during plasma-assisted DRMR. The catalyst pellet is being exposed to a low-pressure glow discharge for which many parameters such as ro-vibrational temperature, electric field, molecules densities have also been measured. The combination of time resolved surface species evolution together with the detailed characterization of the gas phase provides a unique system for describing the plasma/catalyst interaction mechanisms.

2. Experimental

A glow discharge reactor is placed in the beam of a FTIR. The plasma is ignited perpendicularly to the IR beam which is passing through the pellet. CeO₂ was selected as a test material due to its oxygen mobility and redox properties, as well as the literature available for IR bands identification of adsorbed molecules. [3,4] Nanopowder of cerium oxide from Sigma-Aldrich was pressed to form a pellet of ~1 cm diameter.

Each experiment follows the next sequence: the appropriate gas mixture (CO₂ alone or CO₂-CH₄) was sent to the reactor (Before plasma), then plasma at 10 mA was ignited, plasma current increased to 50 mA,

plasma was switched off (After plasma), O₂ gas was sent to the reactor, O₂ plasma at 30 mA was ignited and finally, then O₂ gas. Each step takes 10 minutes.

3. Results

Firstly, CO₂ is observed to adsorb in ceria as tridentate carbonates (TC) and hydrogen carbonates (HC). [3] When plasma is ignited, the carbonates bands are reduced while strong bands corresponding to formates species appear on the spectra. [4] When plasma is off, TC bands reappear but those for HC do not with the same intensity as before plasma. Formate bands remain the same.

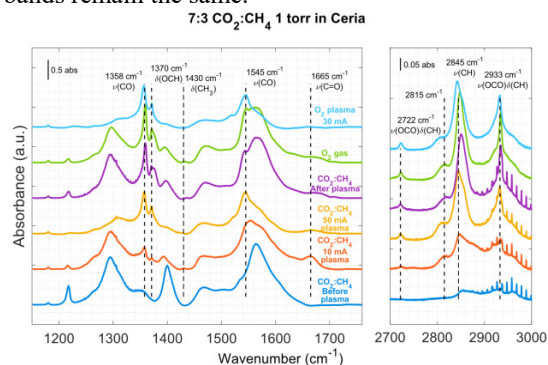


Fig. 1. *in situ* FTIR spectra following each step from bottom to top

Only formate bands remain during the cleaning step with O₂ plasma at 30 mA showing the stronger bond formed to the surface in comparison to carbonate species. Also, it was confirmed that water adsorption on the surface prevents the formation of HC species.

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Investigating picosecond two-photon absorption laser-induced fluorescence in Kr for N- and H-atom density calibration in reactive media

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1. Introduction

The consideration of fast (nanosecond-*ns*) and ultrafast (picosecond-*ps* and femtosecond-*fs*) two-photon absorption laser induced fluorescence (TALIF) for the study of gas flows, flames and plasmas is of a major importance in different research groups [1]. TALIF allows for the quantification (space and time analysis) of key atomic densities (e.g., N, O, H), leading to a better understanding of their role in kinetics. It is thus essential for the optimization of various applications (material synthesis, biomedical, environmental, etc.).

Particularly, the implementation of *ps*- and *fs*-TALIF for quench-free atomic density measurements in collisional media is very promising [1]. In *ps/fs*-TALIF, however, a substantial depletion of the densities of the laser-excited states through photoionization (PIN) and/or amplified stimulated emission (ASE) can happen, leading to saturated fluorescence regimes [2]. These processes must be understood and avoided when absolute density measurements are sought from TALIF signals. Such an understanding is particularly needed in the case of *ps*-TALIF in krypton (Kr). Contrary to *ns*-TALIF, the use of *ps*-TALIF requires further assessment of the conditions where Kr may be still considered for calibration of H- and N-atom densities in reactive media. Our study here focuses on the investigation of the relative predominance of the processes involved in the *ps*-TALIF scheme of a Kr atom.

2. Results

Herein, *ps*-TALIF is applied in Kr at different pressures ($P_{\text{Kr}}=0.1\text{--}10$ mbar). The laser intensity (I , units W cm^{-2}) is tuned ($1\text{--}480$ MW cm^{-2}) and the depletion of the density of Kr $5p'[3/2]_2$ fluorescing state through PIN and ASE is investigated. This is done by combining TALIF experiments with a simple 0D collisional-radiative model. For $P_{\text{Kr}}=3$ mbar and $15 < I \leq 480$ MW cm^{-2} , a saturated

fluorescence signal is obtained, which is largely attributed to PIN, ASE being negligible (figure 1; yellow) [3]. Also, a broadening of the two-photon absorption line ($4p^6\ ^1S_0 \rightarrow 5p'[3/2]_2$) is recorded (not shown) due to the production of charged species through PIN inducing a Stark effect. For $I \leq 15$ MW cm^{-2} , PIN is limited, the absorption line is narrowed, and the quadratic dependence of the TALIF signal intensity versus the laser energy (E_{Laser}) is obtained (figure 1; green). Thus, the Kr TALIF scheme, using the $5p'[3/2]_2 \rightarrow 5s[3/2]_1$ fluorescence channel, can be used for calibration purposes in *ps*-TALIF studies.

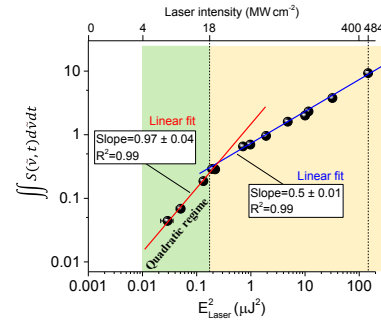


Fig. 1: Space-time integrated TALIF ($\iint S(\vec{v}, t) d\vec{v} dt$) vs squared laser energy (E_{Laser}^2) and I (top) at $P_{\text{Kr}}=3$ mbar.

In this study, the role of PIN on the depletion of the Kr $5p'[3/2]_2$ fluorescing state is investigated and conditions for using Kr as a calibrating gas are shown. These results contribute to the development of *ps*-TALIF for determining absolute densities and quenching coefficients of H- and N-atoms in reactive media such as atmospheric pressure plasmas.

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A low temperature atmospheric pressure plasma comparison of a novel multi-electrode source and surface dielectric barrier discharge

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1. Introduction

Low temperature atmospheric pressure (LTAP) plasmas have interesting physico-chemical properties that have made them useful in a variety of applications, from biological inactivation to alteration of photovoltaics [1, 2]. With different applications, feed gases and engineering requirements these LTAP sources take a variety of forms, such as plasma jets or surface dielectric barrier discharges (SDBDs) [3, 4]. The aim of this work is to investigate and compare two sources, a novel LTAP jet plasma source which utilises multiple powered electrodes (180° frequency phase shift between each) and an SDBD with a triangular valley separating live and ground. The aim of this is to evaluate each for usage in commercial/industrial settings. Here we present the electrical and optical measurements used to characterise the devices, and demonstrate their experimental utility.

2. Method

The plasma in both devices was powered utilising the same method, using a signal generator subsequently feeding into a sinusoidal power supply and transformer. The frequency was varied from 3kHz to 30kHz, and the voltage range tested was from 0 to 10kV. The novel multi electrode source (ID: 1mm - 9mm, OD: 3mm - 11mm) consisted of two phase shifted live electrodes on quartz tubing, with a ground separating them. The SDBD consisted of a brass electrode implanted inside a polyether ether ketone (PeEK) barrier, while a grounded saw-tooth aluminium electrode with spikes of varying dimension (width 2mm – 0.5mm) was attached to the other side (5mm electrode gap). The feed gases for the jet included Helium (99.996%) and Argon (99.998%), while the SDBD was also used with Nitrogen (99.9999%) and air.

The electrical properties were primarily measured using a Tektronix TBS1052B oscilloscope, a Pearson through-wire ratio 1:1 current probe, and a TESTEC HVP-15HF 1000:1 voltage probe. The optical properties were measured utilising an CMOS optical emission spectrometer (OES), (AvaSpec-ULS409CL-EVO-UA-10) which has a resolution of 0.50-0.70 nm, this was used with a 400 µm fibre

optic sensitive in the 200 nm to 1100 nm range.(FC-UVIR400-2-BX). Furthermore, these electrode setups were also tested in-situ via metrology in the National Physical Laboratory (NPL), and spectrometry experiments in Swansea University.

3. Results

Preliminary results, such as figure 1 below, demonstrate that different plasma conditions result in changes of the observed spectral range. Shown here is the optical emission spectra collected from the SDBD, when Argon is used as the feed gas.

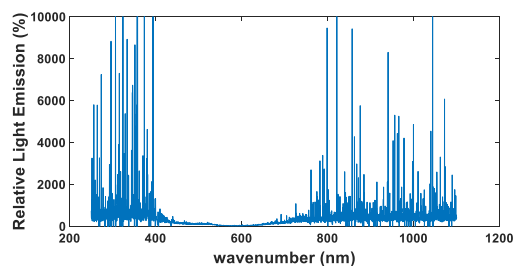


Fig. 1: An emission spectrum measurement using the SDBD with nitrogen plasma, examined under OES

From our results in Swansea University, Liverpool and NPL, we were able to show various utilities involving both the novel electrode setup and the SDBD. Further findings include their advantages, key properties and potential future avenues of research and improvement (e.g. tuning, efficacy, containment) to be presented in conference.

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Capacitively coupled radiofrequency discharge with a structured electrode

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Low pressure gas discharges are often used for the treatment of solid surfaces that are brought in contact with the plasma medium. Applications can target the modification of surface properties like the wettability, chemical composition, biocompatibility, or microbiological sterilization may be aimed for. In most cases a homogeneous treatment over the whole surface area is important, thus every surface element should be exposed to the same flux of the process relevant active plasma species. Depending on the desired process, these active elements can be ions, radicals and UV/VUV photons emitted by excited species. The homogeneity requirement becomes particularly challenging in the case of objects with complex shapes.

The spatio-temporal dynamics of electrons in low-pressure capacitively coupled radiofrequency (RF) discharges with a structured electrode is investigated. We apply Phase Resolved Optical Emission Spectroscopy (PROES) at the 706 nm He line that reveals information about the excitation dynamics with high spatial and nanosecond temporal resolution within the RF period. In contrast to earlier independent experiments [1] and numerical works [2], the present study combines the two approaches. Our experiments are accompanied by numerical simulations of the equivalent system, that are validated against the experimental data and provide further details about the particle fluxes at the different electrode segments.

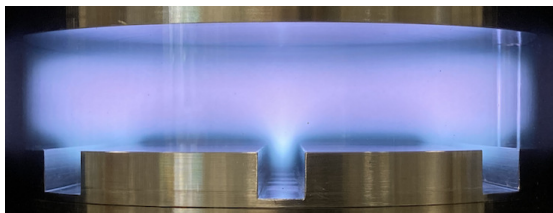


Fig. 1: Photo of the discharge in He at 60 Pa.

The discharges are operated in helium gas. The lower (grounded, $D = 14$ cm) electrode has 1 cm wide and deep parallel grooves machined into its plasma facing side along the diameter and 5 cm apart, as

shown in Fig. 1. The inter-electrode gap is 2.8 cm. The discharges are driven at a frequency of $f_{\text{RF}} = 13.56$ MHz, the pressure is varied between $p = 40$ Pa and 200 Pa, while the peak-to-peak driving voltage is fixed at $V_{\text{pp}} = 340$ V.

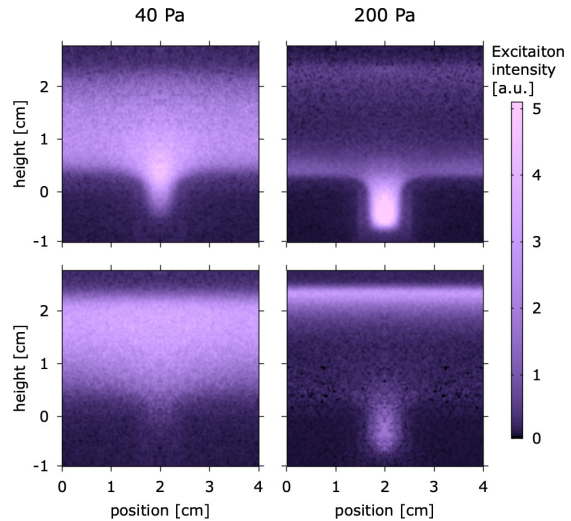


Fig. 2: Snapshots of the lifetime de-convoluted excitation intensities at 40 Pa and 200 Pa.

Fig. 2 shows example snapshots of the excitation intensity distributions at low (40 Pa) and higher (200 Pa) pressures at opposing phases of the RF period, where the intensities peak near the grounded electrode (upper row), and near the powered electrode (lower row). The penetration of the discharge into the trench and enhanced excitation inside the trench is found with increasing pressure.

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Time Resolved Spectroscopy of Discharges In Conducting Liquids

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Plasmas in liquids have many applications in areas such as material science and health care [1]. Recently the focus of the plasma-liquid interaction research has expanded to include various applications such as nanoparticles synthesis, material surface functionalization and water treatment [2,3,4]. With low voltage pulses and higher conductivity ionic solutions, the discharge normally follows the formation of a bubble or gas film around the electrode tip [5,6].

In this work, short HV pulses of ~7kV were applied to aqueous solutions. Typically pulse widths were < 10 μ s and frequencies were ~ 1 Hz or lower. The frequency was kept low to minimise heating of the liquid. Two tungsten wire electrodes separated by a micro-gap of less than 1 mm were immersed in the solution. The production and evolution of plasma breakdown in saline (NaCl) and potassium bromide (KBr) solutions were investigated. An intensified charged-coupled device (iCCD) camera, iStar Andor, was used to take Images at various delays, enabling the creation of a sequence of frame-by-frame images to monitor the evolution of the vapour layer over time. The time evolution of emission spectra was also monitored with the iStar camera attached to an Oriel MS125 spectrometer. H α , H β and OI emission lines are the most noticeable lines in the spectra. Fig. 1 shows the time resolved spectroscopy results for H α emission at KBr solution at different delays with a fixed time width 0.5 μ s. These results are being analysed to estimate electron densities and temperatures.

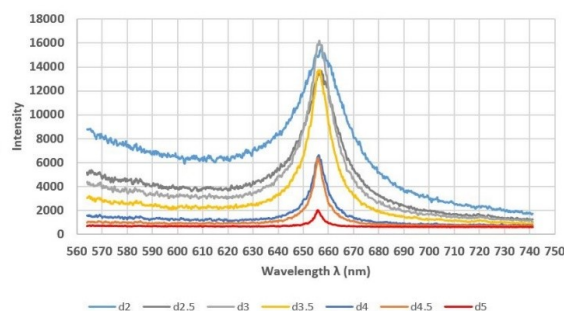


Fig.1 H α results for KBr solution at different delays; the labels dx refer to time windows of x to x+0.5 μ s; e.g., d4 indicates a time window of 4.0 to 4.5 μ s after the initiation of the discharge.

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Plasma Immersion Ion Implantation of PEEK and PDMS: Using Optical Properties and Surface Roughness to Predict Ion Dynamics

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1. Aim/Introduction

Plasma Immersion Ion Implantation/Deposition (PIII/D) is surface modification technique, which utilises a pulsed dielectric barrier discharge (DBD) to implant energetic ions into polymer, silicon and metallic substrates. The novelty of this technique is its ability to alter the material properties of polymers, such as their hardness, surface energy and wear resistance. Since the plasma is formed using a pulsed discharge, the system never comes to thermal equilibrium [1]. Thus, traditional plasma diagnostic techniques, both optical spectroscopies and diagnostic probes, are often not sufficient to determine the plasma parameters on their own. In this work, the dynamics of the plasma were investigated using changes in the surface morphology, optical properties, and UV absorption/transmission spectra of treated PEEK and PDMS.

2. Method

The dielectric function of both native and PIII/PIID treated poly-ether-ether-ketone (PEEK) is measured using Ellipsometry and fitted with both Lorentz and Gaussian Oscillator models. The resultant optical properties, specifically the imaginary part of the dielectric function (ϵ_2) are investigated for unique trends as a function of treatment time, hollow cathode voltage and pressure which could prove useful in providing a non-destructive measurement of the ion dose into an opaque polymer. The surface roughness is also measured for the same parameters.

3. Results/Discussion

Figure 1a depicts a plot of ϵ_2 as a function of energy for a range of treatment times from 0-30 min as described in the figure legend and caption. The shift in the dielectric function to lower energies is indicative of a change in the molecular structure of the PIII treated layer, and a narrowing of the band gap of PEEK [2]. Figure 1b shows the ratio of peak heights h_2/h_1 plotted with respect to treatment time, showing a mostly monotonic relationship until saturation at 30 min. Increasing the pressure, yields a greater ion density in the plasma, accompanied by the

aggregation of ions. However, only to an extent since collisions become more dominant and lower the average ion kinetic energy. This behavior is evident in figure 2 which shows a plot of the average surface roughness as a function of nitrogen gas pressure.

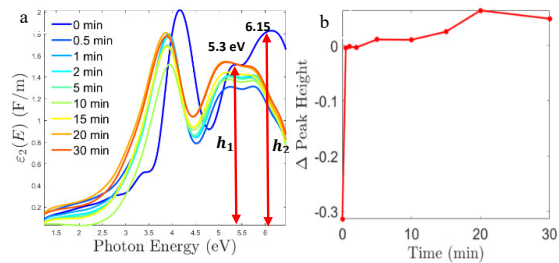


Fig. 1: (a) $\epsilon_2(E)$, plotted for treatment times of 0-30 min calculated from ellipsometry. (b) shows a plot of the ratio of peak heights.

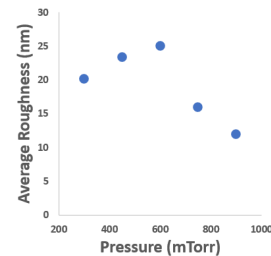


Fig. 2: Plot of Average Surface Roughness vs Pressure of Nitrogen Gas.

The average roughness and depth of the valleys and peaks in the modified polymer are indicative of; the kinetic energy of the ions, aggregation of particles and a measure of the relative ion fluence/density into the substrate. Using a combination of the two techniques, the relative ion energies and densities can be inferred, providing excellent diagnostic information for optimising future treatments for materials.

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Novel Dielectric Barrier Discharge Based Plasma Pen for Skin Treatment

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1. Introduction

Various plasma systems suitable for the living tissue surface treatment were constructed during the last decade. Majority of them is based on dielectric barrier discharge (DBD) in different configurations, mainly in form of jet. Helium is the mostly used working gas, but some systems were optimized also for use of Argon. The presented contribution gives plasma diagnostics results of novel planar DBD based system working in argon for larger area tissue treatment.

2. Experimental setup

System is based on circle grid electrode of 10 mm in diameter flowed by the working gas mixture. System is supplied by pulsed HV generator. Discharge was generated in various Argon-Helium and Argon-Air mixtures with the total gas flow of 100 Sccm.

The first experiments showed very good discharge homogeneity across whole electrode area, so detailed studies were focused on the radial distributions of active particles and plasma parameters, only. Radial profiles of intensities of Argon, Helium and Oxygen lines as well as OH, NO-gamma, and nitrogen bands (first and second positive and first negative) were measured in dependence on working gas mixture composition perpendicularly to the discharge plate. Based on them, argon excitation temperature, vibrational temperature of the $N_2(C^3\Pi_u)$ state and rotational temperatures of OH and nitrogen second positive system were calculated.

3. Results

The intensities of plasma active species are measurable in area exceeding the electrode diameter, as it is shown in Fig. 1. The primary working gas is effectively mixed with surrounding ambient air that leads to the energy losses into vibrational and rotational excitations of molecules. Thus, temperature of neutral gas in plasma outer part is higher than at its center (see Fig. 2). Results show that addition of helium into working gas mixture leads to the increase of argon excitation temperature as well as to the increase of rotational temperature that exceeds values applicable for the tissue treatment (see Fig. 2). Also vibrational nitrogen temperature increases. What is also important for the

tissue treatment, concentrations of all plasma active species (O , N_2^* , NO and OH) except nitrogen molecular ion significantly decreases with the increase of helium amount in the working gas. Addition of synthetic air leads to increase of all temperatures and concentration of active species except OH radical. All the obtained results show that the novel studied device should be operating ideally in the pure argon without addition of any other gases. The first pilot studies were carried out for the skin decontamination of selected pathogens like *Propionibacterium acnes* or *Propionibacterium granulosum*.

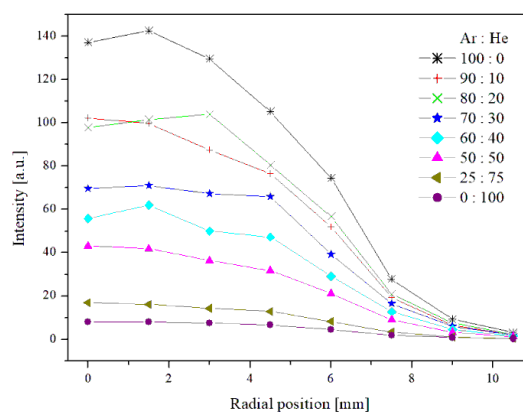


Fig. 1: Radial profile of OH radical intensity

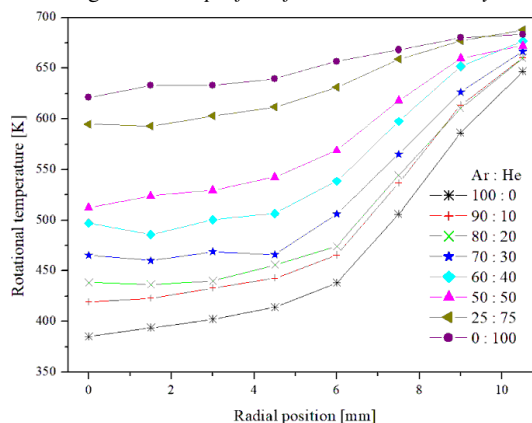


Fig. 2: Radial profile of OH radical rotational temperature

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Second Poster session

Second Poster session	
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Nanosecond surface dielectric barrier discharge: Experimental comparison of streamer to filament transition for O₂ and CO₂

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1. Introduction

Transition from streamer to filament is a common feature of nanosecond surface dielectric barrier discharge (nSDBD) at high voltages and high pressure [1]. The transition is characterised by the abrupt (1-2 ns) apparition of filaments. A filament, around 20 μm in diameter and with electron density about 10^{18} - 10^{19} cm^{-3} , appears for every 4-5 streamers already developing along the dielectric. These streamers are 200-300 μm in diameter and with electron density 10^{15} cm^{-3} . The transition is as fast as 1-2 ns.

2. Experimental set-up

The nSDBD was ignited in the system with a 20 mm diameter high voltage disk electrode, resting on a 50 mm diameter grounded electrode. The dielectric consisted of a 0.3 mm PVC sheet maintained on the grounded electrode by a 0.3 ± 0.1 mm layer of silicon glue. Single-shot high-voltage (HV) pulses of negative or positive polarity, 20 ns in duration (FWHM), 2 ns rise time and 20-60 kV on the HV electrode were produced by FPG20-03PM or FPG20-03PN pulsers (FID Technology). Voltage waveform was controlled by custom made calibrated back current shunts. ICCD images ($\lambda = 300$ – 800 nm) were taken by a Pi-Max4 Princeton Instruments ICCD camera. The gate of the ICCD was selected to be 12 ns from the start of the discharge, similar to [1].

3. Results and discussion

For the negative polarity, the transition point appears almost decoupled from the gas pressure. Addition of 1% of CO₂ or O₂ to nitrogen only causes a slight increase in said transition, with observable changes occurring at concentrations of 20% O₂. The apparent negative slope of these lines is mitigated by results obtained in [1], where the trend for N₂ inverted when increasing the pressure past 6 bar.

For positive polarity, for every mixture composition, a similar linearly decreasing behaviour of the transition threshold for increasing pressure is observed. Gas composition however appears to play a strong role, with the addition of 20% of O₂ leading to a 66% increase in the transition voltage amplitude.

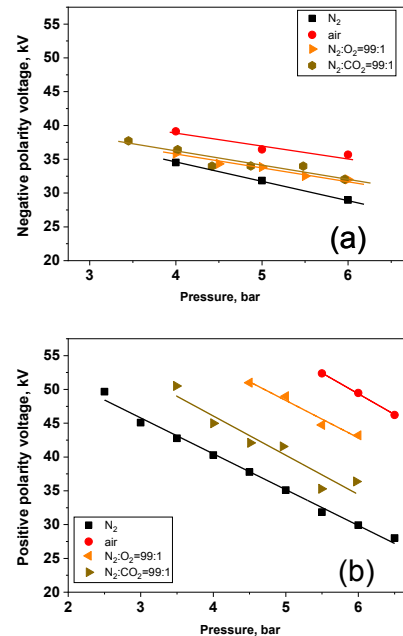


Fig. 1: Transition curves of (a) negative and (b) positive polarity discharge for varying pressure and gas composition. Values for N₂ and air taken from [1]

Differences between the CO₂ and O₂ curves in positive polarity can be indicative of two parameters thought to play a role in the transition physics, namely photoionization and electronegativity of the gases. Further experiments are required to determine this and are planned for the future.

4. Acknowledgments

This work was partially supported by IRP KaPPA (CNRS) and EP-DGA convention 2790.

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B-dot probe measurements of a periodic E-field structure for estimation of the power coupled into the plasma

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1. Introduction

A conceptually different mechanism for collisionless electron heating and plasma generation at low pressures was theoretically proposed [1]. It is based on a lattice of vortex fields, which produce certain resonances in velocity space. The particular periodic structure of the induced electric field is vital for its operation in the stochastic mode. Here we show measurements of the induced electric field in the inductively coupled array (INCA) discharge, a realization of this concept, for different coil configurations and the influence of the field forms on the plasma heating.

2. Theory

The power coupled into the plasma is calculated using the kinetic conductivity in Fourier space:

$$\left\langle \frac{\partial P}{\partial A} \right\rangle_{t,A} \sim \frac{\sigma_0 s}{A} \int d^2 k \left| \hat{E} \right|^2 \frac{k_0}{k} \exp \left\{ -\frac{k_0^2}{k^2} \right\} \quad (1)$$

(A – area of the discharge, σ_0 – RF conductivity). The characteristic wave number $k_0 = \omega_0/v_{th}$ (ω_0 – RF frequency, v_{th} – thermal velocity of the electrons) corresponds to spatial structures, which the majority of the electrons traverses within one RF period. The 2d Fourier transform of the external electric field $\left| \hat{E} \right|^2$ is assumed to decay exponentially into the plasma along the 3rd dimension with a decay length s .

3. Experimental

The INCA discharge consists of an array of 6×6 coils. The current in the coils is measured with a home-made pick-up coil placed beside the feed-through connecting the matchbox and the array [2]. The B-dot measurements are performed with a home-made measurement system without plasma. From these results the electric field structure in a region in the center covering 2×4 coils is obtained.

The performance of two different spiral coil forms in an ortho configuration (current through all coils in phase) is compared. The first one has the form of a standard spiral coil (gen. 1) and was originally used in INCA [2], whereas in the second configuration (gen. 2), the windings are concentrated towards the outer radius, approaching the form of an ideal current loop.

4. Results and discussions

The spatial electric field structure of both coil configurations (fig. 1) deviates rather strongly from the initially expected field distribution due to the spa-

tial structure of the individual coils and the finite lattice size.

The square of the absolute value of the Fourier transformed electric field (fig. 2), which is the input parameter for the calculation of the power coupled into the plasma (eq. (1)), reveals that for the gen. 1 coils the cardinal resonances are much stronger than the diagonal ones, although in the theory those resonances were expected to contribute equally. For the gen. 2 coils this effect is still present but less pronounced. Also the component at $k = 0$, which does not contribute to the plasma heating, compared to the other modes, is lower for the gen. 2 coils. Indeed, eq. (1), the power coupling with gen. 2 coils is more efficient.

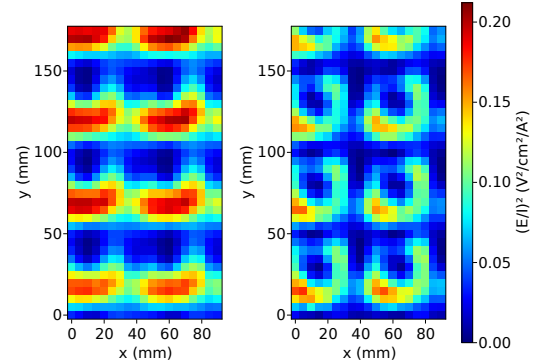


Fig. 1: E^2 from B-dot measurements for gen. 1 (left) and for gen. 2 (right) coils.

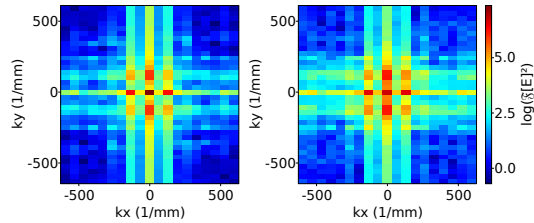


Fig. 2: Logarithm of the magnitude of the Fourier transformed fields from fig. 1.

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Fluorescence of oxygen induced by electron impact

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Optical emission spectroscopy was used to study electron impact induced fluorescence of oxygen. Oxygen is the third most abundant element in the universe, it is present in the atmospheres of Earth, Europa and Ganymede [1], or cometary comae. This study is a part of spectroscopic data analysis of the comet 67P/Churyumov–Gerasimenko coma acquired by Rosetta's Optical, Spectroscopic, and Infrared Remote Imaging System (OSIRIS) [2]. One of Rosetta's most intriguing findings was the persistent detection of O₂ in the coma of 67P in high abundances. These emissions are not only driven by fluorescence of atomic oxygen, but also contain contributions from dissociative excitation of other oxygen-bearing molecules [3].

The experimental apparatus built at Comenius University in Bratislava is based on the crossed electron and molecular beams method and is further described elsewhere [4]. The emission spectrum was measured within the wavelengths of 200–650 nm with 50 eV electron energy. The spectrum is shown in Fig. 1. The emission band in the wavelength range from 200 to 450 nm corresponds to the second negative system of O₂⁺ (A²Π_u - X²Π_g). Individual vibrational transitions of the system were identified as well. Several bands of the first negative system of O₂⁺ (b⁴Σ_g⁻ - a⁴Π_u) were detected within 490 – 650 nm. The emission lines of atomic oxygen O I and O II were identified in the spectrum as well. All the emission lines and bands were identified according to the previous research [5,6].

The emission cross sections Q_{em} of all identified transitions were determined for discrete values of electron energies within 19–100 eV, making a major contribution to the database of previously published experimental results. The aim of this experiment is to extend the collection of the emission cross sections to be sufficient for the study and theoretical modelling of the processes that occur on extra-terrestrial bodies.

Acknowledgements

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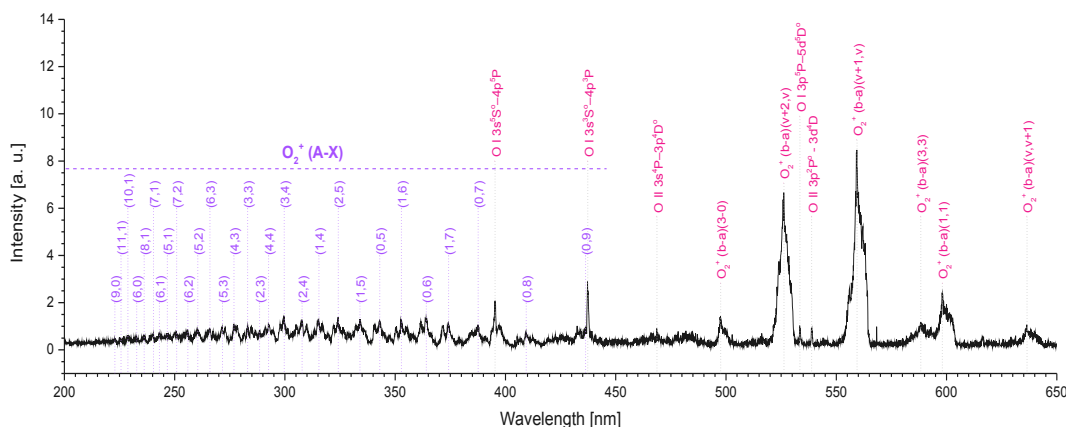


Fig. 1: The emission spectrum of O₂ measured at 50 eV. The transitions correspond to the systems O₂⁺ (A²Π_u - X²Π_g) (purple) and O₂⁺ (b⁴Σ_g⁻ - a⁴Π_u), O I and O II lines (pink).

Electrical and optical diagnostics of novel multi-electrode low temperature atmospheric pressure plasma system

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1. Introduction

Low temperature atmospheric pressure (LTAP) plasmas have interesting physico-chemical properties that have made them useful in a variety of applications, from biological inactivation to alteration of plastics [1, 2, 3]. Often different applications mean the plasma sources have various electrode configurations, applied waveforms, and feed gases. The aim of this work is to evaluate and quantify the properties of a novel LTAP plasma source which utilises multiple powered electrodes. The voltage/frequency applied is 180° phase shifted between each electrode. The aim of this source is to reduce the applied voltage on an individual electrode while maintaining the potential difference, making the system safer for use in commercial/industrial settings. Here we present the electrical and optical measurements used to characterise the breakdown voltage and other plasma properties.

2. Method

The plasma was powered using sinusoidal power supply, transformer and signal generator. The frequency was varied from 3kHz to 30kHz, and the voltage range tested was from 0 to 10kV. The plasma source (ID: 1 mm - 9 mm, OD: 3mm - 11mm) consisted of two phase shifted live electrodes on quartz tubing, with a ground separating them. A standard dual electrode formation was also used for comparison.

The primary feed gas was Helium (99.996%) and small admixtures of Nitrogen were also blended in. The electrical properties were primarily measured using a Tektronix TBS1052B oscilloscope, a Pearson through-wire ratio 1:1 current probe, and a TESTEC HVP-15HF 1000:1 voltage probe. Using these measurements and equations from literature [4, 5] estimations of the electron density and other plasma parameters were made.

The optical properties were measured utilising an CMOS optical emission spectrometer (OES), (AvaSpec-ULS409CL-EVO-UA-10) which has a resolution of 0.50-0.70 nm, this was used with a 400 µm fibre optic sensitive in the 200 nm to 1100 nm range. (FC-UVIR400-2-BX).

3. Results

An example of the electrical diagnostics is shown in figure 1, where the nitrogen content of the feed gas is varied from 0 sccm to 100 sccm. From this it is clear that the multi-electrode system can handle larger N₂ concentrations while maintaining lower breakdown voltages.

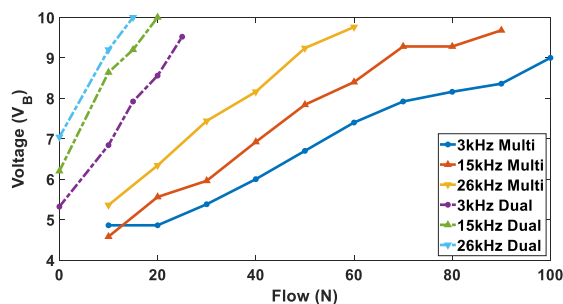


Fig. 1: A comparison of breakdown voltages per electrode for dual vs multi electrode systems

From our results in these experiments, we were able to show the advantageous properties of the novel electrode setup. Further results will be presented at the conference.

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Low energy electron attachment to $\text{Co}(\text{CO})_3\text{NO}$ clusters

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1. Introduction

Interaction of low energy electrons with molecules and molecular clusters is one of the elemental processes in plasma and plasma technologies. In our study we are focusing on electron attachment (EA) and dissociative electron attachment (DEA) on cobalt tricarbonyl nitrosyl ($\text{Co}(\text{CO})_3\text{NO}$). $\text{Co}(\text{CO})_3\text{NO}$ is used as a precursor gas for deposition of Co in Focused Electron Beam Induced Deposition (FEBID) and following Electron Beam Induced Surface Activation (EBISA) [1].

2. DEA to $\text{Co}(\text{CO})_3\text{NO}$ clusters

Ion yields from our cluster measurements (created by co-expansion with Ar gas) will be compared to previous EA and DEA gas phase $\text{Co}(\text{CO})_3\text{NO}$ studies [2] and He nanodroplet cluster studies [3]. The gas phase DEA products from Ar clusters agree well with previous works [2,3], comparison with existing gas phase data [2] is shown in Fig. 1.

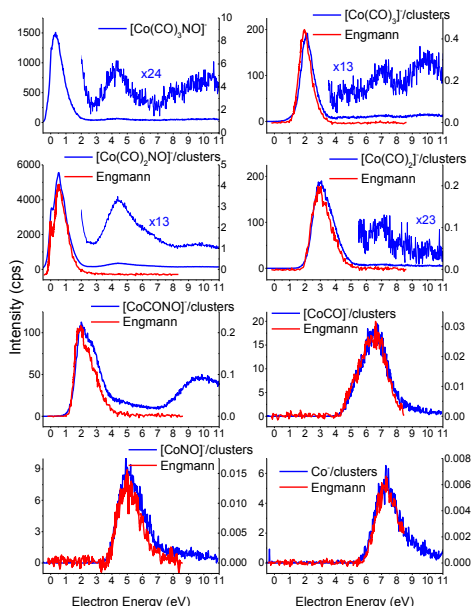


Fig. 1: Comparison of gas phase products from our Ar cluster measurements (blue) with pure gas phase products (red) [2]

However, formation of cluster products in He nanodroplets [3] and our co-expansion with Ar reveal

some differences mostly the shape of the ion yield curves due to the higher energy resolution of the electron beam used in our experiment. The exclusive formation of the molecular ion of $\text{Co}(\text{CO})_3\text{NO}$ in clusters was confirmed in our measurements but we have detected the ~ 0 eV resonance for this ion contrary to previous work [3], similarly to other cluster fragments shown in Fig. 2.

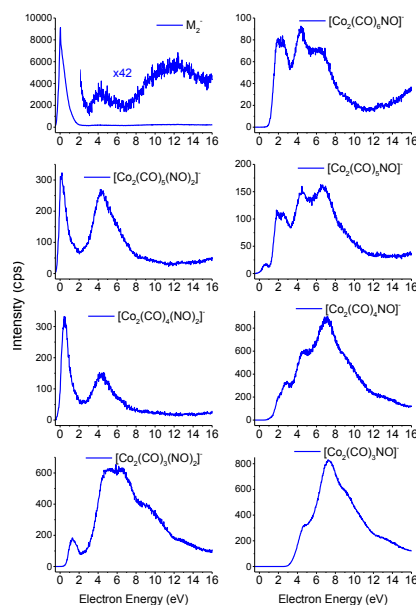


Fig. 2: Selected cluster fragments from our Ar cluster measurements

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Optical Emission Spectroscopy Study of Plasma Parameters in Low-Pressure Hollow Cathode Plasma Jet and Planar Magnetron Powered by Pulsed DC Power Supply

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1. Introduction

The passive optical emission spectroscopy (OES) is one of the most widely used cheap, versatile and non-invasive diagnostic method for low-temperature low-pressure plasmas study. The OES has proven to be a versatile techniques for the determination of electron density (n_e), electron temperature (T_e), and within certain energy limits also the electron energy distribution function (EEDF) in low pressure discharges. In our work the OES diagnostic technique is utilized for the analytical study of T_e and n_e of low-pressure Argon (Ar) plasmas as a function of applied power and Ar flow rate in a hollow cathode plasma jet (HC) and planar magnetron (PM) systems using pulsed DC power source.

2. Experimental setup

The scheme of the HC and PM for plasma parameters studies is shown in Figure. 1. HC is installed in a grounded vacuum chamber configuration which is designed explicitly for UHV conditions. The setup consists of a cylindrically shaped iron (99.995%) target with a through-hole along the axis. HC was mounted horizontally to decrease the number of microscopic fragments from the cathode. The length of the cathode is 29 mm, and the inner and outer diameters are 5 and 8 mm, respectively. The hollow cathode was fixed by the water-cooled copper blocks and insulated from the rest of the system employing a ceramic shield.

3. Results

The emission spectroscopic diagnostic method applied in our work combines measurements of certain argon line-intensity with collisional–radiative modeling of the excitation kinetics at low pressures. The modified Boltzmann method [1] and the line-ratio method [2] were deployed for T_e and n_e measurements, respectively. In figure 2 there are displayed T_e and n_e acquired in the active phase of the discharge pulse. The pulse repetition frequency was 100 Hz, the duty cycle 1:10. The displayed power is an average over the whole repetition period. However, here the electron temperature and density in the PM system is noticeably higher than that for the HC system.

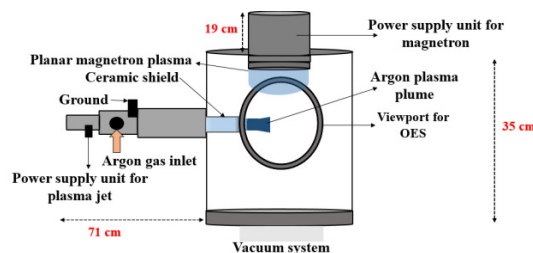


Fig.1: Scheme of hollow cathode plasma jet and planar magnetron sputtering system.

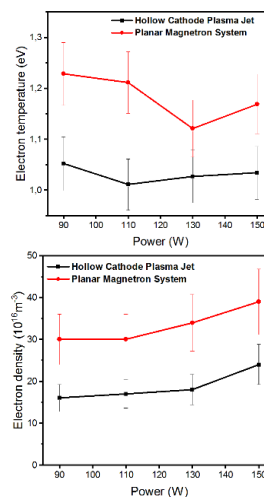


Fig 2: Electron density and temperature plots in HC and PM systems with changing power in pulsed DC operation for constant flow rate of 140 sccm and working pressure of 20 Pa.

3. Conclusion

The T_e and n_e PM data was greater than the HC data. The explanation may be that the plasma in the HC system decays faster than plasma in the PM system due to its smaller volume. That leads to smaller values when averaged over the whole pulse period.

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Collisional energy transfer-laser induced fluorescence (CET-LIF) on pulsed nanosecond discharges

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1. Introduction Plasma-driven conversion of the major greenhouse gases, namely CO₂ and CH₄ [1], represents a valuable technology to simultaneously store renewable energy and convert greenhouse gases into value-added compounds. Among the several plasma discharges, the efficiency of the atmospheric nanosecond repetitively pulsed (NRP) discharges is among the highest in converting CO₂ and CH₄ [2, 3].

An essential feature concerns how the energy is provided to the system. It was shown [2, 3] that a sequence of packets of close pulses in time (bursts) gives higher performance than a pattern of equally spaced pulses, for the same energy input. Time-resolved optical emission spectroscopy was employed to reveal the physical properties of a pure CO₂ splitting process in an NRP discharge [4]. In this contribution, collisional energy transfer-laser induced fluorescence (CET-LIF) is applied to the same system to probe the gas composition in the first microseconds of the discharge, where the presence of excited molecules, atoms and ions is still relevant to the kinetic of the process [4].

2. Experimental methods. The CET-LIF technique is used to measure the time evolution of the gas composition in the discharge gap. Following the addition of a trace of water, the discharge generates OH by dissociating H₂O. The excitation of the electronic state OH(A²Σ⁺, ν'=1) is obtained with a frequency-doubled dye laser (TDL50, Quantel; Rhodamine 590 chloride dye) pumped by a 10 Hz Q-switched Nd:YAG laser (YG580, Quantel). An intensified CCD (ICCD, DH334T-18U-03, Andor iStar) collects the fluorescence spectrum. The experiment is carried out at 745 Torr. The information on the gas composition, namely the instantaneous CO₂ conversion, is derived from the ratio between the two bands (1,1) OH(A²Σ⁺, ν'=1)→OH(X²Π, ν=1) and (0,0) OH(A²Σ⁺, ν=0)→OH(X²Π, ν=0) [5].

3. Results The investigation is focused on the first microseconds of the discharge pulses. Specifically, the burst mode characterized by short inter-pulse times (<40 μs) is studied. An example of fluorescence spectra is shown in Fig. 1. The reported spectra are collected after a time τ = 100, 480, 4000 ns from the beginning of the discharge. In the first 2 μs of the first pulse, the conversion raises to reach a value around 55%. The subsequent pulses of the burst contribute to a build-up of the dissociation that stabi-

lizes around the value of 70%. Preliminary results show that the burst mode allows achieving a maximum conversion comparable to the equally spaced pulsing scheme but provides less energy to the system. This finding may represent a further confirmation that the discharge acting on a pre-treated gas is more efficient than a discharge intercepting new gas [2, 4].

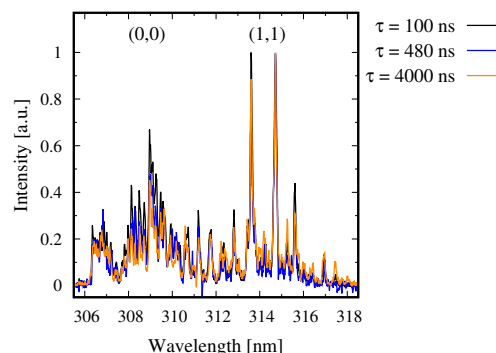


Fig. 1: Fluorescence spectra collected in the first pulse of a burst. τ indicates the elapsed time from the beginning of the discharge. The transition used to excite the OH(A²Σ⁺, ν'=1) state is the P₁(3).

Acknowledgments

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Diagnostics of void in capacitively-coupled rf dusty discharge

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Void in capacitively coupled rf dusty plasmas, i.e. a dust-free area formed in the central area of the discharge, represents the most common disturbance of homogeneity in microgravity experiments [1] as well as in dust-growing plasmas [2]. Balance of the electrostatic and ion drag forces on the microparticles at the void boundary has been since long time considered as a physical mechanism of void formation. However, recent experiments and simulations [3] have shown that this approach is incomplete.

The experiments [4] were conducted in a capacitively-coupled rf discharge with parallel-plate geometry. Two electrodes were driven in a push-pull mode by a sinusoidal signal with the frequency of 13.56 MHz. The experiments were conducted in the range of argon (fed with 3 sccm flow) pressures 20 to 80 Pa. We used plastic spheres with the diameter of $1.95 \pm 0.05 \mu\text{m}$ as dust particles.

Under ground laboratory conditions, the microparticles concentrate themselves in the vicinity of the lower electrode. To obtain large volumetric microparticle suspensions, we compensated the gravitational force by means of thermophoresis [5]. The temperature gradient between the electrodes was controlled by heating the bottom electrode with an electric heater and cooling the top electrode with fans. The temperature difference was 14 – 15 K during the experiments.

Diagnostics of the dusty discharge involved observation of dust, time-averaged and rf-period-resolved optical emission spectroscopy as well as laser-induced fluorescence.

Comprehensive diagnostics revealed that the void in capacitively-coupled rf dusty plasmas can exist in two qualitatively different regimes, which were termed “dim” and “bright” (Figure 1). The dim regime is characterized by the smooth emission profile: No feature in the plasma emission is associated with the void or void boundary. On the contrary, in the bright regime, the plasma inside the void exhibits stronger emission inside the void. Transition between the void modi was achieved by the variation of the discharge power and was manifested by a kink in the power dependence of the void size. Variation of RF-period-resolved optical emission spectroscopy revealed that the presence of the microparticles increases the emission in the entire axial extension of the discharge every half of the period in the case of

the dim void, whereas for the bright void, the emission increase appears only between the expanding sheath and the opposite void boundary.

Simplified 1D model of a dusty rf discharge has shown that only the bright regime can be explained by the balance of electrostatic and ion drag forces. The dim regime could be obtained only when the radial ion diffusion was phenomenologically introduced into the ion continuity equation: Only then, a stable plasma configuration provided by a very small axial electrostatic barrier, responsible for the void formation, could be formed.

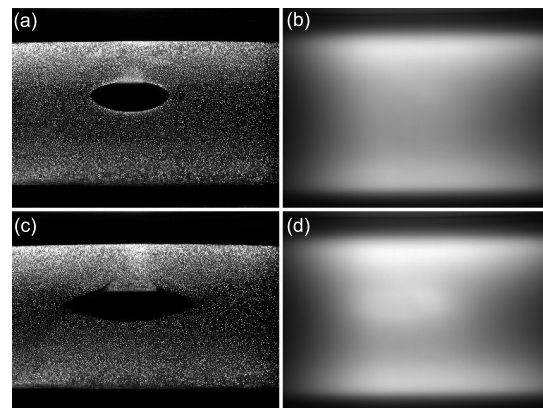


Fig. 1: (a), (c) Dust and (b), (d) plasma emission profiles in a capacitively coupled rf discharge in argon at 37 Pa pressure and (a), (b) 500 and (c), (d) 700 mW power corresponding to dim and bright void regimes, respectively.

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Design of a mid-infrared continuous-wave cavity ring-down spectrometer for in-situ trace gas detection

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For detailed temperature studies of molecules at trace gas level concentrations instruments of simultaneously high sensitivity and broad tunability are required. Established high-sensitivity laser absorption techniques are based on the theory of resonant optical cavities e.g., cavity enhanced absorption spectroscopy (CEAS) and cavity ring-down spectroscopy (CRDS) [1]. In the mid infrared, these techniques often utilize narrow radiation sources such as lead salt diode lasers or quantum cascade lasers, both of which are limited to tuning ranges below 10 cm^{-1} [2]. External-cavity quantum cascade lasers (EC-QCL) improve upon this with a mode-hop free tunability in the range of 100 cm^{-1} and at least an order of magnitude increased emission power [3]. Subsequently recent spectroscopic studies reported the use of EC-QCL sources for cavity enhanced spectroscopy setups at low and atmospheric pressures [2,4-6].

We present a broadband CRDS setup in the mid infrared region between 2000 and 2300 cm^{-1} (5 to $4.35\text{ }\mu\text{m}$) designed as a compact, robust, and adaptable spectrometer. Fig. 1 shows the encased optical board, which supports the laser source and wavemeter-based calibration. On triggering, an AOM directs the beam on a focal mirror and in-coupling optics for the subsequent cavity. Note the option for remote coupling adjustment during laser operation using i.e., piezo actuated mounts. The cavity is formed by a high-reflectivity mirror pair placed circa 0.6 m apart and flanged to a reactor using micrometer-screw adjustable port aligners. For tuning of the free spectral range, three strain-gauged piezo actuators simultaneously sweep the mirror spacing at sub-nanometer resolution. On coincidence with the fixed laser frequency, the AOM is deactivated and an InAsSb detector records the ring-down transient.

In this contribution we present our progress with the design as well as a performance evaluation with CO and CO₂ gas samples at room temperature and Pa-level pressure. Further we evaluate the effect of mechanical vibrations, as introduced by the operation of vacuum systems, on the performance of the optical cavity. Finally, we discuss how remotely adjustable optical mountings in our application improve laser safety.

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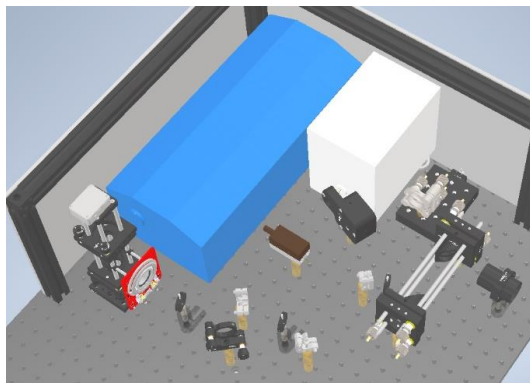


Fig. 1: Rendering of the setup before coupling into the cavity. The wavemeter (blue) tracks the spectral position of the attenuated EC-QCL (white). On triggering, an AOM (brown) directs the beam into the cavity (off right).

Back current shunts for electrical diagnostic of Nanosecond Pulsed Discharges

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1. Introduction

Nanosecond Pulsed Discharges are a promising method to convert CO₂ into value-added chemicals and fuels. The conversion via renewable electricity would allow storing it, recycling CO₂, and obtaining carbon-neutral synthetic fuels. The accurate estimation of the energy deposited in the plasma has a crucial role in evaluating the efficiency of the plasma-mediated processes.

For short high voltage (~ 40 kV) electrical pulses, the measurement of the voltage drop across the plasma reactor, necessary to calculate the energy dissipated by the plasma, is not trivial. Probes with a wide bandwidth (~ 100 MHz) are needed, but they are not commercially available [1].

Hence, custom made devices, like back current shunts (BCS), must be developed and characterized.

2. Back current shunts

BCSs allow measuring the electrical pulse in the middle of the transmission line connecting the generator to the load (i.e., the plasma reactor). They are built by making a small gap in the outer conducting braid of a coaxial cable and replacing it with resistive elements [1].

Measuring the voltage drop across the shunt, we can intercept the incident electric pulse followed by its reflections at the load and the generator due to impedance mismatches. The voltage signal U_{osc} measured at the oscilloscope is proportional to that in the central wire U_{HV} . According to the transmission line theory, the potential on the HV electrode, and so the voltage drop across the reactor, can be calculated as the sum of the incident and reflected wave potentials. If the transmission line is long enough, it is possible to separate the incident and the reflected pulses in the oscilloscope's signal and calculate their energy. The difference between the energy of the incident and reflected pulses in the transmission line is equal to the energy deposited in the plasma.

The resistors used to build the BCS must have very low stray capacitance and inductance; their frequency response must also be flat up to the desired frequency.

Furthermore, R_{BCS} must be much lower than Z_{cable} , and the cylindrical symmetry must be kept. We could obtain a BCS with these characteristics using 18 10Ω thick film SMD resistors in parallel. This BCS was calibrated using a VNA and tested with low-voltage nanosecond pulses. Its behavior at high frequencies is shown in Fig. 1. It turned out to be a good candidate for our applications.

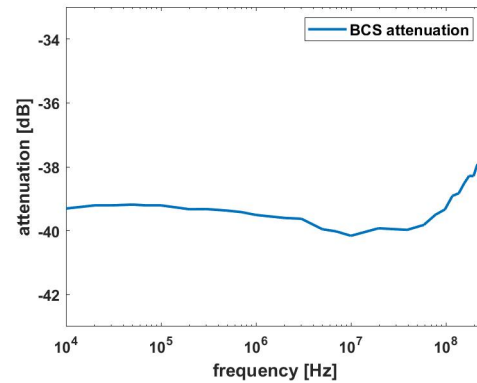


Fig. 1: BCS's frequency response.

In this contribution, a comparison between this technique and conventional probes will be presented, applied to a plasma reactor for Nanosecond Repetively Pulsed Discharges at atmospheric pressure in pin-to-pin configuration, similar to the one used for CO₂ splitting [2] and dry reforming of methane [3].

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Ion dynamics at the hollow cathode for abnormal argon glow discharges

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1. Introduction

Laser-Induced Fluorescence (LIF) measurements of the ArII $3d^2G_{9/2}$ metastable state in an abnormal glow discharge using a hollow cathode in argon provide new insight into ion dynamics near the hollow cathode. All experiments were performed within the same magnetic multipole chamber using argon discharges between pressures 0.1 and 10 mTorr sustained by electron emission from negatively biased tungsten filaments. LIF measurements were taken along the axis of symmetry inside and outside various hollow cathodes.

2. Results

A hollow cathode consisting of two parallel rings biased at -200 V with a common axis of symmetry shows diverging ion motion originating from the cathode center (Fig 1) [1]. The cathode demonstrates the hollow cathode effect with high ionization density between the two equipotential rings. This result is in disagreement with the commonly held view of predominant convergent ion motion in similar cathodes used to produce nuclear fusion from deuterium discharges (See [2] for example). Experimental results are used to argue that the spherical grid cathodes used to operate such fusion devices also operate as a hollow cathode discharge despite significantly more negative cathode biases.

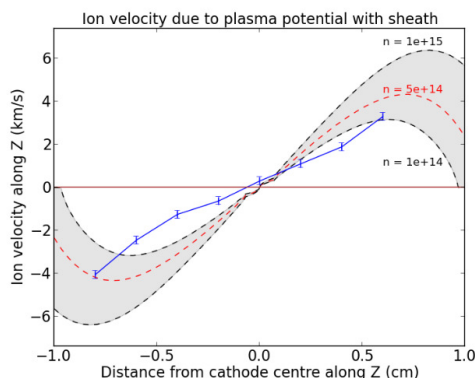


Fig. 1: LIF measurements of ArII ion velocity (blue line) as a function of axial distance from the cathode centre (z) in the two-ring hollow cathode, along with ion velocities predicted by a fluid discharge model (grey area)

Similar LIF measurements on the immediate exterior of a conventional hollow cathode biased between -200 and -1000 V indicate an anomalous increase of the metastable ArII ion density while approaching the cathode sheath (Fig 2). These results correspond with published experimental results [3] obtained from a floating plate within an abnormal argon discharge in the same experimental apparatus, yet they remain unexplained.

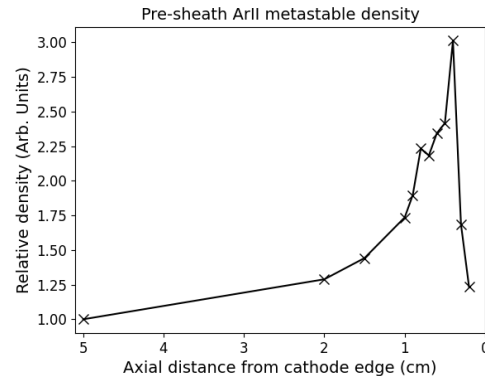


Fig. 2: Demonstrative LIF measurements of ArII $3s^2G_{9/2}$ metastable relative density along the axis of a conventional hollow cathode biased at -1000 V (edge at 0 cm). The ion density has been set to 1 in the bulk plasma.

The ion divergence phenomenon follows predictions from fluid discharge modeling of an ionic sheath whilst the increasing ion density could perhaps arise from the presence of a spatially dependent secondary electron distribution within the pre-sheath.

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CO₂ dissociation in plasma-assisted chemical looping process

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In view of GHG emissions mitigation, efficient methods for CO₂ splitting are necessary. The advantage of powering this process with renewable energy is twofold; the use of CO₂ as raw material and the prevention of CO₂ emissions from combusted fossil fuels. Among the different electrified technologies, non-thermal plasma offers an available technology in gas processing [1].

In CO₂ dissociation processes driven by non-thermal plasma, electron impact reactions enable the split of CO₂ molecule in CO and oxygen through dissociation and vibrational excitation reactions. In nanosecond pulsed discharges, for instance, CO₂ is readily dissociated in the plasma, leading to instantaneous conversions of 60%, which however drop significantly to ~12% within a few μ s during pulse intervals mainly due to CO/O recombination [2]. To increase the conversion level, hybrid plasma-catalyst systems are widely investigated. In classical plasma catalysis, the plasma intensifies mostly O extraction from the crystal lattice of a material that is placed inside the plasma zone. CO₂ molecules are adsorbed at the oxygen vacancies and dissociate into CO and O via dissociative electron attachment. [3]

We propose and demonstrate a different approach: the integration of a material for the chemical looping process. CO₂ is first dissociated inside plasma into CO/O at supra-equilibrium conversions (up to 60%) at bulk gas temperature 773 K and 403 kJ/mol energy cost. Then a nanostructured CeO₂/Fe₂O₃ oxygen scavenger, pre-reduced by H₂ plasma, downstream of the plasma zone, enables the capture of produced oxygen species, suppressing the CO/O recombination reaction. Supra-equilibrium CO₂ conversions are achieved at the reactor outlet.

A reactor based on a coaxial nanosecond pulsed discharge has been developed to investigate the aforementioned process. Electrical characterization of the discharge has been performed as in [1]. The outlet gases were analyzed by chromatographic techniques.

The nanostructured oxygen scavenger material, CeO₂/Fe₂O₃ 10:90 %_w, was synthesized according to the procedure described by Galvita et al. [4]. The material has been characterized in fixed bed experiment at the temperature of 623 K, 723 K and 773 K, after reduction with 5% H₂ in N₂. The material has been placed in the plasma reactor downstream of

the discharge, measuring the conversion for the plasma assisted chemical looping (PACL) configuration. The results for material-alone, plasma alone and PACL (indicated as average synergistic effect) are shown in Fig. 1. The PACL conversion is 29%, higher than the cumulative effects of the plasma-alone and material-alone. The maximum synergistic effect corresponds to the estimated conversion just after the end of the pulse inside the discharge streamer by spectroscopy.

Without plasma-material synergy, such average CO₂ conversion can only be attained at temperatures ≥ 2775 K, according to chemical equilibrium calculations. This concept of plasma-assisted chemical looping allows reaching threefold higher conversions than state-of-the-art plasma technologies.

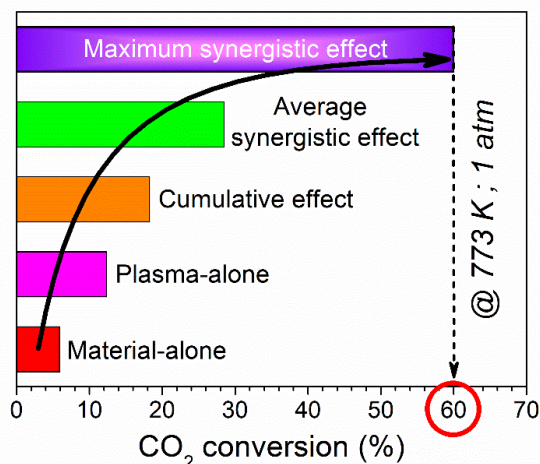


Fig. 1: the measured CO₂ conversion for the material-alone, the plasma-alone and the PACL reactor.

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Langmuir probe measurements in a dual-frequency capacitively coupled rf discharge

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The standard (industrial) frequency for common rf plasmas used in technology is 13.56 MHz. A difference in the area of the electrodes and the difference in the electron and ion fluxes due to their mobility in respect to the excitation frequency, result in a negative dc potential, the self-bias voltage, at the powered electrode. The gas pressure in the device and the dc self-bias mainly determine the plasma sheath extent as well as the potential and, thus, the ion current density and the ion energy towards the electrode surface. An independent control of these important properties - especially in industrial applications - is desirable but usually not possible. By adding a second frequency (1st harmonic, 27.56 MHz), a so-called electrical asymmetry effect (EAE) is created, which enables the control of the bias voltage and, thus, the ion energy almost independent of the ion flux by varying the phase angle between the two harmonics, while other discharge parameters, e.g. gas pressure, stay constant [1]. Since the EAE is a relatively new approach to separately control these two parameters, the number of diagnostics performed in such a (2f)-discharge amounts to a minimum.

Regarding this, measurements with optically trapped microparticles in an optical tweezer have been carried out, which provided a spatially resolved force distribution of the electric field force of the sheath region by moving the microprobe through the plasma (Fig. 1), both in a dual- and a single-frequency discharge [2].

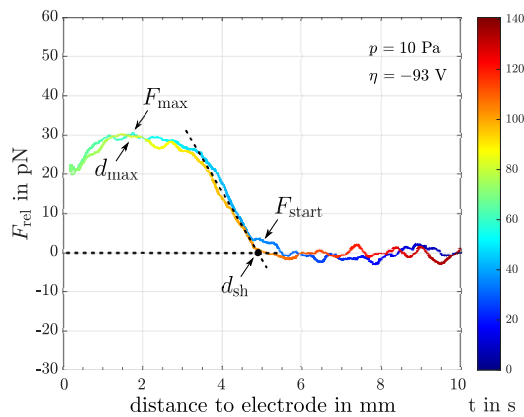


Fig. 1: Measured force profile obtained by the use of an optically trapped microparticle for a certain pressure p and a specific dc self-bias η in a 2f discharge. The color

gradient illustrates the time required for the measurement. The surface of the electrode is at 0 mm. The points d_{sh} and F_{start} mark the position of the sheath edge. F_{max} and d_{max} indicate the value and the position of the force maximum [2].

Based on these profiles, clear differences of the marked parameters d_{sh} , F_{max} and d_{max} could be identified between the three frequency regimes (Fig. 2).

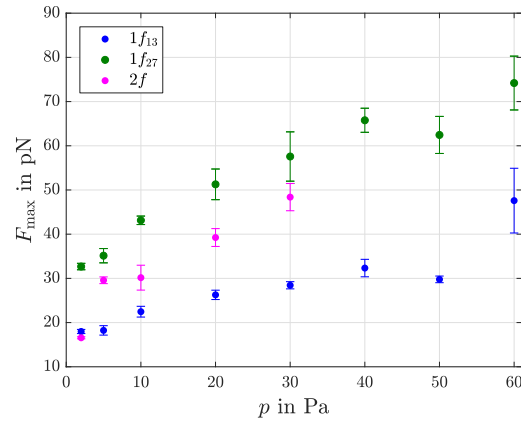


Fig. 2: Value of the force maximum F_{max} derived from the force profiles as a function of the pressure p for three different frequency regimes ($1f_{13}$: 13.56 MHz and $1f_{27}$: 27.12 MHz) [2].

By using a specially designed Langmuir probe in this dual-frequency (2f-) plasma, it can be determined to what extent the important plasma parameters, i.e. electron density and electron temperature, change with a variation of the phase between the two harmonics. This work aims to provide an initial insight into the differences between a single- and dual-frequency plasma based on Langmuir probe measurements and therefore provide indications of possible causes of the differences in the measured force profiles between these frequency regimes, as well as offer a comparison of theory and experiment.

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Absolute oxygen atom density in a DC discharge in pure O₂ : 2: Two-photon absorption Laser-induced Fluorescence

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1. Introduction

Accurate measurements of the absolute density of reactive atoms are essential for the validation of models of plasmas in molecular gases. However, even for such a ubiquitous species as oxygen atoms, few attempts have been made to compare the results from different measurement techniques. Two-photon absorption Laser-induced Fluorescence (TALIF), with the calibration scheme proposed by Niemi et al. [1], is widely used. However, high absolute accuracy (better than a factor two in either direction) is difficult to achieve due to the many complex steps in the calibration, and the fluctuations inherent in a non-linear technique. Above all, the absolute densities obtained depend upon the ratio of the O atom to the Xe two-photon absorption cross-section, measured only in the paper by Niemi. More recently, Drag et al. [2] directly measured the two-photon absorption cross-section for the Xe transition used, and found a value of about one half of that implied by the Niemi measurement. This would imply (assuming that the oxygen cross-section calculated by Saxon and Eichler [3] is correct), that the oxygen atom densities measured by TALIF should be revised downwards by this factor. In order to determine the accuracy of the cross-section ratio, we have performed a careful comparison of TALIF measurements with cavity ringdown measurements (presented separately), using the forbidden $^3P_2 \rightarrow ^1D_2$ transition at 630 nm [4]. Measurements were made in the positive column of a DC glow discharge in pure oxygen, which provides a uniform discharge over 50 cm.

2. Experimental results

The O atoms were excited by an unfocused 225.5 nm laser pulse, and fluorescence signal was detected by the photo-multiplier (PMT). The number density of O atoms at ground state was obtained by:

$$n_O = \frac{\sigma_{Xe}^{(2)}}{\sigma_O^{(2)}} \cdot \frac{v_O^2}{v_{Xe}^2} \cdot \frac{P_{Xe}}{P_O} \cdot \frac{F_{Xe}}{F_O} \cdot \frac{A_{Xe}/(A_{Xe}+Q_{Xe})}{A_O/(A_O+Q_O)} \cdot \frac{S_O/I_O^2}{S_{Xe}/I_{Xe}^2} \cdot \frac{1}{\eta} \cdot n_{Xe} \quad (1)$$

where the quantum efficiency P for PMT and filter transmission F was taken from the literature; the emission coefficient, quenching rate and fluorescence integral were calculated from the time/wavelength resolved TALIF spectrum as shown in Figure 1.

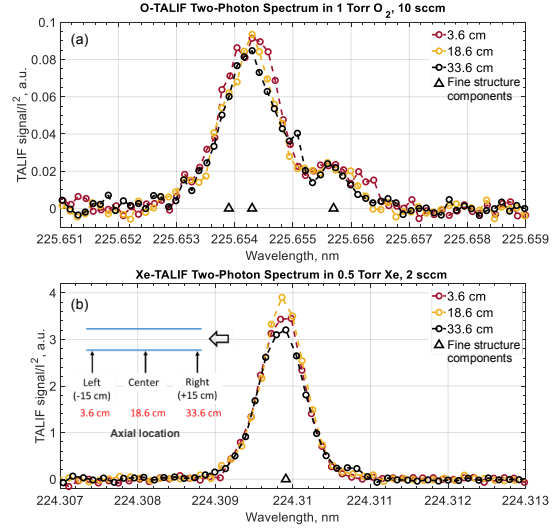


Fig. 1: (a) TALIF spectrum in 1 Torr, 40 mA O₂ discharge stabilized after 4 hours under wall temperature of 20 °C (b) TALIF spectrum in 0.5 Torr Xe under 20 °C

The TALIF measurements indicate a central O atom number density of $1.7 \times 10^{15} \text{ cm}^{-3}$, compared to $(3.5 \pm 0.2) \times 10^{15} \text{ cm}^{-3}$ by CRDS (at 4 hours, allowing for the axial profile), implying that the Niemi cross-section ratio is in fact too small by a factor of about 2.

Acknowledgements

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Microwave interferometry as a tool for electron density estimation in atmospheric CO₂ microwave plasma

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Chemical storage of renewable energy by means of efficient conversion of stable CO₂ and CH₄ molecules is a perspective route for the mitigation of green-house emissions and closing the carbon cycle. Among the variety of Power-to-X technologies, gas conversion technologies using plasmas show high potential for the efficient use of intermittent renewable energies. To date, the highest efficiency (above 80 %) of the plasma assisted conversion of CO₂ into CO is reported in experiments with microwave-sustained plasmas [1].

Particular for CO₂ atmospheric plasmas both Langmuir probes and Thompson scattering diagnostics are not well applicable. The former one, because of gas temperature ranging up to 7000 K, and for the latter one, because of the strong Rayleigh scattering signal that dominates the Thompson scattering signal. In this regard, microwave interferometry could become the alternative tool for the measurement of electron density in compact atmospheric plasmas. This method is well known from larger plasma experiments, e. g. it is used in fusion plasmas (tokamaks and stellarators) where the interferometer is one of the standard diagnostics [2].

In the present work, the applicability of microwave interferometry to atmospheric plasma sustained in the surfaguide microwave reactor [3], in a 16 mm inner diameter glass tube is investigated. The interferometer features two elliptical focusing mirrors and pyramidal gain horn antennas and operates from 47 GHz up to 77 GHz. A specific challenge is the presence of parasitic interferences due to wave propagation through the glass tube and the standing wave effects. To get information about the refractive index of the plasma from the transmitted signal, modelling of electromagnetic wave propagation is inevitable. It is found that the 3D problem can be well approximated with 2D problem considered in a plane of antennae and glass tube axes. Geometry of problem approximated to 2D dimension in Tamic RτH full-wave code [4] is shown in Fig. 1. Note, in such 2D approximation only single polarization of e/m wave is taken into account and scattering effects at glass tube surfaces out of plane of consideration are neglected. The validation of this approximation was proven by comparing the results obtained with Tamic-RτH 2D full-wave code against 3D model in CST Studio Suite [5]. It is found that

with solving the inverse problem based on the 2D full-wave modelling with Tamic-RτH (Fig. 1), the correct interpretation of electromagnetic wave signals transmitted through the plasma in terms of plasma electron density is possible. The method and results will be presented and discussed for selected plasma scenarios with pure CO₂ plasma as well as with admixture of argon gas in different concentrations.

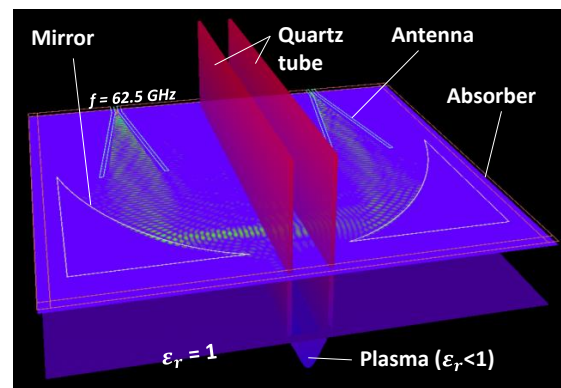


Fig. 1: Full wave 2D model in Tamic-RτH solver. Boundary conditions: ideal conductor (antennae and mirrors), dielectric with $\epsilon_r=3.75$ (quartz tube), plasma permittivity is approximated for Gaussian profile as $n_e=n_0 \cdot \exp(-(x/\Delta x)^2)$. Microwave electric field distribution shown for $n_0=3E19 \text{ cm}^{-3}$, $\Delta x=7 \text{ mm}$ and excitation frequency of 62.5 GHz.

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Mueller Polarimetric Imaging as a new tool for detecting the effect of Non-Thermal Plasma treatment on the skin

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Abstract

Non-Thermal Plasma (NTP) is a promising technique studied for several medical applications such as wound healing or tumor reduction. The detection of microstructural variations in the skin is currently performed by histological methods, which are time-consuming and invasive. This study aims to show that Mueller polarimetric imaging is suitable for fast and non-intrusive detection of skin microstructure modifications induced by high-power NTP treatment [1].

In this study, pig skin samples are treated by a He plasma jet generated by 10 kHz AC voltage in a glass capillary. Mueller polarimetric imaging is performed before and after NTP treatment with different power/dose. The results show that, with plasma power higher than 1 W and more than 1 min treatment time, the thermal contribution of NTP-treatment may play a role in modifying tissue polarimetric properties. The thermal effects of NTP-treatment, cause an increase of the tissue anisotropy. In particular, the total depolarization and linear birefringence are the most important parameters for quantifying and monitoring skin microscopic modifications induced by thermal effects. Figure 1 shows that NTP treatment with plasma power of 1.1W for up to 4min induces a ring shape area where the linear birefringence increases and the total depolarization decreases. In the center of the ring shape area, the linear birefringence decreases and the total depolarization increases, and both parameters in the center become more homogeneous. Such change of the polarimetric parameters is possibly related to the structure modification of collagens. NTP treatment with lower power (200 mW) /dose, shows no change to the total depolarization and linear birefringence, thus has possibly no damage to the microstructure of pig skin samples. This is consistent with recent publication showing *in-vivo* wound healing properties of a 10 – 100 mW plasma jet operating at 50 Hz [2].

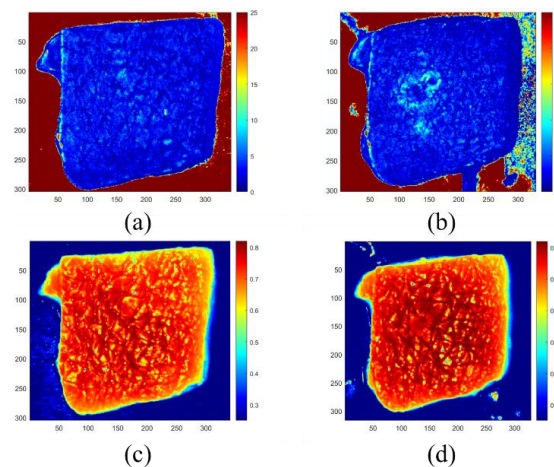


Fig. 1: (a) linear birefringence of the pig skin before treatment. (b) linear birefringence of the pig skin after treatment. (c) total depolarization of the pig skin before the treatment. (d) total depolarization of the pig skin after the treatment. The skin is treated by plasma jet with power of 1.1W and treatment time of 4min. The treated area is heated by the plasma to 59°C

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