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The dynamics of radio-frequency driven atmospheric pressure plasma jets

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Abstract. The complex dynamics of radio-frequency driven atmospheric pressure plasma jets is investigated using various optical diagnostic techniques and numerical simulations. Absolute number densities of ground state at omic oxygen radicals in the plasma effluent are measured by two-photon absorption laser induced fluorescence spectroscopy (TALIF). Spatial profiles are compared with (vacuum) ultra-violet radiation from excited states of atomic oxygen and molecular oxygen, respectively. The excitation and ionization dynamics in the plasma core are dominated by electron impact and observed by space and phase resolved optical emission spectroscopy (PROES). The electron dynamics is governed through the motion of the plasma boundary sheaths in front of the electrodes as illustrated in numerical simulations using a hybrid code based on fluid equations and kinetic treatment of electrons.

1. Introduction

Non-thermal non-equilibrium atmospheric pressure plasmas are commonly characterized by reduced spatial dimensions of the confining structures, e.g. electrodes, stabilizing the discharge and preventing the transition to a ‘thermal’ discharge. Typical dimensions vary from a few micrometres up to a few millimetres. The wide range of these discharges is, therefore, often sub-summarized under the topic ‘microplasmas’ [1–5].

High concentrations of atoms in the order of $10^{15}$ cm$^{-3}$ and a low gas temperature significantly below 100°C, suitable for many applications especially in localized modifications of sensitive surfaces, can be provided by jet-like devices without the requirement of complicated and expensive vacuum systems [6]. Many of these discharges operate in a mixture of a noble gas, preferably helium, and a small molecular component selected depending on the envisaged application [7]. Despite enormous potential for technological applications, the fundamentals of these stable homogeneous non-equilibrium plasmas at ambient pressure are only poorly understood. This is mainly due to the complexity of these discharges composed of electrodes in the close vicinity of the confining walls and a mixture of neutral and charged atomic and molecular components. Furthermore, radiation can play an important role in the discharge balance. These jets are typically divided into the core plasma where plasma processes are driven and the effluent with reactive components suitable for technological applications.

The discharge concept adopted here is the atmospheric pressure plasma jet (APPJ) introduced by Selwyn and co-workers [8] in 1998. This technically relatively simple capacitively coupled device is
typically operated at an excitation frequency of 13.56 MHz and an electrode spacing of about 1 mm. A mixture of a noble gas as the base gas and a small percentage addition of a molecular component serves as the operating gas. The molecular admixture is limited by instabilities in the discharge. At typical gas velocities of some 10 ms$^{-1}$ the atomic oxygen concentration in the effluent of an APPJ is in the order of n$_O$ $\sim$ 10$^{15}$ cm$^{-3}$ [9]. These reactive oxygen species are a prominent candidate for the surface modifying properties of the jet [10, 11].

Figure 1 shows a photograph of a typical coaxial APPJ illustrating the potential for technological applications of the effluent at comparatively low gas temperature. The front view picture shows the geometrical design of the coaxial APPJ with an inner radio-frequency driven rod-electrode and an outer grounded electrode.

![Figure 1: Photograph of a coaxial APPJ. The side view illustrates the potential for technological applications of the effluent at low gas temperature. The front view shows the geometrical design with an rf driven inner rod and an outer grounded electrode.](image)

Figure 2 shows a photograph illustrating the capability for localised surface modifications using APPJs. A simple example is the observable significant change in wettability of a treated Petri-dish. Efficient exploitation, further development, and tailoring towards future high benefit technological applications require detailed understanding of the dynamics of APPJs. This includes the complex dynamics of plasma production and energy carrying species in the plasma core. Crucial aspects are energy transport mechanisms from the core plasma to the effluent, in particular in the vicinity of their interface region.

![Figure 2: Photograph illustrating the change in wettability of a locally treated Petri-dish.](image)
The sketch of figure 3 shows the constituents determining the dynamics of energy dissipation in the core plasma and energy transport mechanisms to the effluent region. The plasma chemistry inside the discharge volume is strongly influenced by the dynamics of the plasma boundary sheaths in front of the electrodes and corresponding surface processes. The plasma boundary sheaths energize electrons which dominate plasma ionization and the production of electronically excited particles through electron impact. Subsequent Penning ionization and three body collisions, due to the high pressure collisional environment, also play important roles in the particle and power balance of the core plasma. Energy transport mechanisms to the effluent region include transport of radicals, excited particles, and radiation. It has been shown that the effluent region is practically free of charged particles [8].

Figure 3: Sketch of the components determining the dynamics in the plasma core and energy transport mechanisms from the core to the effluent region. The wall represents a surface to be treated in technological applications.

2. Measurements in the effluent region

Ground state atomic oxygen radicals and (V)UV radiation play key roles in energy transport mechanisms in the effluent region and are also crucial energy carriers for technological surface modifications. Measurements in the effluent region of a planar APPJ are discussed in the following.

Advantages of the planar configuration over the coaxial design shown in figure 1 are better diagnostics access to the discharge core and the simpler geometry allows better comparison with modeling and simulation results. A detailed description of the planar APPJ can be found in S Reuter et al. [12]. The two parallel stainless steel electrodes are 80 mm in length and 40 mm in width. The electrode gap is 1.1 mm. One electrode is connected to a 13.56 MHz radio-frequency power supply, while the other is grounded. The equal area electrode design results in symmetric plasma operation with corresponding plasma boundary sheaths in front of both electrodes. The APPJ is operated with helium feed gas and small oxygen admixtures.

The atomic oxygen ground state density is measured by TALIF spectroscopy. Tunable UV laser radiation is used to excite oxygen atoms by simultaneous absorption of two UV photons. The fluorescence radiation emitted when the atoms revert to an energetically lower state is measured in order to gain information about the atomic oxygen density. The measurements are calibrated with TALIF measurements on xenon, according to earlier investigations on the concentric APPJ [9, 13]. Collisional quenching, temperature effects and vignetting of the signal are essential aspects to consider in the analysis of the measurements [9, 12].
Figure 4 shows the atomic oxygen ground state density distribution in the effluent of the planar APPJ for a helium flux of 2 m³ h⁻¹, 0.5 vol% O₂ admixture and an RF power of 150 W (reading at the power supply). The effluent stays confined over several centimetres. The initial atomic oxygen ground state density directly at the APPJ’s nozzle is as high as \(~1 \times 10^{16}\) cm⁻³. Due to the planar APPJ’s simple geometry, these measurement results close to the nozzle are more precise than earlier measurements on the concentric version of the APPJ [9, 13]. Furthermore, the TALIF measurements show that at 10 cm distance from the nozzle still \(~1\)% of the initial atomic oxygen density remains.

![Figure 4: 2-dimensional spatial map of absolute densities of ground state atomic oxygen in the effluent region of a planar APPJ. The measurements are carried out in a controlled helium/oxygen environment using absolutely calibrated two-photon absorption laser induced fluorescence spectroscopy (TALIF).](image)

VUV radiation in the effluent is measured using a 0.2 m monochromator (Minuteman Laboratories Inc. 302VM, grating 1200 mm⁻¹) and a solar blind photomultiplier with a CsTe-photocathode (EMI G-26H315, MgF₂ window, spectral response 110 to 340 nm). The main spectral features in the VUV spectrum are the Schumann–Runge bands of O₂ and the atomic oxygen line at \(\lambda = 130\) nm.

Figure 5 shows that considerable VUV and UV radiation - highest at the jet’s nozzle - can be detected even at several centimetres distance from the nozzle. An atomic oxygen line at \(\lambda = 130\) nm and an O₂ emission at \(\lambda = 181\) nm are presented. (V)UV radiation measured off axis is below the detection limit of the measurement setup. It can be concluded that the major fraction of the (V)UV radiation originates from the jet’s discharge region and is not produced in the effluent. The radiation is not absorbed by the surrounding air, because the jet emits a plume of helium (and molecular oxygen), in which VUV radiation can be transported. The differing progressions of the two emission peaks’ intensities result from the difference in absorption by the molecular oxygen [14].
3. Measurements in the plasma core and the interface region

Measurements of the plasma dynamics in the core region are carried out in the so-called μ-APPJ. The μ-APPJ is an especially designed microscale version of the APPJ providing excellent access for optical diagnostics, in particular to the core plasma. Details of the design can be found in references [13], [15], and [16]. The reduced discharge dimensions (1x1x30 mm$^3$) significantly reduce the required power input (5 – 20 W) and gas flow (1 slm) keeping the gas velocity in the same range of some 10 ms$^{-1}$. Figure 6 is a photograph of the μ-APPJ in operation. Optical emission is observable from the core plasma, interface region, and effluent region. The red square indicates the region investigated by space and phase resolved optical emission spectroscopy (PROES).

PROES provides deep insight into the ionization and excitation dynamics of radio-frequency plasmas with excellent spatial and temporal resolution - down to microns [17] and nanoseconds [18]. A typical PROES experiment is described in reference [19]. Figure 7 displays spectrally integrated (ICCD sensitivity range: 300 – 800 nm) 2-dimensional spatially resolved images at different phases of the radio-frequency cycle. The images exhibit a pronounced dynamics - particularly in the plasma core. A symmetric plasma boundary sheaths dynamics, typical for capacitively coupled plasmas with equal area electrodes, is clearly visible. Interesting structures are observable in the interface region to
the effluent. These structures may be explained by enhanced electric fields at the edges of the electrodes.

![Figure 7: 2d-PROES measurements on the µ-APPJ. The images at different phases exhibit pronounced dynamics during the rf-cycle. The plasma is geometrically symmetric with plasma boundary sheath dynamics in front of both electrodes.](image)

Details of the excitation and ionization dynamics in the plasma core are shown in figure 8. From this spatio-temporal map conclusions can be drawn on the electron kinetics within the discharge between the electrodes. A first observation is the excellent symmetry of the discharge showing three pairs of emission features (indexed as 1–3) of equal intensity and distance to the electrodes. The respective emission at the opposite electrode is displaced by half of the period (~37 ns). The structures 1 and 2 in the symmetric half-phases of the cycle can be attributed to the emission induced by electrons accelerated by the expanding and collapsing sheaths. They are typical for a volume ionization-based α-mode discharge. The sheath collapse is also correlated with an electric field reversal facilitated by the reduced mobility of electrons in the collision dominated environment of atmospheric pressure plasmas. Feature 3 can be attributed to electron acceleration and multiplication within the high voltage plasma boundary sheaths. Initial electrons starting this mechanism in the sheath regions can be gamma-electrons from the electrode surfaces or electrons created through Penning ionization.

![Figure 8: Excitation and ionization dynamics in the plasma core of the µ-APPJ. Energetic electrons are observable during sheath expansion (1), sheath collapse (2), and maximum sheath expansion (3). The intensity rises from bright to dark colours.](image)
4. Numerical simulation of the electron dynamics

Numerical computer simulations can provide more detailed insight into the plasma dynamics, in particular the electron and plasma boundary sheath dynamics. Here we present a one-dimensional hybrid model across the discharge gap of the µ-APPJ with rf-excitation using the finite element method. The electrons are described kinetically and all other plasma species (ions, neutrals, excited particles) hydro-dynamically. The transport coefficients for electrons and chemical reaction rates involving electrons are calculated in advance using the Boltzmann solver BOLSIG+ [20] and stored in look-up tables depending on the mean electron energy. The governing fluid equations are Poisson’s equation, the continuity equations of the considered species in the drift-diffusion approximation and the electron energy equation. The numerical simulations are done for Helium (He) with an added impurity of 0.1% Nitrogen (N$_2$). The species e, He, He$,^+$, He$_2$*, He$_2$*, N$_2$, and N$_2$* are taken into account. He and N$_2$ are treated as constant background gases, all other species densities are determined using a set of chemical reactions equivalent to the one in reference [21].

The current voltage characteristics of atmospheric pressure glow discharges exhibit multiple solutions for a given voltage in the transition region from α-to-γ-mode operation [22]. Therefore, often the external current is given in numerical simulation rather than the voltage as under experimental conditions. By specifying the power rather than the current it is still possible to simulate a discharge with an externally applied voltage. This can be realised using a feedback control loop for the power coupled into the plasma [23].

Figure 9 shows a typical space and phase resolved map of the electron density in the µ-APPJ. The voltage amplitude is 245 V and the maximum electron density is 1.16 $10^{17}$ m$^{-3}$. The symmetric character of the plasma boundary sheaths in front of both electrodes is clearly visible. The rapid movement of the sheath edges energises electrons during sheath expansion and sheath contraction - as observed in the PROES measurements.

Figure 9: Numerical simulation of the electron dynamics in the discharge gap of the µ-APPJ operated in helium with a nitrogen impurity of 0.1%. The voltage amplitude is 245 V and the maximum electron density is 1.16 $10^{17}$ m$^{-3}$. Red colour indicates high and blue low electron densities, respectively. The symmetric character of the plasma boundary sheaths in front of both electrodes is clearly visible.
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