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## Fast Track Communication

# Atmospheric pressure streamer follows the turbulent argon air boundary in a MHz argon plasma jet investigated by OH-tracer PLIF spectroscopy

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## Abstract

An open question in the research of the dynamics of non-equilibrium cold atmospheric pressure plasma jets is the influence of ambient species on streamer propagation pathways. In the present work, by means of planar laser-induced fluorescence (PLIF), an atmospheric pressure argon plasma jet is investigated in a laminar and turbulent gas flow regime. The flow pattern is investigated with plasma on and plasma off. It is shown that in turbulent mode, the streamer path changes according to the flow pattern changes and the resulting changes in air abundance. From a comparison of an analytical diffusion calculation and LIF measurements, the air impurity boundary is determined. Most importantly, the origin of the streamer pathway is investigated in detail, by recording the flow pattern from OH-PLIF measurements and simultaneously measuring the streamer path by an overlay technique through emission measurements. It is shown that the streamer path is correlated to the turbulent flow pattern.

Keywords: atmospheric pressure plasma jet, planar laser-induced fluorescence, discharge propagation, plasma medicine, plasma jet effluent regime

## 1. Introduction

Only a few years ago, so-called plasma bullet [1, 2] and guided-streamer [3] jets came to be known. Recently, researchers have identified the fundamental processes behind these dynamics (see e.g. [4, 5]). However, their propagation in air has continued to raise questions. In this work, the connection of gas flux regime, ambient species and streamer head propagation in a MHz atmospheric pressure argon plasma jet is investigated by laser spectroscopic methods. The diagnostic

technique is based on planar laser-induced fluorescence (PLIF) on hydroxide molecules (OH) generated by the plasma jet itself due to water impurities. With stereoscopic imaging, both flow structure and streamer propagation are investigated simultaneously for a single discharge event. The study reveals that the flux field with plasma differs strongly from the flux field without plasma. Furthermore, it is most prominently and for the first time shown that the streamer propagates in the noble gas channel of a turbulent plasma jet effluent itself and follows the boundaries of ambient species impurities. Especially

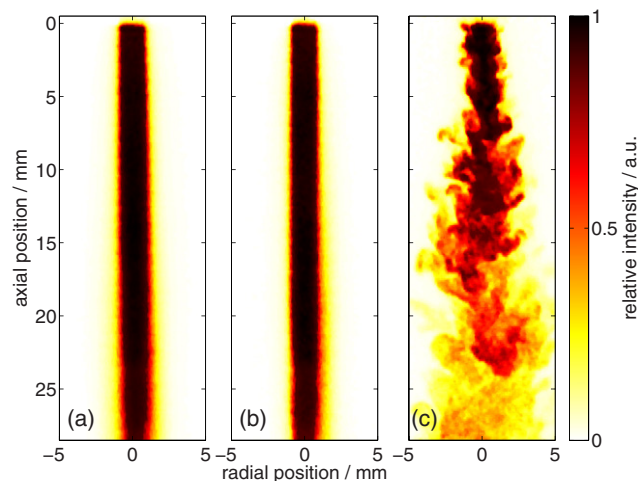
revealing are the differences observed for the laminar and turbulent case.

## 2. Experimental setup

In this work, the flux and streamer propagation of a MHz atmospheric pressure plasma jet (kINPen, neoplas tools GmbH, Germany) [6, 7] have been investigated. The feed gas is argon (Ar purity 99.999%), which may contain water impurities of up to 20 ppm originating from the gas pipe as reported in [8]. The OH PLIF measurements are performed with a frequency doubled dye laser (Cobra Stretch, Sirah Laser und Plasmatechnik GmbH, Germany) pumped with a 10 Hz pulsed Nd:Yag laser (Spectra-Physics, Inc.). The pulse duration is measured to be 8.1 ns after frequency doubling at 283 nm. The laser linewidth has been recently measured on a similar dye laser from the same company to be 1.4 pm [9]. Although this parameter remains crucial for the determination of absolute density [10], it does not influence qualitative measurements. The P1(4) transition of the OH(A, X)(1,0) at 283.5 nm is chosen as the energy transition due to its good absorption efficiency. Additionally, it is well separated from other lines. The linearity of the OH fluorescence signal versus the laser energy has been checked. The laser beam is expanded to a sheet via a cylindrical quartz lens and guided through the plasma jet effluent. The laser energy integrated along the 12 mm high sheet is measured to be 13 mJ. An aperture is mounted in front of the jet in order to illuminate only the area of interest. A CCD camera Imager Pro X (LaVision GmbH, Germany), combined with an external gated image intensifier and a 100 mm f/2.8 UV lens, is used as detection system. The fluorescence emission is observed at a 90° angle to the p-polarized laser beam in order to reduce unwanted signals due to Rayleigh scattering. Additionally, a stereoscopic optical imaging system is coupled to the lens in order to record two images with each laser shot. By installing different filters for the two images respectively, different information can be gathered simultaneously. Here, on the first optical input, a BK7 neutral density filter is mounted, which allows observation from 350 to 1100 nm, where most of the plasma emission is found due to argon lines [8, 11]. On the second optical input, a 10 nm FWHM band-pass filter centered at 313 nm is used to collect the fluorescence only of OH. Both signals are recorded on a vertical split CCD chip, so that the exposure time is identical for both signals.

The plasma jet frequency is phase locked with the laser pulse as well as the detection system, which allows phase-resolved optical emission spectroscopy (PR-OES). The exposure time is set to 200 ns, which is sufficient to obtain enough intensity for single shot acquisition. The optical aberrations such as distortion produced by the stereoscopic imaging system are corrected by Davis software [12] after the calibration procedure described in the manual [13]. The inhomogeneity of the laser sheet is measured by Rayleigh scattering and corrected by intensity normalization.

The observation of the flow pattern with plasma off is performed by PLIF using acetone as a tracer. As described in [14], acetone fluorescence is a suitable tracer to image

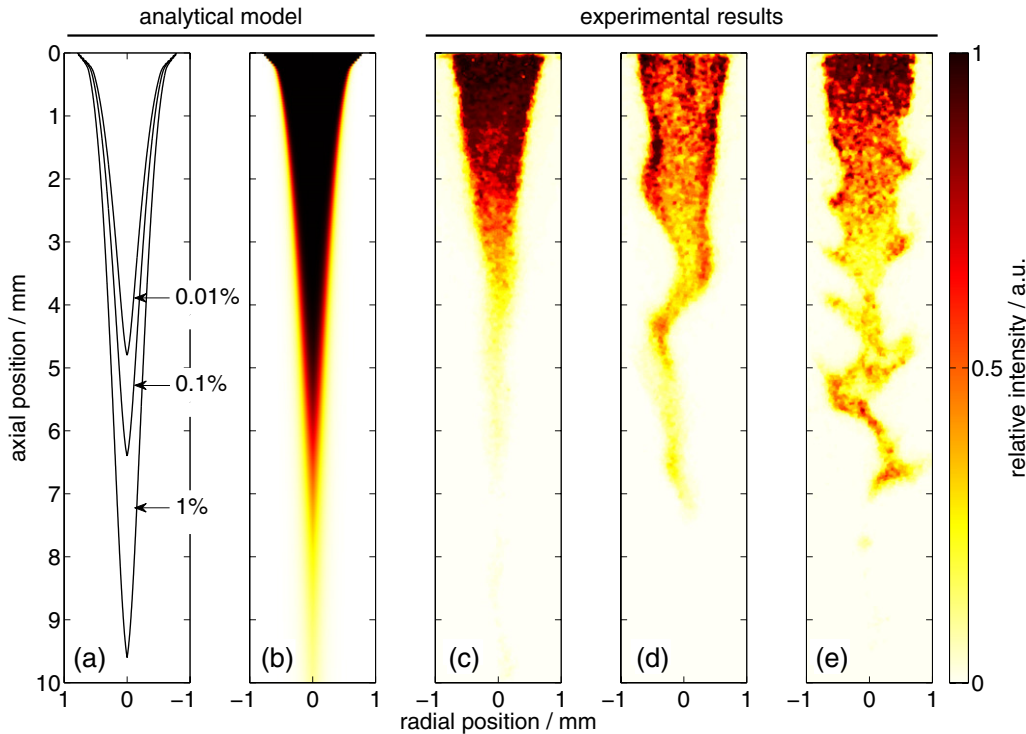


**Figure 1.** Space resolved single shot fluorescence signal of acetone for different argon gas flow rates of (a) 0.5 slm/ $Re=465$ , (b) 1.0 slm/ $Re=930$  and (c) 3.0 slm/ $Re=2790$ . The jet nozzle is located at an axial distance of zero. The plasma was switched off.

turbulent flows. During the measurements, the feed gas is bubbled through acetone into the plasma device. The acetone molecules are excited by the laser beam at 283.5 nm, and the fluorescence from 350 to 800 nm is collected by an intensified CCD camera and recorded. Observing the flow pattern with plasma-on requires the use of a different tracer, since acetone would be destroyed by the reactive species in the plasma. In the past, hydroxyl fluorescence was already used in combustion with good success on turbulence studies [15]. Moreover, one report stated that the hydroxyl molecule lifetime is 1 ms to a few milliseconds at atmospheric conditions [16, 17]. The hydroxyl radical is produced by the plasma itself from water impurities within the feed gas [8], meaning that the plasma generates the tracer itself and no external species need to be admixed for the flow measurements. Recently, hydroxyl molecule densities on-axis were measured by UV absorption on the same discharge [18]. The results show that the density remains constant up to a 7 mm distance from the nozzle. It can thus be assumed that the hydroxyl density does not decay significantly along the first 10 mm from the nozzle [18]. For these reasons, the hydroxyl radical, for the present studies, can be used as a fluorescent tracer.

## 3. LIF on acetone—plasma off measurements

Figure 1 shows the fluorescence of acetone for three different feed gas flow rates. As can be seen, at both 0.5 and 1 standard liter per minute (slm) the flow is laminar, while at 3 slm the flow is turbulent. The indicated Reynolds numbers  $Re = 2r_0v_{av}/\nu$  are defined via the capillary radius  $r_0$  and the kinematic viscosity  $\nu$ . The critical Reynolds number ( $Re$ ) for the transition from the laminar to turbulent regime in pipe flows is  $Re_C = 2300$ , which is consistent with the observed flow regimes.



**Figure 2.** Analytical estimation of the ambient species density (a) and the resulting analytical OH emission pattern (b). Space resolved fluorescence signal of hydroxyl for different argon gas flow rates of (c) 0.5 slm, (d) 1.0 slm and (e) 3.0 slm. The jet nozzle is located at an axial distance of zero. The plasma was switched on. Single shot acquisition.

#### 4. OH fluorescence and quenching by ambient air—plasma on measurements

The radiative lifetime of the OH(A, X)(1,0) is known to be around  $\tau = A^{-1} = 748$  ns, where  $A$  is the Einstein coefficient for spontaneous emission. The emission is strongly quenched by atmospheric species [18]: the combined quenching rate ( $Q$ ) and vibrational energy transfer (VET) coefficient of OH(A, X)(1,0) by argon ( $0.27 \times 10^{-11} \text{ cm}^3 \text{ s}^{-1}$  [19]) is two orders of magnitude lower than for nitrogen ( $k_Q^{\text{N}_2} = 23.3 \times 10^{-11} \text{ cm}^3 \text{ s}^{-1}$  [20]), oxygen ( $k_Q^{\text{O}_2} = 20.6 \times 10^{-11} \text{ cm}^3 \text{ s}^{-1}$  [20]) and water ( $k_Q^{\text{H}_2\text{O}} = 66 \times 10^{-11} \text{ cm}^3 \text{ s}^{-1}$  [20]). Despite that, the quenching rate of OH fluorescence by water is in the same order of magnitude as dry air, and ambient humidity has a negligible effect on the OH fluorescence quenching due to a much lower density compared to air. In consequence, OH(A, X)(1,0) has a longer lifetime when it is mixed with argon than in presence of air. The OH originates dominantly from the dissociation of water impurities in the feed gas. Even at what is considered to be dry feed gas conditions, the quantity of water molecules lies in the order of some 10 ppm. When the gas leaves the jet, the OH concentration is distributed relatively homogeneously along the diameter of the nozzle (see figure 2(c)). Similar observations have been reported earlier. Here the main hydroxyl production is localized near the powered electrode in case of water impurities in the feed gas [21]. Additionally, the hydroxyl density remains presumably constant until several mm distance [18]. The spatial distribution can be expected to resemble the acetone distribution as shown above (see figure 1). From these as well

as the quencher considerations, it can be safely assumed that the LIF signal reveals rather the influence of the quenching by the ambient species than the ground state OH molecule distribution. The measured data therefore reflects the argon flow pattern and the argon air boundary shape, as will be described in the following.

The diffusion of ambient species into the effluent of the plasma jet can be estimated by solving the convection diffusion equation

$$v(r)\partial_z n_A + D\Delta_r n_A = 0$$

with the boundary condition  $n_A = \theta(r - r_0)n_A^0$  for the ambient species density, where  $\theta(x)$  is the Heaviside step function  $\theta(x) = 0$  for  $x < 0$ ,  $\theta(x) = 1$  for  $x \geq 0$ . Here,  $D$  is the diffusion coefficient and  $v(r)$  a velocity field with a radial dependence and  $\Delta_r$  the radial part of the Laplace operator. The solution in the case of a constant velocity profile is given in [22] and can be evaluated using an infinite series. For a laminar jet flow, the velocity field in the near field of the jet can be approximated by a Poiseuille flow profile  $v(r) = 2v_{\text{av}}(1 - r^2/r_0^2)$ . In order to account for this flow profile, non-dispersive path mapping [23] can be applied, mapping the constant velocity solution according to the flow field. The contour lines of the resulting ambient air density  $n_A/n_A^0$  for an average velocity of  $v_{\text{av}} = 4.1 \text{ m s}^{-1}$  and diffusion coefficient  $D = 0.2 \text{ cm}^2 \text{ s}^{-1}$  are shown in figure 2(a). Assuming a constant OH-density in the jet's effluent, the fluorescence intensity of OH(A, X)(1,0) can be estimated to be proportional to

$$I_{\text{LIF}} \sim A / (A + n_{\text{O}_2} k_Q^{\text{O}_2} + n_{\text{N}_2} k_Q^{\text{N}_2} + n_{\text{H}_2\text{O}} k_Q^{\text{H}_2\text{O}})$$

as depicted in figure 2(b). In figures 2(c)–(e), the single shot LIF signal at flow rates of 0.5, 1 and 1.5 slm are displayed. The signal is a pure LIF signal, as spontaneous emission from excited hydroxyl  $\text{OH}(A, X)(1,0)$  is negligible. The analytically obtained triangular emission pattern (figure 2(b)) agrees very well with the experimentally observed OH emission at a gas flux of 0.5 slm (figure 2(c)). A sharp edge in the emission pattern occurs at an ambient air density of approximately 0.01–1%. This shows that due to the strong hydroxyl fluorescence quenching by air, the contour of the argon–air boundary can be determined and reflects the relative space-resolved argon density. The plasma does not change the flow regime at this flow rate.

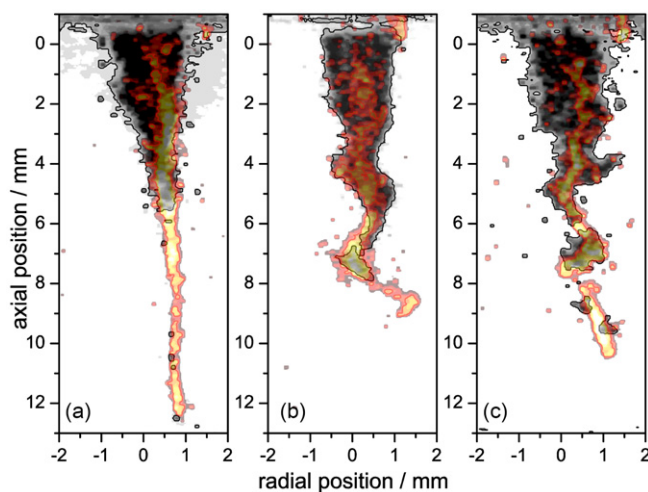
For a flow rate of 1 slm, the occurrence of Kelvin–Helmholtz (KH) instabilities is observed, leading to a breaking of the laminar jet downstream. KH instabilities are likely to appear at the mixing layer of two fluids with different densities and/or in a shear flow [24]. Both mechanisms may be triggered when heating of the gas by the plasma occurs: its density is decreased, while its expansion leads to an increased shear.

Electro hydrodynamic forces can also trigger flow regime transitions. In [25] it is observed that the flow regime of a helium jet is significantly affected by the plasma while the temperature remains at around room temperature. It is noted that helium jets may be affected more strongly by equal electro hydrodynamic forces, as their momentum is ten times less compared to argon jets at identical flow conditions. It was recently observed that the air mixing in an argon plasma jet similar to the one used in the present study is significantly increased when the plasma is turned on, which agrees with our findings [26]. Their gas temperature is around 480 K. They also attribute this effect to the occurrence of KH instabilities and/or electro hydrodynamic forces.

At 3 slm, the flux seems to remain fully turbulent as in the plasma-off case.

## 5. Turbulent flow and streamer propagation

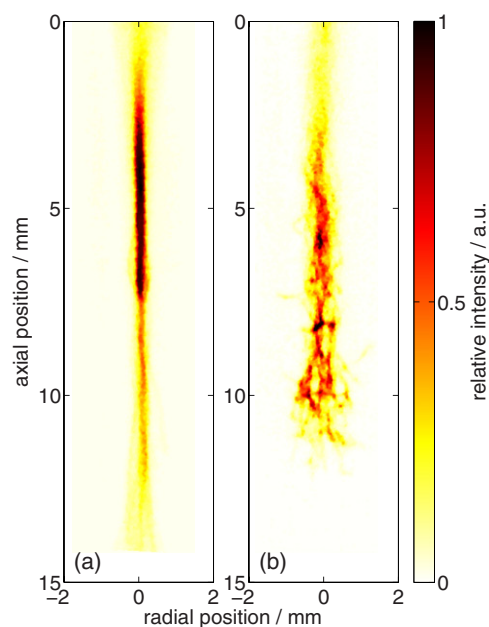
The observation of the turbulent behavior of the effluent flow is possible only if the acquisition time is much faster than the turbulence time scale. At 200 ns exposure time, the flow dynamics is static, but the discharge propagation is fast enough to grow and develop itself. Therefore looking at the effluent turbulence pattern as well as the streamer path within the same single time shot leads to fundamental information about the discharge propagation. Figure 3 shows a superimposed image of the streamer and the hydroxyl fluorescence. The hydroxyl fluorescence resembles the argon flow pattern. Both are following the same path and—especially at greater distances to the nozzle, where the argon abundance has the same dimension as the streamer thickness—the patterns are identical. On figure 3(b) and (c), which corresponds to 1 slm and 3 slm flow rates respectively, the flow patterns are not laminar and result in density gradients of argon which form a non-rectilinear argon channel. Also the streamer propagation does not follow a straight path. For the 3 slm case, it is clear that the turbulent flow has an impact on the streamer morphology (see figure 1(c)). This argon channel is about



**Figure 3.** Overlay of the spatial OH fluorescence signal and of the spatial plasma emission (visible and near-infrared) for different argon gas flow rates of (a) 0.5 slm, (b) 1.0 slm and (c) 3.0 slm. Both signals were obtained simultaneously on the same detector.

2 mm in size at the nozzle and thins down as it propagates along the jet-axis. The experimental results show that the streamer propagates into the argon channel from the wider side and ends its propagation within the very thin argon channel surrounded by air impurities from diffusion, at the space where the argon density remains the highest. This means that the streamer is always bound by argon abundance, as shown in figure 3. In the case of 0.5 slm (figure 3(a)), the argon flow is laminar, inducing a rectilinear argon channel where the streamer propagates linearly. The argon flow is only clearly visible along the first 5 mm because of the induced fluorescence, which is quenched by the diffusion of ambient species as described previously. Moreover, we observe that the streamer position is well aligned on the axis of a right circular cone resulting from the gradient of the air diffusion into the argon channel. It is especially true towards greater distances from the nozzle. These observations are in perfect agreement with the fact that the streamer propagation follows the highest argon density which obviously is located on the axis of the channel. However, those measurements have been made over a 200 ns gate which means that the flow dynamics remains static with respect to the streamer head propagation. In order to confirm the results observed from single shot acquisitions, successive acquisitions of streamer propagations are recorded with the same gating time with an interval of 100 ms between two shots. At this acquisition frequency, the flow dynamics has time to develop from one shot to the other. Thus, it is ensured that between two consecutive acquisitions, the flow pattern must differ from one to the other in the case of turbulence or non-stationary flow.

Accumulation of 50 streamers is recorded for 0.5 slm and 3 slm flow rates, which correspond to a laminar and a turbulent flow, respectively. The results are presented in figure 4. In the turbulent case at 3 slm, the streamers' spatial distribution is spread over a wider area compared to the laminar case at 0.5 slm. This observation is even more pronounced after a 6 mm distance from the nozzle, where the structure



**Figure 4.** Spatial streamer distribution in the jet effluent for an argon gas flow rate of (a) 0.5 slm and (b) 3.0 slm. The number of accumulations was 50. (The slight vertical tilt is due to the alignment of the plasma jet.)

of the turbulence is developing, as shown in figure 2(c). For the laminar case, (figure 4(a)), the position of the streamers remains constant. This is in agreement with the laminar flow pattern measured for the same conditions and presented in figure 2(a). It thus becomes evident that the atmospheric streamers are bound by argon and propagate through this argon channel. A previous study on streamer propagation in argon flow surrounded by air has been published [27]. The authors report that the reason for the propagation of a guided streamer in argon in the ambient air is due to the interface between these two gases. They observe the brightest emission on the edge on the plasma effluent and identify it as an emission from the second positive system of nitrogen,  $N_2(C)$ , mainly resulting from the efficient energy transfer of the near-resonant transition between the argon metastables of the ( $^3P_{0,2}$ ) states and  $N_2(X)$  to produce  $N_2(C)$  [28]. In this work,  $N_2(C)$  emission position differs from the referred study and is located in the center of the effluent. Moreover, the highest emission intensity was detected in the range from 4 mm up to 7 mm from the nozzle. This is in excellent agreement with figure 2(c), where one sees that OH quenching by ambient molecules reaches the on-axis. Therefore it can be assumed that metastables of argon are, in the present case, located within the argon channel where the streamer develops. One reliable reason for the difference observed to [27] is attributed to the presence of a grounded ring electrode in the plasma source which must induce a different electric field and thus different ionization mechanisms. Due to the high frequency in the present case, streamer events will benefit from previous ionization and excitation phenomena. The memory effect of metastables and ions contributes largely to the development of the discharge within the argon dominating channel. This is also valid for the turbulent cases as the vortices remain

constant on the time scale of streamers. A simulation of discharge development in the case of a pure helium laminar flow surrounded by air *without diffusion* explains that so-called streamer bullet propagation is more favourable within the helium tube than in air because of the ionization coefficient differences of each gas [29].

The similarity in streamer path and gas flow path was also found by [30] in pulsed atmospheric pressure plasma streams operated in helium and neon. Interestingly, they found clear evidence that under their experimental conditions, the plasma determines the gas channel behavior and not, as commonly thought, vice versa [25, 30, 31]. Since the mass density of argon is ten times higher than for helium, this plasma-induced gas channel forming is not directly transferable to the argon plasma jet investigated in this work. Hence, for the laminar-to-turbulent transition in the 1.0 slm case when the plasma is switched on, it is not clear whether the streamers are responsible for inducing the turbulence of the argon flow or whether the argon flow becomes turbulent itself by pure fluid dynamics. Experiments with the focus on this effect need to be performed in future. The studies presented in the present work clearly show that the path of highest argon density, lowest air impurity density and streamer propagation are identical.

## 6. Conclusion

In this study, it was shown by means of PLIF on an atmospheric pressure argon plasma jet that in turbulent mode, the streamer path changes according to the flow pattern changes and the resulting changes in air abundance. Furthermore, with known OH quenching coefficients from a comparison of analytical diffusion calculation and LIF measurements, the air impurity boundary can be determined. Comparing the plasma-on and plasma-off case, it was shown that in the laminar case, the development of KH instabilities is promoted, while in the turbulent flow, the flow patterns remain comparable. Most importantly, it was shown that the streamer pathway is defined by the ambient air diffusing into the effluent. From a convection diffusion model, a steep increase of ambient air particle density in radial direction was revealed. This gradient forms a boundary which leads to the observed streamer enclosure within the argon channel.

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