Control of the intensity of the induced flow in a liquid by a dual-frequency APPJ

R. Fiorotto[1], E. Shakerinasab[1], A. Andreetto[1], A. Patelli[1]

[1] *University of Padova, Dept. Physics and Astronomy, Padova, Italy*

*e-mail: riccardo.fiorotto.1@phd.unipd.it

Abstract

Dual-frequency HF+RF atmospheric-pressure plasmas can exhibit hybrid operating regimes that couple bulk Ω-mode ionisation with a surface-driven γ-mode via adjustment of the sheath voltage. Although such regimes have been examined in parallel-plate configurations and plasma jets, it has remained uncertain whether they can persist on liquid substrates. Here, we demonstrate that they can, and we show that the onset of the γ-mode is accompanied by a switch in the interfacial coupling mechanism, resulting in a pronounced incorporation of sodium in the discharge. When the γ-mode is triggered, the power density delivered to the plasma region adjacent to the liquid increases, producing a charge deposition that is roughly $\sim 10\times$ higher than in the RF-only operation. The liquid evaporation rate, as well as the flow field observed via particle tracking, is primarily governed by the RF power, consistent with thermal heating effects. However, at low RF power, a vertical flow induced also by the HF component is visible.

Introduction

Atmospheric-pressure plasmas (APPs) are versatile platforms for applications in plasma medicine, water treatment, and surface functionalization [1]. Dual-frequency excitation is of particular interest because it enables quasi-independent control of the parameters governing plasma–substrate interactions [2]. When radio-frequency (RF, 13 or 27 MHz) and high-frequency (HF,$10-20$ kHz) voltages are applied simultaneously, the discharge exhibits a transition between two distinct ionisation regimes as a function of the HF amplitude, which regulates the sheath thickness. Magnan et al. [3] identified a critical voltage associated with the $\Omega \rightarrow \gamma$ transition in plane-to-plane dielectric barrier discharge (DBD). This framework was subsequently generalised to coaxial DBD atmospheric-pressure plasma jets (APPJs) in the open air by Patelli et al. [2], who showed that also on the exposed dielectric substrate the RF component sustains the Ω-mode in the bulk, while the HF component controls the electron drift and initiates the γ-mode once a critical sheath voltage drop is exceeded.

In the case of a plasma–liquid interface, the sheath controls both the penetration of the electric-field into the liquid and the interfacial charge transfer [4], whereas the physico-chemical properties of the substrate determine whether electrohydrodynamic (EHD) or Marangoni stresses dominate the dynamics of interfacial flow [5].

In the present study, we address the application of a dual frequency jet on the liquid, focusing on evaluating the ignition of the γ-mode over a liquid surface by means of time resolved optical emission correlated to electrical diagnostics. Furthermore, we analyse the induced flow through particle tracking.

Materials and methods

The APPJ source (Plasma Stylus Noble, Nadir srl) adopts a coaxial configuration with two annular electrodes around an alumina tube (inner diameter 9 mm). The two upstream HF electrodes are spaced 2 mm apart, the downstream one grounded, while the RF electrode lies 13 mm further downstream and 3 mm from the outlet [2]. A quartz cuvette is placed 4 mm below the outlet, with the jet impinging vertically on the liquid surface. The argon flow is 2.0 slm; the HF supply (Nadir HV) operates at 17 kHz (max 15 kV) and the RF generator (SEREN R301) at 27.12 MHz.

Time-resolved optical emission is acquired through a horizontal 500 μm slit above the liquid surface through a photomultiplier tube (Hamamatsu H11901-20). The liquid-phase flow is characterised by PIV using a ~400 µm LED light sheet and a high-speed camera (Basler ace). The tracers are GO microflakes (~20 µm) for a demineralised water and a saline solution (NaCl + PBS).

Results and discussion

γ-modes onset

The time-resolved PMT photon counts, synchronized with both the HF and RF signals, are acquired in 200 sequences close to the liquid surface. The resulting data are accumulated into count histograms as a function of time and then analysed by FFT (Fig. 1). The signal at 54 MHz, which corresponds to the second harmonic of RF, is characteristic of the Ω-mode, while the fundamental component is associated with the γ-mode. At the beginning of the cycle, as the HF voltage increases and the surface becomes negatively charged, the Ω-mode dominates, but its intensity gradually decreases as the sheath slowly expands. Once the voltage reaches the breakdown threshold, the γ-mode ignites and the liquid surface turns positively charged. In this regime, the sheath thickness is again reduced and the Ω-mode becomes dominant once more. As the HF electrode voltage subsequently drops, the γ-mode ignition shifts to the electrode side; consequently, even though the sheath above the liquid remains thin, a temporal modulation of the emission is still observable at the liquid surface.

All these measurements are carried out using the Na doublet emission lines at 589 nm, which cannot be detected under RF-only operation. Moreover, the γ-mode becomes considerably more prominent compared to when the full emission spectrum is collected. The extraction of Na from the liquid is therefore more significantly associated with an electrical extraction process than with simple evaporation.

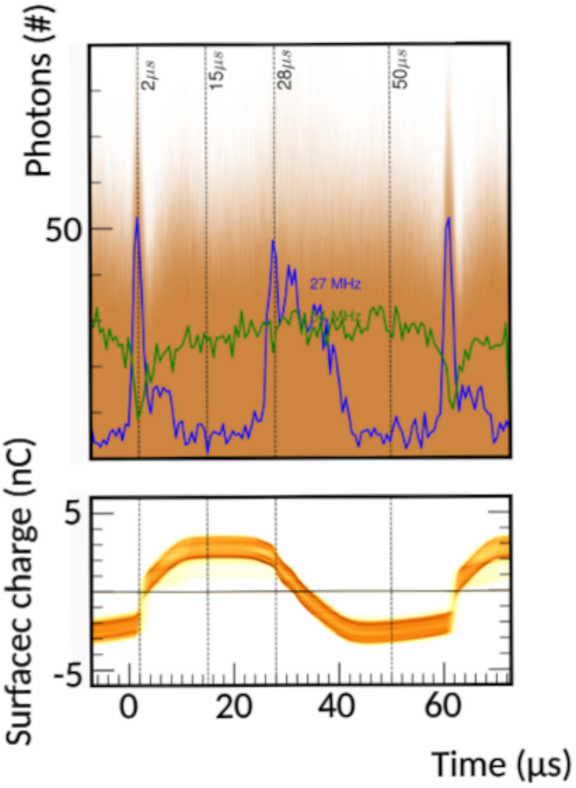

Figure 1: (top) Photon counts histogram in a HF cycle, in green the FFT magnitude linked to the Ω-mode and in blue the one of the γ-mode. (bottom) The charge collected on the liquid substrate.

The power delivered to the plasma close to the liquid surface can be estimated considering the charge collected on the liquid surface as a function of the voltage of the HF electrode. The results reveals a clear dependence of the dissipated power on the substrate conductivity. For DW the loop is narrow and the coupling predominantly capacitive, with a dissipated power of 0.93 ± 0.06 W; the power rises to 1.19 ± 0.04 W at $24.2\,\mathrm{mS\,cm^{-1}}$ and to 1.60 ± 0.05 W at $33.6\,\mathrm{mS\,cm^{-1}}$. Yet the most interesting features in terms of γ-mode as a switching knob can be observed by comparing RF-only excitation, in which the charge deposited has a maximum of ~ 0.9 nC whereas in the dual frequency case reaches the ~ 13 nC.

Induced liquid flow

PIV measurements reveal a pronounced plasma-induced bulk flow, negligible under control conditions ($<1\,\mathrm{mm\,s^{-1}}$ without gas or voltage, $\sim 2\,\mathrm{mm\,s^{-1}}$ with argon only). RF forward power is the primary control parameter: increasing it from 15 to 30 W roughly doubles the flow. The nearly identical DW and saline profiles indicate that conductivity is of minor influence in the Ω-mode, pointing to gas-phase momentum transfer and thermal convection as the principal drivers [6]. The thermal effect is clearly visible also on the evaporation rate which scales with the RF power.

The HF superimposed alters the evaporation of less than 3% and also the upstream flow of the liquid is only slightly affect. However, close to the liquid surface the difference is more significative as by reducing the RF power where the HF ignition can raise the velocity by up to ≈$6\,\mathrm{mm\,s^{-1}}$.

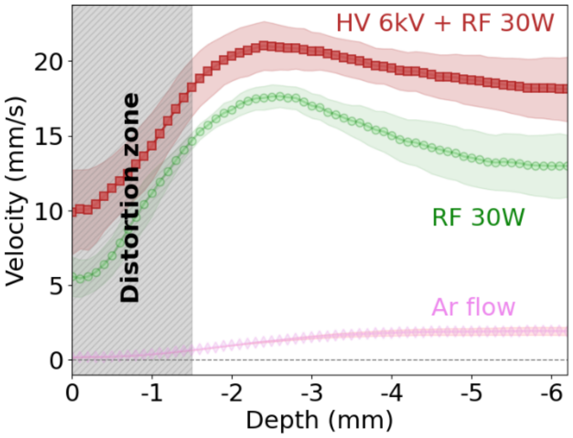

Figure 2: Electrohydrodynamic induced flows in a 33.6 mS/cm saline solution.

Conclusions

We showed that the γ-mode can also be generated on a liquid surface using a dual-frequency APPJ. Time-resolved PMT measurements combined with FFT analysis highlight a temporal alternation between the Ω- and γ-modes within a single HF cycle. The onset of the γ-mode appears when the sheath thickness and its voltage drop exceed a threshold value. The existence of a strong electric field is further supported by the detection of Na ions specifically when the γ-mode is active. Furthermore, the onset of the γ-mode allows the control of the dissipated power in the plasma close to the substrate.

PIV measurements show the critical role of the RF power. The γ-mode influences the flow only when the RF power is low or close to the surface.

References

[1] P.J. Bruggeman et al., Plasma Sources Sci. Technol. **25**, 053002 (2016).

[2] A. Patelli et al., Plasma Sources Sci. Technol. **34**, 025010 (2025).

[3] R. Magnan et al., Plasma Sources Sci. Technol. **30**, 015010 (2021).

[4] P. Vanraes and A. Bogaerts, J. Appl. Phys. **129**, 220901 (2021).

[5] A. Dickenson, J.L. Walsh and M.I. Hasan, J. Appl. Phys. **129**, 213301 (2021).

[6] J.F.M. Van Rens et al., IEEE Trans. Plasma Sci. **42**, 2622 (2014).